<u>Response to Reviewer #1</u>

We thank the reviewer for her/his comments. Please find below point-by-point response to the reviewer's comments. The major changes in the manuscript are highlighted.

Main comments:
1) As the authors stated in the introduction, a number of regional climate models coupled with chemistry and aerosols already exist, e.g. RegCM3-CAMx, WRF-Chem, and others. It would be interesting to summarize the assumptions in the existing models regarding the aerosol-cloud interactions, and their performances in reproducing CDNC, if some studies are available. This can help to highlight what is new in this study, e.g. is SALSA already implemented (offline/online) in any other regional climate model?
The aerosol-cloud interactions are either implicity or explicitly included in all online models. Schemes (for ex. Abdul-Razzak and Ghan , 2002) that explicitly resolve the activation of CCN to cloud droplets are currently included only in a handful of online coupled models (ENVIRO-HIRLAM, WRF-Chem etc). Instead, the droplet number concentrations are derived empirically and is used in the parameterization of droplet radii and autoconversion processes. SALSA is not implemented in any regional model, but, only in global and large eddy models. This is added to the text.

2) In Andersson et al (2014) on MATCH-SALSA model, it is stated that "The lack of ammonium nitrate condensation in the aerosol microphysics could cause underestimation of cloud droplet number concentration (CDNC)." I think a comment on this should be added also in your model description section and the possible impact on your results.
We add the following to the manuscript " Particulate nitrogen is described outside SALSA, i.e. ammonium salts are not taken into account in the modeling of the aerosol microphysical processes. The lack of ammonium nitrate condensation in the aerosol microphysics could cause underestimation of cloud droplet number concentration (CDNC). Currently there are no parameterizations available that take into account co-condensation of ammonia and nitric acid. "

3) It is not clearly described in sections 2.3 and 4, how the first and second IAE are quantified separately. From Page 910, Lines 6-9, it seems that one or two more simulations were performed to turn off the individual IAEs by prescribing the fixed values for CDNC. This should be explained better, probably in Section 2.3. Did you run both PI and PD simulations turning off the IAEs? Did you turn off the first and second IAE at the same time or you had to perform two separate experiments?
To explain this in a better way, the following sentences are added in Section 2.2 Experimental setup-2.
" To evaluate the individual contribution from the first and second indirect aerosol effects to the total radiative forcings, two additional simulations each (for PI and PD climate) were carried out. We turn off the individual IAEs by prescribing the constant CDNC values for the calculation of one IAE at a time, for example, to evaluate the sole contribution from the first IAE, 3D CDNC fields are used in the computation of CD radius to assess the changes in cloud albedo (1st IAE) and constant CDNC values are used in the scheme for the autoconversion process (2nd IAEs) and vice versa."

4) In section 3.3 it is not very clear the last paragraph (Page 907, 6-10). It seems that in the

standard RCA4 version (fixed CDNCs), large scale precipitations are not possible, due to droplet radius below the critical level of 10 μm. At this point, a figure on the impact on precipitations due to the CTRL and MOD simulations could be added.

This is described in Appendix-A. The CD radii used in the radiation scheme are not used in the autoconversion process. The CD radii are re-calculated based on the updated tendencies from dynamics and turbulence that re-calculates cloud water, humidity etc up on entering the microphysics scheme. The Appendix-A is referred towards the end of Section 3.3

5) Regarding the comparison with MODIS and CERES measurements, the authors showed improvements in the frequency distributions (Figure 10 and 12), but I would include when possible also a comparison of the spatial distribution of the measured and simulated fields, to evaluate eventually improvements in the spatial distribution of the simulated fields.

We have now included spatial comparison of cloud liquid water path (also see reply to Reviewer #2). The improvements are best seen in the LWP fields rather than radiation budget. Moreover, LWP is central to the estimation of aerosol-cloud interactions including those relating to radiation changes.

6) Is there a plan to directly implement SALSA into the RCA4 model, to have the online coupled aerosol-cloud interactions? It should be mentioned in the conclusions if such development is foreseen.

This study shows a substantial improvement in the cloud microphysical properties and radiative fluxes with this offline coupled model set up. Hence, we recognize the need for an online coupled model system and we plan to implement SALSA into RCA4 in the future. It is mentioned in the conclusions in the revised manuscript.

Minor comments:
Title: The word "development" in the title is not appropriate, as this work uses existing models doing an offline coupling between them.

The title is now updated as "Integration of prognostic aerosol-cloud interactions in a chemistry transport model coupled offline to a regional climate model".

Page 898, Line 1: remove comma after hence
Page 898, Line 6: ".. , RCA4, . . ." or use parenthesis "(RCA4)"
Page 898, Line 9-10: I would replace with "In the stand-alone RCA4 version CDNCs are constants, distinguishing only between land and ocean surface."

The above suggestions are incorporated in the updated manuscript.

Page 898: Line 23: which months are included in summer half of the year and winter half of the year? Rephrase.

The terminologies 'summer half of the year' and 'winter half of the year' are removed from the text and replaced by DJF seasonal mean and JJA seasonal means.

Page 898, Line 25: ".. reaching 13μm, whereas in the stand-alone version the values reached only 5 μm."
Page 898, Line 28: explicit SD.
Page 899, Line 5: "are estimated in the MOD simulation in comparison to pre-industrial aerosol emissions (1900)"
Page 899, Line 7: You could add in parenthesis the estimates for the first and second IAE.
Page 899, Line 25-26: I would replace "The study of these . . . great detail requires ..", with "An accurate estimate of these effects requires the coupling . . ."
Page 899, Line 27: use "due to their coarse resolution", instead of lack of resolution
Page 900 Line 10-13: consider the following rephrasing: "Their study also showed that the inclusion of climate-chemistry/aerosol coupling led to significant improvements in climate

models."
All the above suggestions are incorporated.

Page 900, Line 21-28: The three points highlighted are not unique to your study. Other regional models exist, which have coupled aerosol-cloud interactions. The last point is also too general, in your case also you don't have online coupling between climate and chemistry, so you would have to run a further simulation with MATCH-SALSA using the meteorology from MOD simulation.
The uniqueness mentioned here is not just meant for this coupled model setup, but also to the online/offline coupled models that already exist. The term 'unique' will be removed from the text.
Page 901, Line 9: remove "through this coupling."
Page 901, Line 10: Section title, "Models description and experimental setup"
Page 901, Line 11: Section title: "Models description"
Page 901, Line 14: "an Eulerian CTM"
Page 901, Line 25: remove "precursor"
All the above suggestions are incorporated.

Page 902, Line 1: which year of EMEP emissions is used?
EMEP emissions from the year 2003 are used in this study. This is now clarified in the text.

Page 902, Line 1: Is there a reference for the PM chemical speciation, vertical distribution and in which aerosol size bins the anthropogenic emissions are injected?
This division of the primary PM is used based on the TNO-MACC emissions of EC and OC (Kuenen et al., 2011; Pouliot et al., 2012; see also the MACC project web page http://www.gmes-atmosphere.eu/). The emissions were given as annual totals. Seasonal, weekday and diurnal variations of the emissions are sector specific and based on results from the GENEMIS project (http://genemis. ier.uni-stuttgart.de/; Friedrich and Reis, 2004). The vertical distribution is also sector specific and based on the vertical distribution used by the EMEP model. The particle emissions of EC and OC are distributed over different particle sizes according to sector resolved mass size distributions described by Visschedijk et al. (2009). See Andersson et al. (2014; 2015) for more details on how the emissions are distributed.
The above text is added to the manuscript.

Page 902, Line 11: consider replacing with "..Ghan (2002), specifically designed for aerosol representation with sectional bins, is embedded . . ."
Page 902, Line 13: remove "the" in "the efficiency of the an aerosol particle"
Page 902, Line 18: "model, RCA4 (. . .), that provides us .."
Page 902, Line 21-25: "the total number of cloud particles were set to constant values over the whole domain, based on .. vertically. These constant values were further used. . . (. . . rain). In this work the 3D CDNC fields obtained from the cloud activation model in MATCH-SALSA are now used in the RCA4 simulation.".
All the above suggestions are incorporated in the updated manuscript.

Page 903, Line 3: why you use 6-hourly CDNC to re-run RCA4? IN principle you can provide CDNC at higher time resolution (1 hour?). Is RCA4 interpolating between 6-hour values? Higher time frequency could improve the RCA4 simulation?
We used 6 hourly CDNC values to re-run RCA4 as this would reduce the post processing time significantly. One must note that the model is run for 8 years. The CDNC data is interpolated at every time step in the RCA4 model. This information would be added in the manuscript.

Page 903, Line 18: remove "precursor"
This is updated in the manuscript.

Page 903, Line 19: what is meant for physiography files?

The physiography files include the landmask, fraction of forest, low vegetation, surface roughness, different vegetation classes etc.

Page 903, Line 25-27: should be better explained how the first and second IAEs can be estimated separately, see also comment above.
Refer to Q.3 in the response to 'Main comments'.

Page 904, Line 6: Section 3.1, how is the monthly distribution of SO2 and PM total emissions?
Andersson et al., 2014 compared the annual mean distribution of aerosol number concentrations at several stations with observations. It was found that the regions of high aerosol number concentrations coincided with regions of high SOx emissions and was similar to the results obtained in previous studies (Spracklen et al., 2010, Yu and Lou, 2009).

Page 904, Line 17: To which figure do you refer with summer half of the year? JJA only? You should check throughout the text this expressions, you use summer or winter half of the year in other points, but the figures shows JJA DJF seasonal means.
The terminology 'summer half of the year' and 'winter half of the year' has been removed from the text and replaced by boreal summer months and boreal winter months or 'JJA means' and 'DJF means'.

Page 905, Line 1-2: It is not clear if you are commenting on the summer or autumn/winter distributions. The reference Yttri (2014) is not appropriate here, but you should check the EMEP emissions used in this specific study, to see which anthropogenic sector is mainly contributing to aerosol distribution in different seasons.
Residential biomass burning is more prominent in late autumn-winter-early spring months over eastern Europe and Russia. Whereas, biogenic VOC emissions are higher in these regions during summer season and this is reflected the in the seasonal distribution of cloud droplet number concentrations (Yttri, 2014). This is clarified in the text.

Page 905, Line 3-5. I would move this sentence at the beginning of the section.
It is updated in the manuscript.

Page 905, Line 6-12. Would be good to have also a comparison of MODIS vs simulation CDNC geographical distribution, to have a better understanding of the model performances.
Unfortunately, one-to-one comparison of CDNC from MODIS and model is very difficult. MODIS observes only few hundred meters of the cloud top and the retrievals are done for only fully cloud covered pixels. Emulating such conditions in the model without the use of satellite simulator is challenging. Therefore, we focus instead on evaluating LWP which is an integral measure and also tightly related to all other cloud microphysical properties.

Page 905, Line 14: replace with "Northern Atlantic" (also from Caption of Figure 5)
This is replaced in the updated manuscript.

Page 905, In the text and in the caption of Figure 6 it is not explained what is shown with the colorscale.
Page 906, Line 22: Also for Figure 8 caption and in the corresponding text, it is not explained exactly what is shown with the colorbar (Normalized frequency?).
The colorbar refers to the normalized frequency. This is clarified in the text and in the figure captions.

Page 906, Line 2-4: the authors claim the impact of transported pollution on the clouds in the North Atlantic, citing the HTAP project report. On the other hand, how the boundary conditions are simulated in the MATCH-SALSA for the aerosol and gases? This should be maybe added in the

simulation setup description. Is the main pollution transport pathways from North America to Europe located at such North latitudes, where R4 is also located?

The following text is added in the manuscript:

"The aerosol and gaseous concentrations at the lateral and top model boundaries are set as described in (Andersson et al., 2007). The boundary concentrations are based on both observations at background locations and large scale model calculations and are prescribed as monthly or seasonally varying fields. However, the boundary concentrations of organic matter (OM) are set to the seasonally varying mass size distributions and totals of marine OM as described in O' Dowd et al., (2004). The aerosol number concentrations are also introduced at the southern, western and northern lateral boundaries. These values are prescribed at the first model level and interpolated linearly to the top and eastern boundaries where the concentrations are set to zero."

 Page 908, Line 8-14: would be helpful to compare also the geographical distribution of simulated and observed LWP in order to see if the differences in the lower tail (50-100) is located are a specific region?

This is done in the manuscript.

Page 909, Line 15: consider rephrasing: "The present day (PD) perturbed case climate scenario is using the MOD simulation setup"

Page 909, Line 17: remove parenthesis from "(PD-PI) case" and replace with "PD-PI differences"

Page 909, Line 18-19: use "from the response" instead of "due to the response" In the same sentence, it is not clear what you mean by response of the land surface without other climate feedbacks.

Page 909, Line 21: "PI simulation" instead of "PI emissions"

Page 909, Line 26: "present day (PD)" instead of "present climate"

Page 909, Line 27: "The spatial distribution of these differences are reflected .."

Page 910, Line 6-9: The separate estimation of the first and second IAEs should be explained better as commented above.

Page 910, Line 14: a better reference should be included instead of "IPCC models", from which AR?

Page 910, Line 24-27: consider rephrasing of the sentence. It is long and difficult to read.

Page 911, Line 11: consider rephrasing "a more sophisticated representation of aerosol distribution (emissions, transport, and microphysical processes) can be included at a higher resolution . . ."

Page 911, Line 6, point 1, consider rephrasing "Investigate the improvements in a regional climate model simulation of the cloud microphysical properties, using spatially and temporally resolved 3D CDNC fields from a detailed aerosol and cloud activation model."

Page 912, Line 7-8: you should include the estimated numbers in parenthesis for the first and second IAEs.

All the above suggestions are incorporated in the revised manuscript.

The panels in Figures 3, 4, 7, 8, and 9, are small and should be enlarged in the figure

The resolution of Figure 2 is not very good, and difficult to read the text inside.

The resolution of Figure 8 is also low, and the axis labels and titles are difficult to read.

All the figures are revised now improving the clarity.

<u>Response to Reviewer #2</u>

We thank the reviewer for her/his comments. Please find below point-by-point response to the reviewer's comments. The major changes in the manuscript are highlighted.

Main comments:

1.Since there is no on-line coupling between the regional climate model and the chemistry-transport model (with aerosol dynamics), I suggest to remove the word "coupling" from the text to refer to the interactions between RCA4 and MATCH-SALSA,which is quite misleading from my point of view. The term "combination" is much more adapted to this methodology, as it is actually indicated in the title of the manuscript. However, there is a true online coupling between MATCH and SALSA.

We understand that this is not a fully online coupled model. In the text, we have tried to make this clear by either mentioning the word 'combining' or by specifically mentioning 'online' or 'offline' accordingly.

2.With regards to the estimation of the first and second indirect effects, I did not understand how the authors could quantify separately these two effects ? I think additional simulations were needed, but this should be stated more clearly.

This is now clearly mentioned in the text in Section 2.2 Experimental setup-2. Added as the following:

" To evaluate the individual contribution from the first and second indirect aerosol effects to the total radiative forcings, two additional simulations each (for PI and PD climate) were carried out. We turn off the individual IAEs by prescribing the constant CDNC values for the calculation of one IAE at a time, for example, to evaluate the sole contribution from the first IAE, 3D CDNC fields are used in the computation of CD radius to assess the changes in cloud albedo (1st IAE) and constant CDNC values are used in the scheme for the autoconversion process (2nd IAEs) and vice versa."

3. It would be interesting to have an idea of the cost of the different simulations, in order to know (1) if this modelling system can be used for multi-decadal simulations, and (2) if in future this coupling between RCA4 and MATCH-SALSA could be online.

A brief paragraph stating the costs is added in section 5 Conclusions. Read as the following "The calculations were performed on a HP Cluster Platform 3000 with SL230s Gen8 compute nodes, each with two 8-core Intel Xeon E5-2660 "Sandy Bridge" processors at 2.2GHz. Using three nodes and 48 MPI-ranks, a one year simulation with the online coupled MATCH-SALSA including the cloud activation module takes 20 hours (wall clock time). On the other hand, RCA4 takes approximately 1.5 hours for one year simulation using two nodes and 32 MPI-ranks. "

Specific comments:

- page 900 line 2: remove the bracket
- page 900 line 8: I wouldn't be so affirmative, I think indeed online integrated modeling is a relevant option to improve the representation of aerosols and chemistry in future models, but you should mention that it depends on the objective of the study.
- page 901 line 4: the coupling . . . is
- page 901 line 26: please define NMVOC and DMS.
- page 902 line 1: please define EC and OC.
- page 904 line 6: 4 should be an indice.

The comments mentioned above have been incorporated in the text.

- page 905 line 7-11: is it possible to have a more precise comparison with MODIS data ?

Unfortunately, one-to-one comparison of CDNC from MODIS and model is very difficult. MODIS

observes only few hundred meters of the cloud top and the retrievals are done for only fully cloud covered pixels. Emulating such conditions in the model without the use of satellite simulator is challenging. Therefore, we focus instead on evaluating LWP which is an integral measure and also tightly related to all other cloud microphysical properties. The spatial comparison of cloud liquid water path is additionally shown in the revised manuscript. It clearly shows improvements in MOD simulations compared to CTRL simulations. The LWP values are more realistically simulated and the LWP distribution is closer to MODIS in the MOD simulation. The LWP values are however still underestimated in the model over the southern parts of the study area.

- page 905 line 14: what does N. stand for ?
N stands for North. The term 'northern N. Atlantic' has been replaced by N. Atlantic.

- page 905 line 16: CDNC values are not always lower than 500 cm-3, notably in DJF Eastern Europe and JJA Central Europe. Is this value of 500 cm-3 very significant ?
The color scale shows the normalized frequency. So, darker the shading means the highest probability of observing those cloud droplet number concentrations. The value of 500 cm^{-3} is chosen because most predominantly, the regions that are relatively on the darker side mostly correspond with CDNCs below 500 cm^{-3}. It is not particularly a significant number.

- page 906 line 19-21: This sentence explains the decrease of droplet radii in summer compared to winter for the MOD simulation. However, it is not true for the CTRL simulation, how do you explain that droplet radii increase in summer, notably in northern Europe ?
These distinguishable features are not seen in the 'CTRL' simulation mainly because the CDNCs have constant values irrespective of the seasons.

- page 907 line 6: How has this threshold of 10 mm been fixed ? Do you have an idea of the impact of this choice on large scale precipitation ?
Numerous past studies suggest a threshold droplet radius for the onset of auto-conversion of cloud droplets to precipitating (falling rain) droplet) to be in the approximate range 10-13 microns . (e.g. Liu et al. 2003, Pawlowska et al 2003).

   The original critical threshold used in the NCAR GCM implementation of the Rash-Kristjansson parameterisation used in RCA4 was 5 microns. This low value was likely necessary because of the low resolution of the NCAR GCM and the use of grid box mean liquid water content (LWC) for both cloud microphysical and cloud-radiation calculations. Low values of the critical radius threshold result in frequent drizzle from clouds reducing grid box mean liquid water amounts and inducing an acceptable cloud-albedo based on (a biased low) grid box mean LWC. In RCA3, with a model resolution of 20-50km rather than ~200km in the NCAR GCM use of a 5micron threshold resulted in excessively frequent drizzle (of very low rates ~0.5 mm/day) which did not impact enormously on monthly precipitation accumulations but did impact negatively on e.g. frequency of wet days and through its impact on precipitation-LWC relationships also on cloud albedo and the 2nd indirect aerosol effect. Use of a 10 micron threshold reduces the frequency of low drizzle occurrences in RCA3 and also acts to increase the grid box mean LWC values. Use of a larger value of the critical threshold (beyond 10 microns) negatively impacts on precipitation rates by delaying the onset of rainfall while also leading to a positive bias in LWC and cloud albedo.

   We view 10 microns as a reasonable value for this threshold (based on theoretical studies and our own model sensitivity tests) but acknowledge that the actual value is very likely dependent on a multitude of factors, such as ambient pollution, cloud vertical motion and cloud temperatures.

All of these factors may have an influence through to the representation of the 2nd indirect effect but are presently beyond the ability of climate models to simulate.

Liu Y, Daum P and McGraw R. An analytical expression for predicting the critical radius in the autoconversion parameterization GEOPHYSICAL RESEARCH LETTERS, VOL. 31, L06121,

doi:10.1029/2003GL019117, 2004

Pawlowska, H., and J.-L. Brenguier (2003), An observational study of drizzle formation in stratocumulus clouds for general circulation model (GCM) parameterizations, J. Geophys. Res., 108, 8630, doi:10.1029/2002JD002679, D15.

- page 909 line 27: one word may be missing after "these"

The sentence is re-phrased as "The steep increase in the aerosol concentrations may be attributed to the increase in anthropogenic pollutant precusror emissions in these countries in the present day (PD). These differences seen in the spatial distribution are reflected as an increase of almost up to 70% increase in CDNCs and correspondingly, an increase of up to 10% in CLWP. "

- Figure 7: it would be better to keep the same color scale for the MOD and CTRL simulations, in order to make the comparison clearer for the reader.

If one uses the same color scale as the 'MOD' simulation for the 'CTRL' simulation in this figure one would not be able to see the variabilities in the 'CTRL' simulation as the CD radii ranges only from 4 to 5 mm whereas in the 'MOD' simulation, the values range from 4 to 13 mm. Hence, the figure is kept as it is.