



# Development of prognostic aerosol–cloud interactions combining a chemistry transport model and a regional climate model

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## Abstract

To reduce uncertainties and hence, to obtain a better estimate of aerosol (direct and indirect) radiative forcing, next generation climate models aim for a tighter coupling between chemistry transport models and regional climate models and a better representation of aerosol–cloud interactions. In this study, this coupling is done by first forcing the Rossby Center regional climate model, RCA4 by ERA-Interim lateral boundaries (LBCs) and SST using the standard CDNC (cloud droplet number concentration) formulation (hereafter, referred to as the “stand-alone RCA4 version” or “CTRL” simulation). In this simulation, the CDNCs are assigned fixed numbers based on if the underlying surface is land or oceanic. The meteorology from this simulation is then used to drive the chemistry transport model, MATCH which is coupled online with the aerosol dynamics model, SALSA. CDNC fields obtained from MATCH-SALSA are then fed back into a new RCA4 simulation. In this new simulation (referred to as “MOD” simulation), all parameters remain the same as in the first run except for the CDNCs provided by MATCH-SALSA. Simulations are carried out with this model set up for the period 2005–2012 over Europe and the differences in cloud microphysical properties and radiative fluxes as a result of local CDNC changes and possible model responses are analyzed.

Our study shows substantial improvements in the cloud microphysical properties with the input of the MATCH-SALSA derived 3-D CDNCs compared to the stand-alone RCA4 version. This model set up improves the spatial, seasonal and vertical distribution of CDNCs with higher concentration observed over central Europe during summer half of the year and over Eastern Europe and Russia during the winter half of the year. Realistic cloud droplet radii (CD radii) values have been simulated with the maxima reaching 13  $\mu\text{m}$  whereas in the stand-alone version, the values reached only 5  $\mu\text{m}$ . A substantial improvement in the distribution of cloud liquid water path was observed when compared to the satellite retrievals from MODIS for the boreal summer months. The median and SD values from the “MOD” simulation are closer to observations than

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those obtained using the stand-alone RCA4 version. These changes resulted in a significant decrease in the total annual mean net fluxes at the top of the atmosphere (TOA) by  $-5 \text{ W m}^{-2}$  over the domain selected in the study. The TOA net fluxes from the “MOD” simulation show a better agreement with the retrievals from CERES instrument. The aerosol indirect effects are evaluated based on 1900 emissions. Our simulations estimated the domain averaged annual mean total radiative forcing of  $-0.64 \text{ W m}^{-2}$  with larger contribution from the first indirect aerosol effect than from the second indirect aerosol effect.

## 1 Introduction

The scientific understanding of the climate effects of the different aerosol species as well as their representation in models and their physical and chemical transformation under different meteorological conditions is still low (Boucher et al., 2013). Aerosols have a direct radiative effect by scattering and absorbing short and long wave radiation, thereby changing the reflectivity, transmissivity and absorptivity of the atmosphere. They can further act as cloud condensation nuclei (CCN), thereby influencing the microphysical properties of clouds. This, in turn, can impact the optical properties and lifetimes of clouds, thus, indirectly affecting the radiative properties of the atmosphere (Penner et al., 2004). Apart from ambient conditions, the ability of the aerosols to act as CCN depends on the size distribution (Dusek et al., 2006) and, for particles in the size range between 40 and 200 nm, on the chemical composition and mixing state (McFiggans et al., 2006).

The direct effect of aerosols and even more so, their indirect impact on radiative forcing have been identified as the largest sources of uncertainty in quantifying the radiative energy budget and its impact on climate system (Forster et al., 2007). The study of these effects accurately and in great detail requires coupling of atmospheric chemistry/aerosols to global circulation models (GCMs); however, due to the lack of resolution, their accuracy reduces when one start to zoom into regional scales. Hence,

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the recent generation of models use the regional climate models at a higher horizontal and vertical resolution instead of GCMs (for example, WRF-Chem (Grell et al., 2005), ENVIRO-HIRLAM (Baklanov et al., 2008), RegCM3-CAMx (Huszar et al., 2012; Qian and Giorgi, 1999; Qian et al., 2001 etc.). Recently, Baklanov et al. (2014) summarized the status of the online/offline European coupled meteorology and chemistry transport models with varying degrees of complexity in the representation of dynamical and physical processes, aerosol–cloud–climate interactions, radiation schemes etc. The main conclusion was that an online integrated modeling approach is the future and can be adapted to several modeling communities such as climate modeling and air quality related studies (Baklanov et al., 2014 and the references therein). The study also showed that for climate modeling, the inclusion of feedback processes is the most important and significant improvements were noticeable in climate-chemistry/aerosols interactions. Whether the coupling need to be online or offline depends on the specific study. For example, Folberth et al. (2011) showed that in long-lived greenhouse gas forcing experiments, the online approach did not give significant improvements, whereas, for short-lived climate forcers, aerosols in particular, online approach is very beneficial.

Here, we attempt a similar approach by adapting the Rossby Center regional climate model, RCA4 for the offline ingestion of CDNCs from the cloud activation module embedded in the chemistry transport model, MATCH that is online coupled to the aerosol dynamics model, SALSA. Such a coupling is unique in many ways:

1. A more detailed description of the emissions, transport, particle growth, deposition, aerosol processes can be included so as to obtain an accurate evaluation of aerosol radiative effects on a higher spatial resolution compared to global models (Colarco et al., 2010),
2. it is possible to assess the level of detail that is required to describe the effects on a regional scale and
3. it can be used to assess the effects of future climate change on air quality.

In this paper, we present the results from a full fledged working version of the coupling between a chemistry transport model (CTM) with a detailed aerosol dynamics model and a regional climate model. The coupling between these two model systems are offline and is done through CDNCs calculated by the CTM. The drawback of offline coupling is that there is no feedback on the simulation of chemistry and aerosols from changes in meteorology due to altered CDNC/radiation and no coupling to SST. In the following subsections, we introduce the models used in this study, their coupling and the improvements made in the cloud microphysical properties and radiative forcing through this coupling.

## 2 Models used and experimental setups

### 2.1 Models used

The schematic of the model coupling is shown in Fig. 1. In this study, we use the Multiple-scale Atmospheric Transport and Chemistry (MATCH) model (Robertson et al., 1999; Andersson et al., 2007) which is a Eulerian CTM that accounts for transport, chemical transformation and deposition of chemical tracers in the atmosphere based on EMEP emissions (<http://www.ceip.at>). The MATCH model is online coupled to the aerosol dynamics model, SALSA (Kokkola et al., 2008) that takes into account physical processes such as nucleation of particles, growth of particles by condensation and coagulation and computes the size distribution, number concentration and chemical composition of the aerosol species. A sectional representation of the aerosol size distribution is considered and has three main size regimes ((a) 3–50 nm, (b) 50–700 nm and (c) > 700 nm) and each regime is again subdivided into smaller bins and into soluble and insoluble bins adding up to a total of 20 bins. A schematic of the sectional size distribution and the aerosol species considered in each bin is shown in Fig. 2. Anthropogenic precursor emissions such as primary particulate matter (PM), NO<sub>x</sub>, NMVOC, SO<sub>x</sub>, NH<sub>3</sub> and CO, volcanic and DMS emissions are taken from the



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EMEP expert emissions inventory. Primary PM is divided into EC, OC and other emissions. Isoprene emissions are modeled online depending on the meteorology based on the methodology by Simpson et al. (1995). The terpene emissions ( $\alpha$ -pinene) is taken from the modeled fields by the EMEP model. Sea salt is parameterized following the scheme of Foltescu et al. (2005), but, modified for varying particle sizes. This means that Mårtensson et al. (2003) is used if the particle diameter is  $\leq 1 \mu\text{m}$  and Monahan et al. (1986) is used otherwise.

The coupling of MATCH with SALSA and the evaluation of this model set up is described in detail in Andersson et al. (2014). A cloud activation model that computes 3-D CDNCs based on the prognostic parameterization scheme of Abdul-Razzak and Ghan (2002) specifically designed for sectional representation of bins is embedded in the MATCH-SALSA model. In addition to the updraft velocity and supersaturation of the air parcel, this scheme simulates the efficiency of the an aerosol particle to be converted to a cloud droplet depending on the number concentration and chemical composition of the particles. The updraft velocity is computed as the sum of the grid mean vertical velocity and turbulent kinetic energy (TKE) for stratiform clouds (Lohmann et al., 1999) derived from the RCA4 simulation. These CDNCs are then offline coupled to a regional climate model, RCA4 (Samuelsson et al., 2011) that provide us information on the cloud microphysical properties such as cloud droplet radii, cloud liquid water path as well as radiative fluxes. In the stand-alone version of RCA4, the total number of cloud particles were assigned fixed numbers over the whole domain based on whether the surface is oceanic ( $150 \text{ cm}^{-3}$ ) or land ( $400 \text{ cm}^{-3}$ ) and scaled vertically. These are further used in calculation of effective radius of cloud droplets and in the autoconversion process (conversion of cloud droplet to rain). These are now replaced by the 3-D CDNC fields obtained from the cloud activation model.

## 2.2 Experimental setup – 1

For the simulations, RCA4 is run with 6 hourly ERA-Interim meteorology on lateral boundaries and sea surface temperature and the 3 hourly RCA4 meteorological fields

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along with fields necessary to compute the updraft velocity are used to drive MATCH-SALSA-cloud activation model. The aerosol properties are used in the cloud activation model to derive the 6 hourly CDNCs which are then employed to re-run RCA4 with dynamic rather than prescribed CDNCs to obtain cloud microphysical properties and radiative effects. Simulations were carried out with this model set up at 44 km × 44 km spatial resolution for the European domain and 24 levels in the vertical (up to 200 hPa) for an 8 year period (2005–2012). Here, we look into the improvements in the cloud microphysical properties and radiative fluxes with the incorporation of dynamic CDNCs where in only local land surface fluxes can respond to these changes, hereby referred to as the MOD simulation. These results are compared with the control simulation, hereby referred to as the CTRL simulation in which the stand-alone version of RCA4 is used.

## 2.3 Experimental setup – 2

To evaluate the indirect aerosol effects due to the present day anthropogenic aerosols, the pre-industrial (PI) emissions required for this simulation were taken from those developed for the ECLAIRE project (Effects of climate change on air pollution impacts and response strategies) for the year 1900 (<http://www.eclaire-fp7.eu>). The PI anthropogenic precursor emissions were provided for CO, NH<sub>3</sub>, NO<sub>x</sub>, SO<sub>x</sub> and VOC. The physiography files and the other emissions (biogenic emissions, DMS and volcanic) are kept the same as in the original model setup. The particulate organic matter emissions are reduced to 14 % of the current emission levels in the pre-industrial set up based on the study by Carslaw et al. (2013). Simulations were carried out at the same spatial resolution as in the previous setup and for the same European domain and for the same meteorology from 2005–2012. Here, we address the changes in cloud properties with respect to the emissions from the PI period without the climate feedbacks and we analyze the total radiative forcing and the individual contribution from the first and second indirect aerosol effects.

## 3 Model evaluation and results

### 3.1 Aerosol number concentrations

Figure 3 shows the spatial distribution of the seasonal mean accumulation mode aerosol number concentrations from MATCH-SALSA model simulations driven by RCA4 meteorology. The main contribution to accumulation mode particles are from  $\text{SO}_4^{2-}$ , EC, OC, sea salt and mineral dust. In the figure, emphasis is given to accumulation mode particles as they can act as CCN and, depending on the water availability and updraft velocity be efficiently converted into cloud droplets. A clear seasonality is noticeable with highest concentrations during the summer months and lowest during the winter months. Concentrations reach as high as  $600 \text{ cm}^{-3}$  over southern European subcontinent during summer. This may be partly due to relatively large emissions of primary fine particles and gaseous  $\text{SO}_x$ , and partly due to less precipitation in southern Europe compared to the rest of Europe resulting in longer residence times of these particles in the atmosphere (Andersson et al., 2014).

### 3.2 Cloud droplet number concentrations

The seasonal mean in-cloud averaged CDNCs for the European domain is presented in Fig. 4. During the summer half of the year, CDNCs are extremely high, reaching as high as  $225 \text{ cm}^{-3}$  over central Europe mainly because of summer time vertical mixing, high probability of liquid clouds and is also, consistent with high aerosol number concentrations simulated during this time. The land–sea contrast is more prominent during the summer months compared to the other seasons. A closer comparison with Fig. 3 reveals that regions of high CDNCs correspond mostly with regions of high accumulation mode aerosol particles. However, in some regions, this correlation is not very noticeable, especially over the Mediterranean in summer. This may be due to subsidence and lack of cloud in these regions. Higher CDNCs are observed in the Eastern

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Europe and Russia can be mainly attributed to the increased residential biomass burning during the autumn and winter months (Yttri et al., 2014).

Storelvmo et al. (2009) showed that differences in the cloud droplet activation would account for about 65 % of the total spread in shortwave forcing. So, it is important to see if this model setup reproduces the spatial distribution of CDNCs realistically. The high droplet concentrations simulated over land compared to oceans agrees well with the previous studies of Barahona et al. (2011) and Moore et al. (2013). Zeng et al. (2014) analyzed the CDNCs from MODIS and CALIOP sensors and it is evident that the droplet concentrations over European subcontinent would be on an average between  $150\text{--}200\text{ cm}^{-3}$  and over oceans, the concentrations are as low as  $100\text{ cm}^{-3}$ . This agrees well with our simulations.

To understand the vertical distribution of CDNCs, we select the following 4 regions as in Fig. 5 – R1: Mediterranean R2: Eastern Europe R3: Central Europe and R4: northern N. Atlantic. Figure 6 shows the joint histograms of CDNC and height (in km) over the four selected regions for winter (DJF) and summer (JJA) months respectively. It can be seen that the majority of the CDNC values are less than  $500\text{ cm}^{-3}$ , but occasionally values also reach as high as  $800\text{ cm}^{-3}$  irrespective of season and region. The figures also show an interesting seasonality of the PDFs (probability distribution functions) of CDNC. For example over the Mediterranean (R1), two peaks can be observed in summer, one around 3 km and another in the boundary layer, however, in winter, only one peak is present and is located below 2 km. This is mainly due to the stronger vertical mixing of aerosols together with increased buoyancy and convection in summer. It is well known that the frequency of occurrence of very low level water clouds is high over this region during summer, while, in winter, the baroclinic disturbances lead to northward transport of winter storms over this region. Over Eastern Europe (R2) and central Europe (R3), the height at which maxima of CDNCs occur at around 1.5 km in summer, whereas during winter, the maxima is in the boundary layer. The droplet concentrations can vary widely over Eastern Europe compared to over central Europe where the concentrations are mostly towards the higher side irrespective of the

seasons. The opposite is observed over northern N. Atlantic (R4) with maxima CDNC simulated at around 1.5 km in winter and in the boundary layer in summer. This can be attributed to the long range of transport of pollutants from across the Atlantic observed during the winter months (HTAP, 2010).

### 3.3 Cloud droplet radii

As mentioned in Sect. 2, both the CTRL and MOD simulations follows the same parameterization scheme of Wyser et al. (1999) in the radiation and is formulated as in Eq. (1). The effective radius for spherical droplets is estimated as,

$$r_{e,water} = \frac{3CC}{4\pi\rho_l kN} \quad (1)$$

where CC is the cloud condensate content in  $\text{kg m}^{-3}$ ,  $\rho_l$  is the density of liquid water,  $N$  is the number concentration of cloud droplets in  $\text{m}^{-3}$ ,  $k$  is a constant depending on marine or continental clouds. In the CTRL simulation,  $N$  was assigned 150 and  $400 \text{ cm}^{-3}$  depending on the underlying surface, however, in the MOD simulation dynamic CDNCs are used.

The winter and summer mean cloud droplet radii in liquid water clouds for the MOD simulation (top row) and the CTRL simulation (2nd row) is discussed in Fig. 7. At a first glance, apart from the spatial differences, one notice the strong disparity in the range of the radii values. In the MOD simulation, CD radii reach as high as  $13 \mu\text{m}$ , whereas in the CTRL simulation, the maxima observed is  $5 \mu\text{m}$ . It can be seen that the radii of the droplets are much lower in the summer months compared to the winter months basically due to the increased pollution during summer resulting in smaller droplets.

In Fig. 8, the joint histograms of CD radii and height during the summer over Mediterranean and Eastern Europe in MOD simulation are shown. The corresponding pattern in the CTRL simulation is shown as the solid line. It can be seen that the CTRL simulation does not exhibit any variability. However, the MOD simulation shows a distinct



variability in height and range. It can be seen that over the Mediterranean and over Eastern Europe, a wide range of droplet radii can be observed from as low as 5  $\mu\text{m}$  up to 16  $\mu\text{m}$  and the larger droplets are present at around 2.0–4.0 km s. However, there is a higher probability of observing larger droplets over Eastern Europe compared to the Mediterranean.

The critical droplet radius at which large scale precipitation occurs is set to 10  $\mu\text{m}$  in the microphysics scheme. This would mean that with these low CD radii values obtained in the stand-alone RCA4 model, precipitation would be absent. It is important to note that the plotted CD radii values are derived from the model radiation scheme (ie. this is the radiatively active CD radii). For more details, refer Appendix A.

### 3.4 Cloud liquid water path

Figure 9 refers to the percentage change in column integrated cloud water (CLWP) in the MOD simulations compared to CTRL simulations averaged over the winter (DJF) and summer (JJA) months. Positive values mean that the CLWP increased in the MOD simulations compared to the CTRL and vice versa. It can be seen that during the winter months, there is a significant decrease (up to 25 %) in the vertically integrated cloud liquid water over land and a slight increase over the water bodies when the 3-D CD-NCs are used. However, during the summer months, the pattern is reversed with a noticeable increase in CLWP over most of the European subcontinent. The decrease in CLWP with increase in CD radii is consistent and may be partly attributed to precipitation removal. In the model, the critical droplet radius at which autoconversion becomes efficient is set to 10  $\mu\text{m}$ . When the CD effective radius exceeds this critical droplet radius, precipitation occurs. However, over land during summer, an increase in CLWP is observed and can be partly due to the fact that the increase in CD radius is not sufficient to trigger precipitation.

We evaluated model simulated cloud liquid water path estimates using satellite sensor retrievals. We selected LWP for evaluation as it is tightly connected not only to other microphysical properties of clouds, but also to the first and second indirect aerosol ef-

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fects which are the main application focus of the coupling attempted here. We used a decade long data record (2003–2012) from the Moderate Resolution Imaging Spectroradiometer (MODIS) sensor flying onboard NASA's Aqua satellite since 2002 (Platnick et al., 2003; Hubanks et al., 2008). The monthly Level 3 data from the Collection 5 are analysed over the study area to compute probability density function of LWP for the boreal summer months of JJA (June, July and August), when liquid clouds are most prevalent and the quality of satellite retrievals is also best. The comparison is shown in Fig. 10. We observe substantial improvement in the distribution of LWP in the MOD simulation compared to the CTRL-Simulation. The inclusion of the chemistry-aerosol–cloud microphysical link leads to more realistic distribution of LWP that is closer to the observations. At the lower end of the distribution, the model still slightly overestimates the frequency of those optically thin liquid water clouds. But the LWP of optically thick clouds, which are most abundant and play a key role in the radiation budget over the study area, shows substantial improvements in the MOD simulation.

### 3.5 Total radiative fluxes at the TOA

The difference in the annual mean total net fluxes at the TOA due to these changes in the cloud microphysical properties is shown in Fig. 11. A significant change is seen over most of the domain with decreases up to  $-5\text{ W m}^{-2}$  when the CDNC values are assigned fixed numbers depending on the underlying surface.

To evaluate the TOA radiative fluxes, we used a decade long data for comparison from the Clouds and the Earth's Radiant Energy System (CERES) sensor that is flying onboard Aqua satellite as well (Wielicki et al., 1996) (more information is available at [http://ceres.larc.nasa.gov/documents/DQ\\_summaries/CERES\\_EBAF\\_Ed2.8\\_DQS.pdf](http://ceres.larc.nasa.gov/documents/DQ_summaries/CERES_EBAF_Ed2.8_DQS.pdf)). The Level 3 Energy Balanced and Filled estimates of all-sky net top of the atmosphere fluxes (EBAF-TOA, Edition 2.8) are analysed for comparison with MOD and CTRL simulations and shown in Fig. 12. The distribution of fluxes in the MOD simulation is closer to the CERES observations compared to CTRL simulation.

## 4 Aerosol radiative forcing at the top of the atmosphere

In this section, we evaluate the effect of changing cloud albedo and cloud lifetime due to the present day anthropogenic aerosols which is widely known as the first indirect aerosol effect (Twomey, 1959) and the second indirect aerosol effect (Albrecht, 1989) respectively. In a review paper, Lohmann and Feichter (2005) summarized the aerosol radiative effects into positive and negative perturbations to the radiation budget. Both the indirect aerosol effects tend to cool the Earth system by increasing the cloud optical depth and cloud cover, thereby resulting in the reduction of the net radiation reaching the TOA and surface.

The local radiative forcing associated with these IAE are in most cases estimated as the difference between the perturbed and unperturbed radiative fluxes. The perturbed case is the current climate scenario and in the unperturbed case, the fluxes are calculated based on a pre-industrial or pristine scenario with meteorology and SST remaining the same in the both cases. In this study, for the unperturbed case (PI), we use the pre-industrial emissions from 1900s as explained in Sect. 2.3. And the present day climate scenario as in the “MOD” simulations would be our perturbed case (PD). In the following paragraphs, we analyze the changes in CDNC and CLWP in the (PD-PI) case and the TOA aerosol radiative forcing over Europe which arises mainly due to the response of the land surface without other climate feedbacks. Figures 13–15 show the annual mean difference in aerosol number concentrations, CDNC and CLWP respectively with respect to the PI emissions expressed as a percentage.

It can be seen that there is approximately 50–80 % increase in the aerosol number concentrations with respect to PI era over southern and Eastern European subcontinent and around 10–30 % over the rest of Europe and over the oceans. The steep increase in the aerosol concentrations may be attributed to the increase in anthropogenic pollutant precursor emissions in these countries in the present climate. The spatial distribution in these are reflected in the increase of almost up to 70 % increase in CDNCs and correspondingly, an increase of up to 10 % in CLWP. Figure 16 shows

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the spatial distribution of the annual mean indirect aerosol forcing when using the 1900 reference value over the study region. The European domain averaged annual mean radiative forcing is  $-0.64 \text{ W m}^{-2}$ , with values reaching as high as  $-1.3 \text{ W m}^{-2}$ . This is comparable to the estimate of  $-0.96 \text{ W m}^{-2}$  obtained by Carslaw et al. (2013) for the global mean forcing using the same reference period.

We also estimated the individual contribution of the first and second IAE to the total aerosol radiative forcing. This is done by turning off the individual IAEs by prescribing the fixed values for CDNCs as in the stand-alone RCA4 version in the radiation and cloud microphysics calculation (Lohmann and Feichter, 2001).

In IPCC-2013 (Boucher et al., 2013) it was pointed out that the estimated values of the first IAE constitute the largest uncertainty and vary significantly between the different models. The impact of changes of aerosols on cloud albedo through the changes in droplet radius (first IAE) is estimated from our model set up to be  $-0.57 \text{ W m}^{-2}$  when averaged over the European domain. IPCC models estimated the global annual mean first IAE to be  $-0.7 \text{ W m}^{-2}$  and can vary widely from  $-1.8$  to  $-0.3 \text{ W m}^{-2}$ . However, the impact of changes of aerosols on cloud lifetime via the modification of precipitation efficiency is estimated to be  $-0.14 \text{ W m}^{-2}$  when averaged over the European domain used in this study. Studies have shown that the uncertainties in this are even larger compared to the first IAE. Depending on the autoconversion schemes used in the global models, Rotstayn and Liu (2005) showed that the global mean second IAE can vary from  $-0.71$  to  $-0.28 \text{ W m}^{-2}$ , of which the scheme used to obtain the value of  $-0.28 \text{ W m}^{-2}$  is more realistic.

## 5 Conclusions

In this study, we coupled the Rossby Center regional climate model, RCA4 for the offline ingestion of CDNCs from the cloud activation module that is embedded in the chemistry transport model, MATCH that is online coupled to the aerosol dynamics model, SALSA. Such a set up is beneficial mainly because a more sophisticated rep-



resentation of emissions, transport, particle growth, deposition, aerosol processes can be included at a higher resolution compared to global models. Simulations were carried out with this set up for the period 2005–2012 over Europe using present day emissions (PD) from EMEP for the year 2000 and also, using the stand-alone version of RCA4.

5 We carried out two sets of analysis:

1. Investigate the improvements in the cloud microphysical properties with the input of the 3-D CDNCs that replaced the assigned fixed numbers in the stand alone version of RCA4 based on if the underlying surface warm-continental or cold-oceanic.
- 10 2. Evaluate the indirect aerosol effects using this integrated modeling approach using the PI emissions taken from the ECLAIRE project. The particulate matter in the PI period are reduced to 14 % of the current levels based on Carslaw et al. (2013).

This model set up improves the spatial, seasonal and vertical distribution of CDNCs with higher concentration observed over central Europe during summer half of the year and over Eastern Europe and Russia during the winter half of the year. Realistic cloud droplet radii numbers has been simulated with the maxima reaching  $13\mu\text{m}$  whereas in the stand-alone version, the values reached only  $5\mu\text{m}$ . The stand-alone version of RCA4 over-estimated the vertically integrated cloud water by up to 25 % in winter and underestimated by similar magnitude in summer over the European subcontinent. Comparisons with satellite retrievals from MODIS reveals a significant improvement in the LWP distribution; the median and SD values obtained from the MOD simulation is much closer to observations than the CTRL simulation. A significant decrease by up to  $-5\text{Wm}^{-2}$  in the total TOA net fluxes is simulated owing to these changes. The TOA net fluxes obtained with the new model setup show a better agreement with net flux retrievals from the CERES instrument than those computed with the old model setup. This confirms the importance of employing a realistic, dynamic description of aerosol number distribution fields in regional climate models.

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Using the pre-industrial emissions from 1900s, we estimated an increase of around 50–70 % CDNCs over southern and Eastern Europe and below 30 % over the rest of Europe in the present day climate consistent with the increases in aerosol number concentrations and correspondingly, an increase in CLWP is simulated over our study area. These changes resulted in an annual mean TOA radiative forcing over Europe of  $-0.64 \text{ W m}^{-2}$  which is comparable to previous estimates for the same reference period. It was also estimated that the contribution from the first IAE is larger than the contribution from the second IAE.

## Appendix A

In a given timestep,  $t = n$ , the thermodynamic tendencies resulting from resolved dynamics and parameterized vertical turbulent fluxes are first calculated. These terms are stored in the relevant tendency terms,  $\frac{\partial \chi}{\partial t}$  at  $t = n$ . Next, the radiation scheme is called using the thermodynamic values (temperature,  $T$ ; humidity,  $q$ ; cloud water,  $cw$  etc.) valid at  $t = n$ . After the radiation fluxes have been calculated, the model calls the surface scheme, then, convection and finally, condensation and cloud microphysics is called. On entering the microphysics scheme, the thermodynamic variables valid at  $t = n$  are updated by the tendencies from dynamics and turbulence. For example,

$$T = T_n + \Delta t \times \left\{ \frac{\partial T}{\partial t} \Big|_{\text{dyn}} + \frac{\partial T}{\partial t} \Big|_{\text{turb}} \right\}$$

and these variables are used to calculate a new value of  $cw$ ,  $q$  etc. consistent with the updated saturated state of the atmosphere. With this, calculated cloud water can increase locally due to the dynamical and turbulent terms and this re-calculated CD radii for the autoconversion process in the microphysics scheme may be substantially larger than in the radiation scheme (exceeding the threshold for precipitation onset of  $10 \mu\text{m}$ ). With the microphysics scheme, precipitation removal of generated cloud water is also parameterized, hence, the fraction of the newly generated cloud water is removed as



rain. The resulting cw tendency due to condensation and subsequent precipitation removal is then incremented on to  $t = n$  values, along with all other tendencies from  $t = n$  to give new values for the next timestep. In this manner, CD radii in the microphysics scheme can increase above the threshold for onset of precipitation and then, as a result of this precipitation removal of cloud water decrease again below the threshold. The low (precipitation affected) value of cloud water is what the radiation scheme “sees” in each timestep leading to a low estimate in CD radii.

## Code availability

The different model source codes and the coupled system used in this study are not entirely available for open source distribution at present, but can be made available to interested users/investigators upon request. The aerosol microphysics code, SALSA is distributed under Apache 2.0 license, while the regional climate model RCA4 and the chemistry transport model MATCH are available upon request from the SMHI.

*Acknowledgements.* This work was supported and funded by the Swedish Environmental Protection Agency through the CLEO (CLimate change and Environmental Objectives) and partly by the SCAC (Swedish Clean Air and Climate research program) projects. We also acknowledge the funding from FORMAS through MACCII (Modeling Aerosol-Cloud-Climate Interactions and Impacts) project. M. Kahnert acknowledges funding from the Swedish Research Council (Vetenskapsrådet) under project 621–2011-3346.

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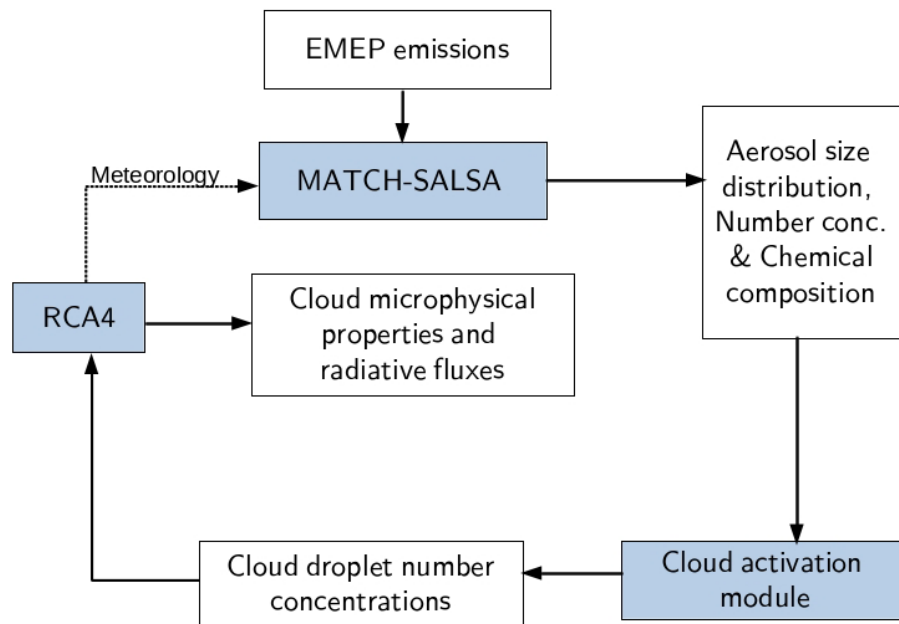
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**Figure 1.** Schematic showing the different model components and their couplings.

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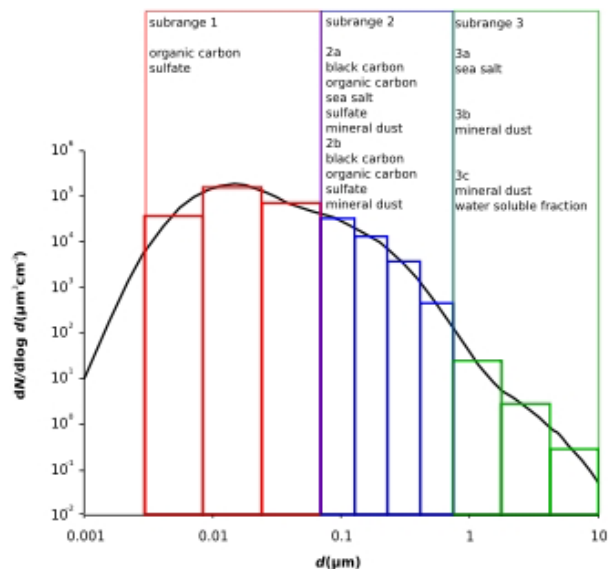
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**Figure 2.** Schematic of sectional distribution of aerosol size bins and the chemical components in the bins (taken from Kokkola et al., 2008).

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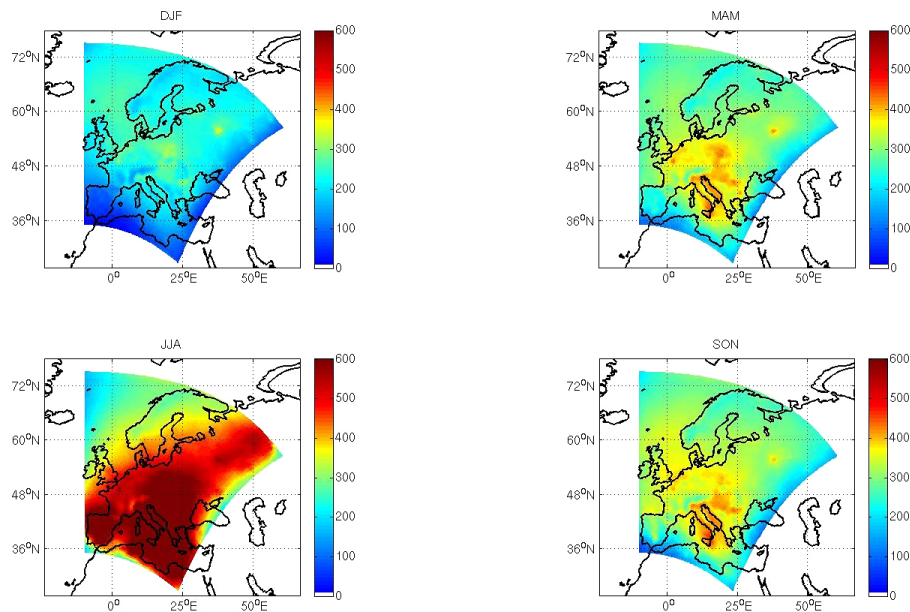
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**Figure 3.** Averaged aerosol number concentrations in the accumulation mode ( $\text{cm}^{-3}$ ).



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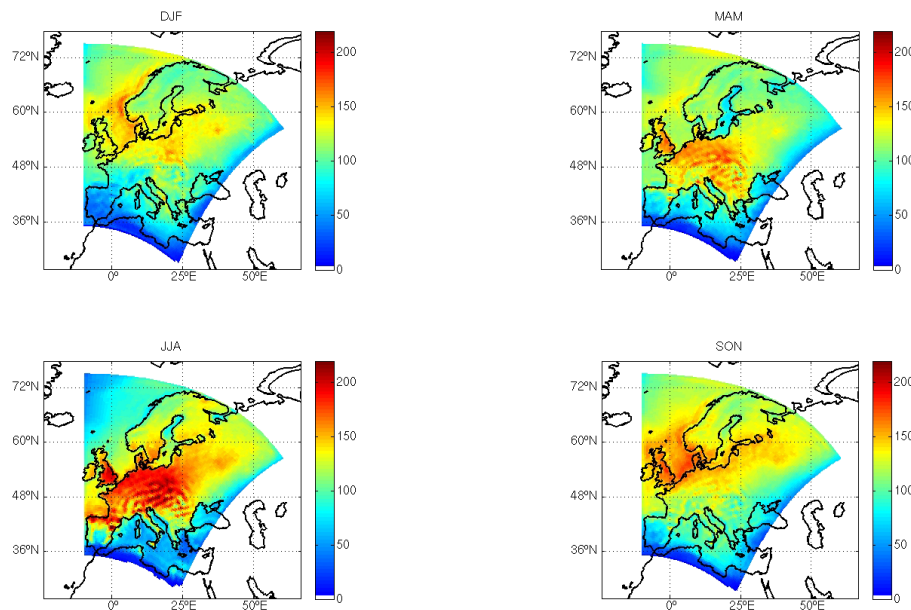
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**Figure 4.** Seasonal mean in-cloud averaged cloud droplet number concentrations ( $\text{cm}^{-3}$ ).

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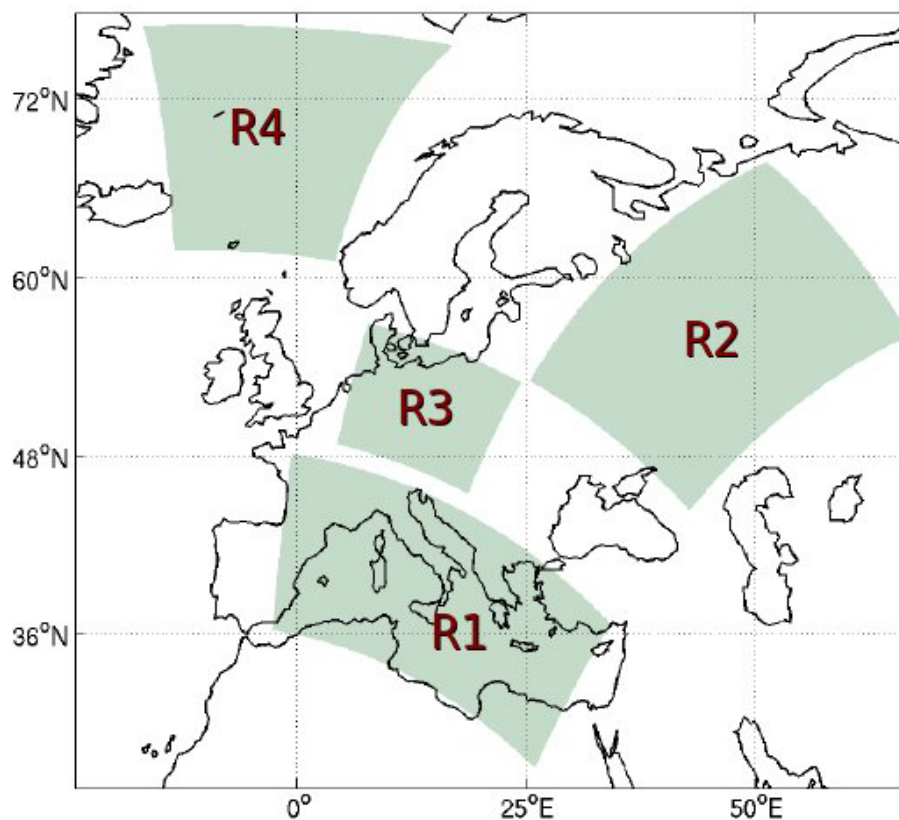
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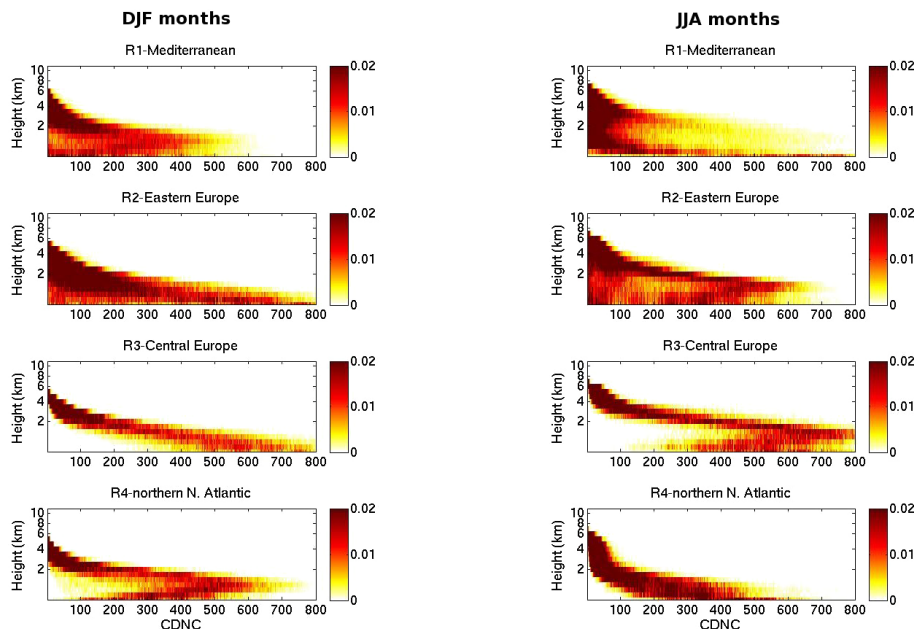
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**Figure 5.** Schematic of the four regions selected for this study. R1 – Mediterranean; R2 – Eastern Europe; R3 – Central Europe; R4 – northern N. Atlantic.

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**Figure 6.** Seasonality in the vertical distributions of CDNC ( $\text{cm}^{-3}$ ) shown as joint histograms averaged over winter averaged (DJF) months (first column) and summer averaged (JJA) months (second column) for the selected 4 regions.

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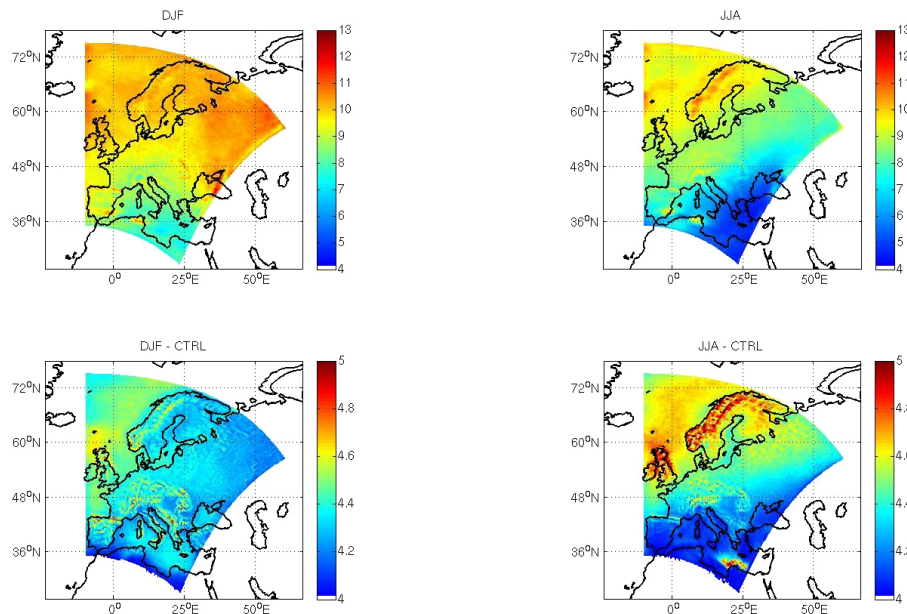
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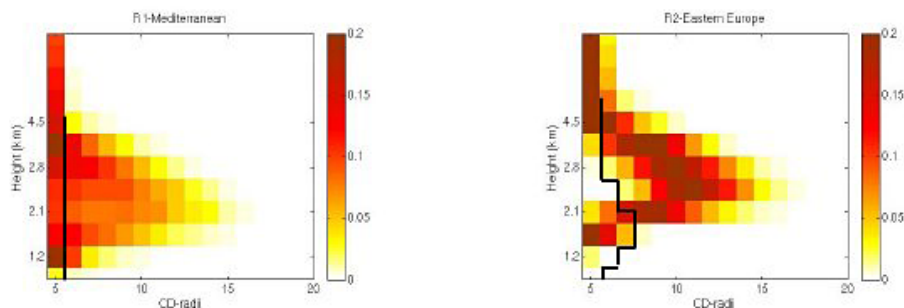
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**Figure 7.** Seasonal mean CD radii ( $\mu\text{m}$ ) averaged over the entire water cloud for DJF mean and JJA mean in the MOD simulations (top row) and CTRL simulation (2nd row). Note the difference in scale.

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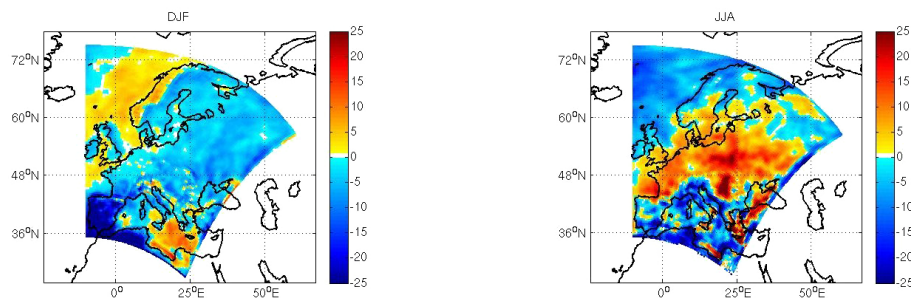


**Figure 8.** Joint histograms of CD radii ( $\mu\text{m}$ ) vs height averaged over JJA months in MOD simulation and the solid line shows the same in CTRL simulation.

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**Figure 9.** The difference in the vertically integrated cloud water in the MOD simulation compared to the CTRL simulation expressed as a percentage over the winter (DJF) and summer (JJA) months.

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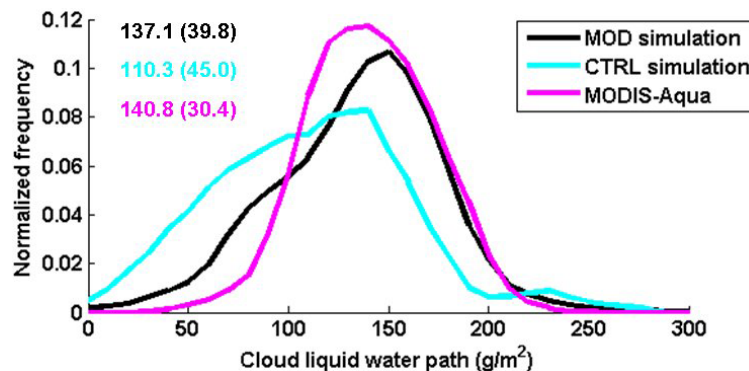
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**Figure 10.** Comparison of cloud liquid water path ( $\text{gm}^{-2}$ ) with observations from the MODIS sensor onboard the Aqua satellite. The histograms are computed over the entire study area and for the JJA months. The median and SD (in brackets) values are shown for all cases.

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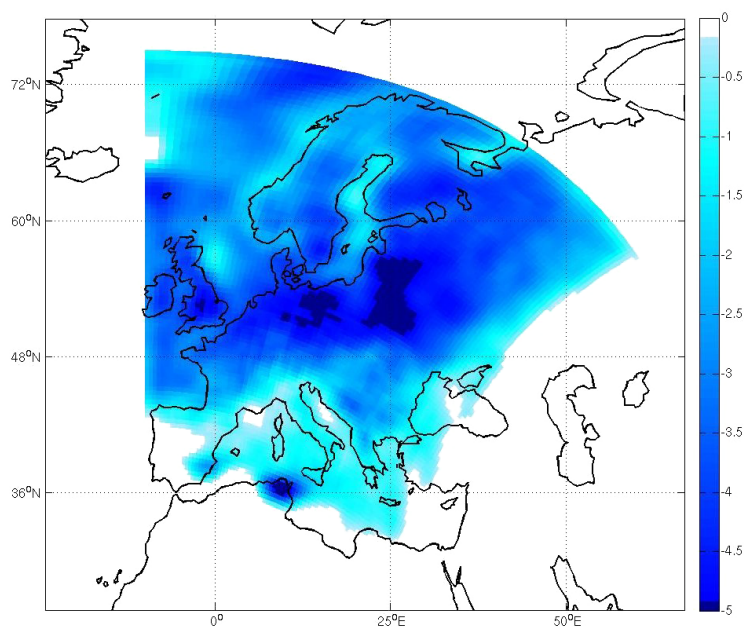
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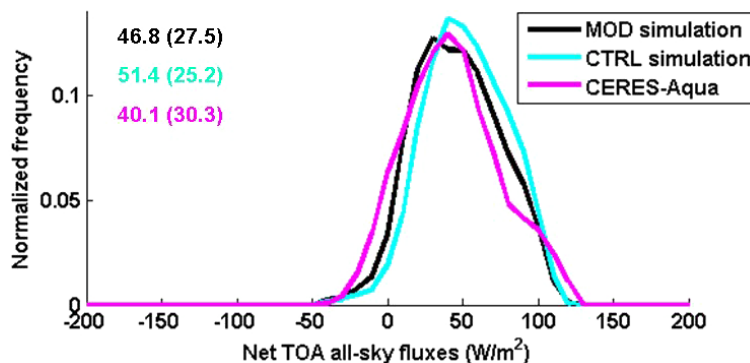


**Figure 11.** Difference in the annual mean total net fluxes ( $\text{Wm}^{-2}$ ) at the TOA in the (MOD-CTRL) case.



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**Figure 12.** Comparison of total TOA fluxes ( $\text{W m}^{-2}$ ) with observations from the CERES sensor onboard the Aqua satellite. The histograms are computed over the entire study area and for the JJA months. The median and SD (in brackets) values are shown for all cases.

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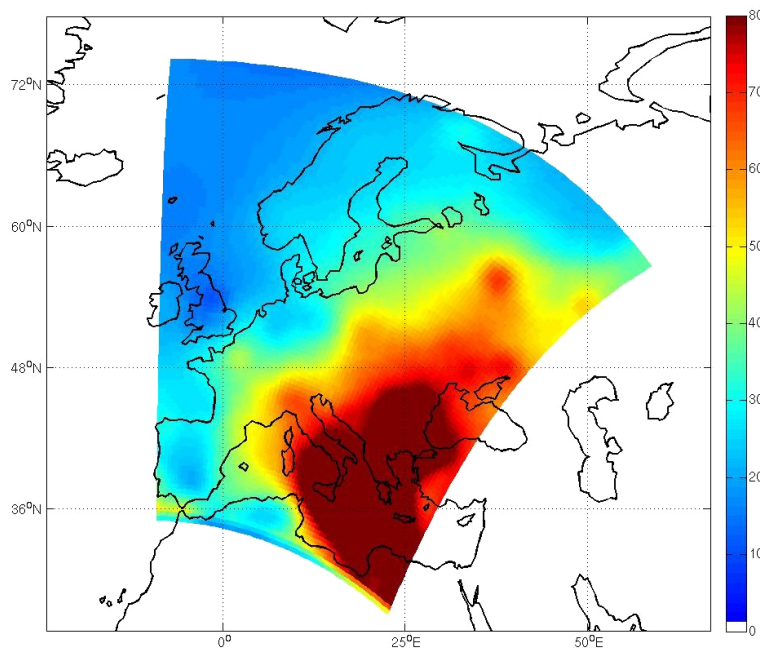
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**Figure 13.** Difference in the annual mean aerosol number concentrations for the (PD-PI) simulation expressed as a percentage.

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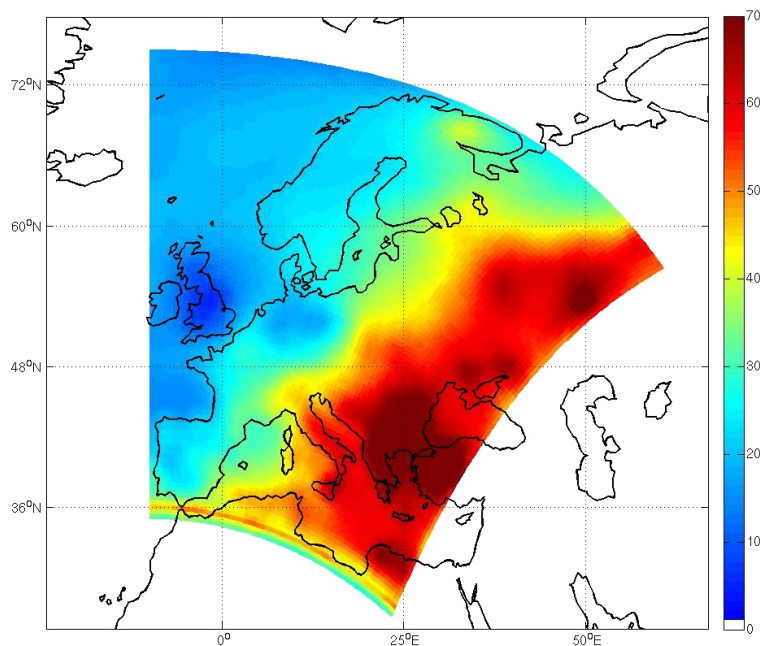
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**Figure 14.** Difference in the annual mean CDNC for the (PD-PI) simulation expressed as a percentage.

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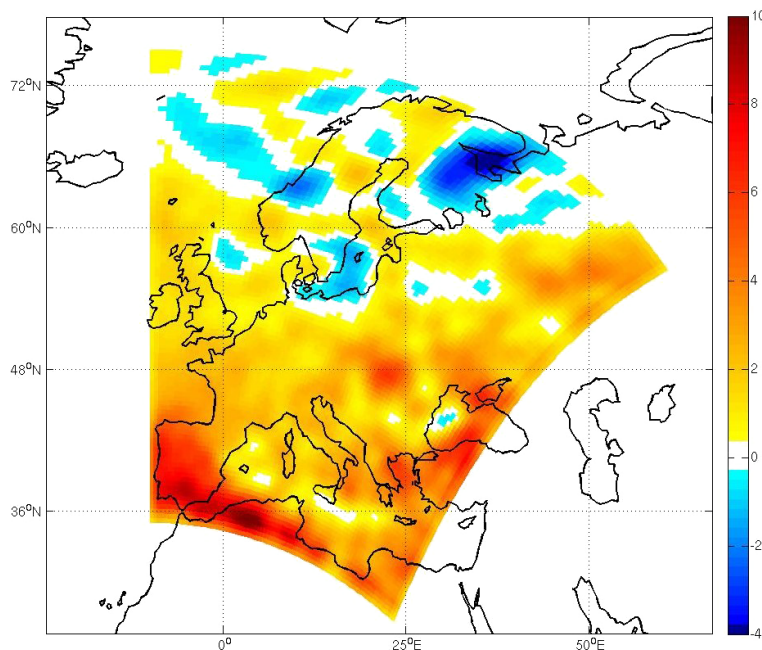
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**Figure 15.** Difference in the annual mean CLWP for the (PD-PI) simulation expressed as a percentage.

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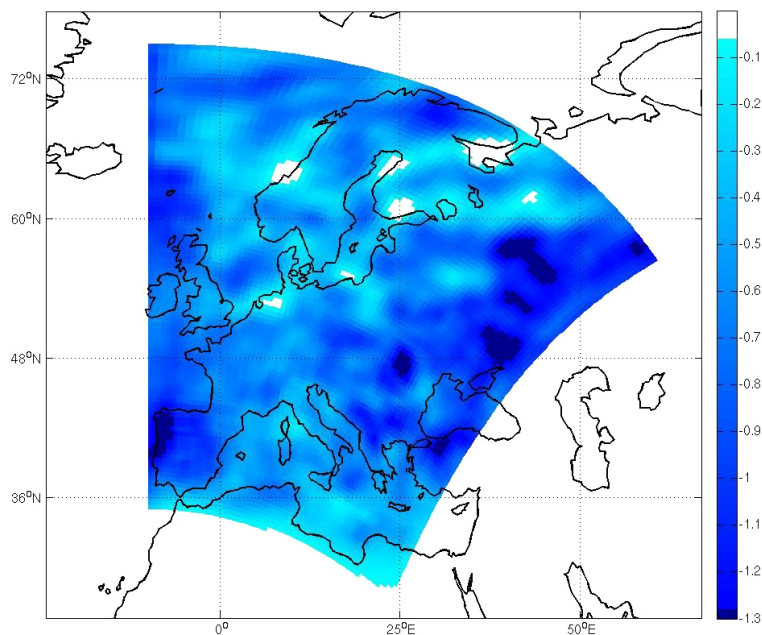
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**Figure 16.** Annual mean aerosol radiative forcing at the TOA ( $\text{Wm}^{-2}$ ).