Anonymous Referee #2 Authors' response

This manuscript, submitted to Geoscientific Model Development, presents a sensitivity study on the role of dynamic aerosols in regional climate simulations over Europe, carried out with the WRF model. The authors consider both present and future simulations, and discuss the role of aerosol-radiation and aerosol-cloud interactions respectively. They conclude that the response of downwelling surface shortwave radiation (rsds) to aerosols is mainly driven by the impact of aerosols on cloudiness. Overall this question is very interesting and needs to be studied, I found the present manuscript presents major problems of methodology, that is the reason why I would suggest not to publish it in GMD.

We do thank the reviewer for the time devoted to read and thoughtfully comment on our work. Below we provide detailed answers to each comment, hoping to have been clear enough in our explanations. Attending these comments and the ones posted by the other reviewers and the Editor, the new version of the manuscript:

1 – Has been entirely revised by a native speaker in order to improve the redaction.

2 – Has a new title. The change intends to avoid that the reader interprets that we are comparing simulations with dynamic vs. static aerosols. The new title is:

"Sensitivity of surface solar radiation to aerosol-radiation and aerosol-cloud interactions over Europe in WRFv3.6.1 climatic runs with fully interactive aerosols"

In this line, we have made an effort to make the scientific purpose of the manuscript clearer throughout the whole text.

3 – Includes further details and arguments on the experimental set-up and the methodology. Section 2 has been divided into 3 subsections.

4 – The formerly labeled as ACI simulations are now named ARCI to emphasize that these include both aerosol-radiation and aerosol-clouds interactions.

5 – Includes a brief validation exercise. We now face the outputs of our simulations with ERA5.

6 – Includes two subsections within the Results section: one for the historical simulations and another for the future projections.

7 – Includes a deeper discussion of the results attending several comments by the reviewers, e.g.:

- the activation of the autoconversion scheme in the ACI simulations hampers a direct attribution of the signals to the aerosol-cloud interactions from a physical point of view (the attribution can be made, from a modeling point of view, to the activation of these interactions in the model);
- the fact that we kept constant the anthropogenic aerosol emissions in future simulations permits to better isolate the signals from the aerosol-radiation-cloud interactions due to the so-called climate change penalty alone, while reduces the reliability of the future projections obtained;

• the signals obtained for different seasons (additional analysis is provided as Supplementary Material);

8 – Includes a link where all the data and codes to reproduce our study have been made publicly available: <u>http://doi.org/10.23728/b2share.682b1c6311134b36a18f59a99a443afd</u>.

We are confident that these major changes have improved significantly the manuscript and provides a larger support to its key findings.

Main comments:

- The authors are ambiguous about the objective of their study, to begin with the title. I do not understand if they want (1) to show the added values of representing interactive dynamic aerosols in regional climate simulations, compared to regional climate simulations with climatological aerosols, or (2) if they want to show the mean impact of aerosols in regional climate simulations compared to simulations which would not have any aerosols. Given the title, I was expected the first option, which is a very interesting question, not very much documented in literature, but this requires a rigorous protocol in which we compare regional climate simulations with the same aerosol content on average. This is not the case here. So I suppose the authors were in the second option, which is much less interesting, as it has already been studied in different publications. In that case, I suggest to remove the word dynamic from the title, and avoid overly affirmative expressions such as "a reduction about 5% in RSDS was found when aerosols are dynamically solved by the RCM".

We understand the title may lead to misinterpretations, so we changed it.

As we are comparing RCM outputs from simulations with and without aerosols, we were, effectively, in the second option mentioned by the reviewer. Although there exist previous works in this second option, none of them attempted to unveil the impact of aerosols from a purely modeling approach such as the one used here, where no prescribed aerosol concentrations are used. So the word 'dynamic' actually makes the difference with previous studies, that's why we included it in the title.

- Another major concern about this study is the fact that the authors draw conclusions on the impact of aerosols on rsds future evolution, while they keep constant anthropogenic emissions in their future simulation. The authors are aware of discrepancies in the rsds future evolution between global and regional climate simulations, which could be due to the use of constant aerosols in RCMs contrary to GCMs (Boé et al. 2020). That is the reason why I do not understand the authors keep anthropogenic aerosol emissions constant in future simulations, while they should evolve as in the GCM simulation.

As stated above, this approach permits to better isolate the signals from the aerosol-radiation-cloud interactions due to the so-called climate change penalty alone, while reduces the reliability of the future projections obtained, in fact. We now we make more emphasis on this point in the manuscript.

- The last major concern is about the RCM used in this study. The version of WRF used here, namely 3.6.1 is quite old (reference paper from 2008), and above all a precise description of how aerosols and their effects on climate are represented is missing. For example, I wonder what aerosol climatology is used in the BASE simulation (if it is not zero). I am also very worried about the very low values of summer AOD shown in Figure 1g-h, which shows that WRF clearly underestimates AOD over Europe. WRF values range from 0.05 to 0.09 over

Europe, while observations typically range from 0.1 to 0.2 (Papadimas et al. 2008, Nabat et al. 2013, Schultze and Rockel 2018). That could lead to an underestimation of aerosol effects. In such a study, an evaluation of AOD (even brief) is needed in order to ensure the consistency of the results.

Some of the simulations included in this work were performed time ago. Others have been performed more recently, but we decided not to change the WRF version to be sure of being comparing the same "thing". At that time, when first simulations were carried out, the last stable version of WRF was the 3.6.1. In any case, the physics of the model is the same, no matter of its version.

Addressing the appropriate reviewer concern about the lack of a deep description of how aerosols are treated by the model, we have now added more details on that. For instance, regarding the BASE experiments, we now say in section 2:

"BASE: aerosols are not considered in the simulations. No aerosol climatology is used and no aerosol interactions are taken into account by the model. WRF-alone considers a constant number of cloud condensation nuclei (250 per cm³, set in the model by default) to enable the formation of clouds."

Regarding the ARCI experiments, we now explain in section 2:

"Aerosol-cloud interactions were implemented by linking the simulated cloud droplet number with the microphysics schemes (Chapman et al 2009) affecting both the calculated droplet mean radius and the cloud optical depth. Although this WRF-Chem version (3.6.1) does not allow a full coupling with aerosol-cloud interactions, the microphysics implemented here is a single moment scheme that turns into a two moments scheme in the simulations denoted as ARCI. One-moment microphysical schemes are unsuitable for assessing the aerosol-clouds interactions as they only predicts the mass of cloud droplets and does not represent the number or concentration of cloud droplets (Li et al. 2008). The prediction of two moments provides a more robust treatment of the particle size distributions, which is key for computing the microphysical process rates and cloud/precipitation evolution. In this sense, although the Lin microphysics is presented as a single moment scheme, the WRF-Chem model allows to transform the single into a double moment scheme. A prognostic treatment of cloud droplet number was added (Ghan et al. 1997), which treats water vapour and cloud water, rain, cloud ice, snow, and graupel. The autoconversion of cloud droplets to rain droplets depends on droplet number (Liu et al. 2005). Droplet-number nucleation and (complete) evaporation rates correspond to the aerosol activation and resuspension rates. Ice nuclei based on predicted particulates are not treated. However, ice clouds are included via the prescribed ice nuclei distribution following the Lin scheme. Finally, the interactions of clouds and incoming solar radiation have been implemented by linking simulated cloud droplet number with the Goddard shortwave radiation scheme, representing the first indirect effect, and with Lin microphysics, which represents the second indirect effect (Skamarock et al. 2008). Therefore, droplet number will affect both the calculated droplet mean radius and cloud optical depth."

References:

Ghan, S. J., Leung, L. R., Easter, R. C., & Abdul Razzak, H. (1997). Prediction of cloud droplet number in a general circulation model. *Journal of Geophysical Research: Atmospheres*, *102*(D18), 21777-21794.

Li, G., Wang, Y., & Zhang, R. (2008). Implementation of a two moment bulk microphysics scheme to the WRF model to investigate aerosol cloud interaction. *Journal of Geophysical Research: Atmospheres, 113*(D15).

Lin, Y. L., Farley, R. D., & Orville, H. D. (1983). Bulk parameterization of the snow field in a cloud model. Journal of Climate and Applied Meteorology, 22(6), 1065-1092.

Liu, Y., Daum, P. H., & McGraw, R. L. (2005). Size truncation effect, threshold behavior, and a new type of autoconversion parameterization. *Geophysical research letters*, *32*(11).

Mitchell, D. L., Rasch, P., Ivanova, D., McFarquhar, G., & Nousiainen, T. (2008). Impact of small ice crystal assumptions on ice sedimentation rates in cirrus clouds and GCM simulations. *Geophysical research letters*, *35*(9).

Skamarock, W.C.; Klemp, J.B.; Dudhia, J.; Gill, D.O.; Barker, D.M.; Wang, W.; Powers, J.G. A Description of the Advanced Research WRF Version 3; Technical Report NCAR Tech. Note TN-475+STR; NCAR: Boulder, CO, USA, 2008.

Tao, W. K., Simpson, J., & McCumber, M. (1989). An ice-water saturation adjustment. *Monthly Weather Review*, *117*(1), 231-235.

And acknowledge in the discussion section:

"In the ARCI simulations, the autoconversion scheme called so that cloud droplets can turn into rain droplets is different to the autoconversion scheme activated in the ARI simulations. This change in the WRF-Chem configuration can lead to ARCI-ARI differences that do not come necessarily from the of the aerosol-cloud interactions from a physical point of view (Liu et al 2005). In fact, the activation of the aerosol-cloud interactions requires further changes in the model configuration (as compared to the configuration used for the simulations labeled as ARI) beyond the autoconversion scheme, such as the activation of aqueous chemistry processes, that could also have an added impact to effect that can be strictly attributed to the aerosol-cloud interactions. However, technically, the encoding of WRF-Chem model hampers to better isolate the effect of the aerosol-cloud interactions from a purely physical point of view, but to the activation of the aerosol-cloud interactions from a modeling point of view, since the autoconversion schemes necessarily change between ARI and ARCI. This should be beared in mind when interpreting the signals."

Reference:

Liu, Y., Daum, P. H., & McGraw, R. L. (2005). Size truncation effect, threshold behavior, and a new type of autoconversion parameterization. *Geophysical research letters*, *32*(11).

Regarding the inclusion of gas-phase chemistry in the simulations, in section 2 we added:

"Chemical reactions in the GOCART model include several oxidation processes by the three main oxidants in the troposphere: OH, NO3, and O3. The OH radical dominates oxidation during the daytime, but at night its concentration drops and NO3 becomes the primary oxidant (Archer-Nicholls et al., 2014). So, the oxidation pathways represented in GOCART include: (a) the dimethyl sulphide (DMS) oxidation by the hydroxyl radical (OH) during the day to form sulphur dioxide (SO₂) and methanesulfonic acid (MSA); (b) the oxidation by nitrate radicals (NO3) at night to form SO₂; and (c) the SO₂ oxidation by OH in air and by H_2O_2 and tropospheric ozone (O₃) in clouds (aqueous chemistry) to form sulphate (Chin et al., 2000). Henceforth, the skilful characterization of gas-phase radicals such as OH and NO3 or compounds like O₃ is essential for the representation of oxidation pathways in the atmosphere leading to the formation of secondary aerosols (Jiménez et al., 2003). Therefore, in this contribution the RACM (Stockwell et al., 1997; Geiger et al., 2003) mechanism has been coupled to GOCART trough the kinetics pre-processor (KPP) in WRF-Chem in order to provide the concentrations of radical and gas-phase pollutants needed by the GOCART aerosol model."

References:

Archer-Nicholls, S., Lowe, D., Utembe, S., Allan, J., Zaveri, R. A., Fast, J. D., Hodnebrog, Ø., Denier van der Gon, H., McFiggans, G. (2014). Gaseous chemistry and aerosol mechanism developments for version 3.5.1 of the online regional model, WRF-Chem. *Geoscientific Model Development*, *7*, 2557–2579.

Chin, M., Rood, R.B., Lin. S.-J., Müller, J.-F., Thompson, M. (2000). Atmospheric sulfur cycle simulated in the global model GOCART: Model description and global properties. *Journal of Geophysical Research*, 105(D20), 24671-24687.

Geiger, H., Barnes, I., Bejan, I., Benter, T., & Spittler, M. (2003). The tropospheric degradation of isoprene: an updated module for the regional atmospheric chemistry mechanism. *Atmospheric Environment*, 37, 1503 - 1519.

Jiménez, P., Baldasano, J.M., Dabdub, D. (2003). Comparison of photochemical mechanisms for air quality modeling. *Atmospheric Environment*, 37, 4179-4194.

Stockwell, W. R., Kirchner, F., Kuhn, M., & Seefeld, S. (1997). A new mechanism for regional atmospheric chemistry modeling. *Journal of Geophysical Research: Atmospheres*, 102, 25 847 - 25 879.

Finally, regarding the 'low' AOD values shown in Figure 1g-h, we must acknowledge that we certainly used wrong AOD values. These had been computed from the TAUAER3 and TAUAER4 variables, which do exhibit a weird evolution along the year. After inspection, we figured out that these and the EXTCOF55 variables had been wrongly recorded in the wrfout files (not new, apparently, see e.g.: <u>https://forum.mmm.ucar.edu/phpBB3/viewtopic.php?t=9313&p=17464</u>). So we have now adopted an alternative method to compute AOD following Palacios-Peña et al (2020), where, in fact, the representation of AOD by these model configurations (ARI and ARCI) were deeply evaluated. The new AOD files were estimated using the reconstructed mass-extinction method (Malm et al 1994) from the well-recorded concentrations of the various aerosol species in the wrfout files, namely: black carbon, organic carbon, dust and sea salt. Sulfates were estimated from SO2 and OH recorded concentrations using the same kinetic reaction as the one implemented in the RACM-KPP module. We want to remark that the mistake occurered during the postprocessing of the wrfout files, while WRF-Chem run satisfactorily. These wrfout files were removed after postprocessed, so we have now generated a sample one (using the ARI configuration) and uploaded it for checking together with all the other data files. Importantly, this change in the methodology for estimating AOD values did not alter the the overall results of the paper. Attached here a figure with the AOD climatologies from ARI and ARCI and those from MACv2.

References:

Malm, W. C., Sisler, J. F., Huffman, D., Eldred, R. A., & Cahill, T. A. (1994). Spatial and seasonal trends in particle concentration and optical extinction in the United States. *Journal of Geophysical Research: Atmospheres*, *99*(D1), 1347-1370.

Palacios-Peña, L., Montávez, J. P., López-Romero, J. M., Jerez, S., Gómez-Navarro, J. J., Lorente-Plazas, R., ... & Jiménez-Guerrero, P. (2020). Added Value of Aerosol-Cloud Interactions for Representing Aerosol Optical Depth in an Online Coupled Climate-Chemistry Model over Europe. Atmosphere, 11(4), 360.

Other comments:

- page 2 line 31: land use change is not specific to regional climate simulations, I think it is even more used in global climate simulations.

We simply intend to note that at higher resolutions, land uses can be represented at a finer scale.

- page 3 lines 57-63: please avoid such long lists of references, and clarify the conclusions of each of them.

This paragraph has been reformulated accordingly.

- page 3 lines 70-71: "which still remain largely a mystery". Other studies such as Giorgi et al. (2016), Sørland et al. (2018) and Boé et al. (2020) have also underlined differences between RCMs and GCMs in future projections. The role of aerosols is even discussed in Boé et al. (2020), which should be mentioned here.

In fact. This is now acknowledged in the paper.

- page 5 lines 140-141: it is not clear for me how aerosol-cloud interactions are represented in the simulations.

We now provide further details in the manuscript. See our response about this above.

-page 6 lines 144-146: This way of calculating clear-sky variables in simulations is not common in modeling studies. It would be appropriate for a comparison to observations (it is exactly how satellites do for example), but in models, you generally compute clear-sky variables at each time step, removing clouds in radiative transfer. This would avoid the numerous missing values.

Unfortunately, we did not save clear-sky values from the model outputs, so we needed to adopt an alternative methodology.

- page 6 section 3: This section should be divided in several sub-sections, with more precise titles than only "Results".

Done.

- page 6 line 165: "The inclusion of interactive aerosols reduce the JJA mean values of RSDS". This is typically an example of my first main comment. This decrease in rsds is likely due to the mean effect of aerosols, and not their interactive pattern.

We agree that the sentence was misleading and has been reformulated. Although that statement is true in the context of our work, it wrongly gave the message that such a reduction in RSDS is due to the interactive aerosols modeling approach adopted here as compared to a more conservative (and common) approach based on non-interactive aerosols, which is something that we did not inspect.

- page 7 line 172: "ARI and ACI lead to more cloudiness in central and northern regions". This is not really the case when looking at the figure.

Blue colors prevail in central and northern regions in Figure 1d-f indeed. Please note that the color palette is inverted here with the aim of facilitating a visual identification of the matching between the patterns of the drivers (CCT, AOD) and those of RSDS.

- page 7 lines 184-187: This conclusion is not justified.

Both the spatial correlations shown in the previous Supp. Figure 4 and the temporal correlations shown in the previous Supp. Figure 5 (Supp. Figure numbers changed in the new version) support that the CTT differences prevail over the AOD differences in driving the RSDS differences between pairs of experiments, not only in the present-day climate simulations, but also under future conditions.

- Figures 1-4: From my point of view it would be easier to understand to have differences in absolute values rather than in percentages. Indeed, I suspect here we look at very low values which could be unsignificant.

Figures 1 to 4 have been replicated to show plain differences. These new figures have been included as Supp. Material and used to describe the results.

- Figures 1-4: Why consider only land points ? It would be interesting to show also ocean points on figures.

We prefer to focus only on land points to get a clearer message tailored to the modeling applications for the solar energy sector.

- Figure 3: When comparing the evolution of rsds, cct and aod in the simulations, I suspect a possible bug in the figure or in the simulation. Indeed, the strong decrease in rsds in northern latitudes (for example in Iceland), is neither explained by cct nor by aod.

The negative signals in rsds must be related to the increase in cct, even if small. Maybe better to see the plots with the differences in absolute values provided in the new version of the Supp. Material. This is also supported by the high spatial correlations between rsds and cct changes.

- Page 8 lines 218-219: If "the anthropogenic component is disregarded", there should be no possible conclusion on the future evolution of rsds.

We do not attempt to provide reliable projections for RSDS, but to unveil how aerosols can affect them. See our response to the main comment #2.

- The manuscript suffers from many typographical and English spelling errors that need to be corrected.

The manuscript has been entirely revised with the help of a native speaker.

AOD climatologies for 1991-2010

