General comments:

Review of "Spatial variability and future evolution of surface solar radiation over Northern France and Benelux: a regional climate model approach"

by Gabriel Chesnoiu, Isabelle Chiapello, Nicolas Ferlay, Pierre Nabat, Marc Mallet, and Véronique Riffault

Dear Referee #2,

Thank you for your suggestions and remarks. Consideration of these comments has helped to improve the manuscript. Below you will find the answers to each comment.

The authors present results evaluating the performance of the regional climate model CNRM-ALADIN64 comparing the simulations to observations of surface radiative fluxes and aerosols properties focusing on the west-European area over Benelux and Northern France (BNF) and provide historical period, mid-term and long-term future scenarios. The analysis evaluates the simulations from ALADIN hindcast from 2010 to 2020 allowing the comparison of observations at several sites within the BNF region. Their methodology starts with the assessment of clear-sky frequency based on the methodology of Long and Ackerman (2000) method considering the annual cycle variation at three different sites. Then they focus on the analysis of surface solar radiation by later delving into the annual variation of aerosol properties. Then the paper puts the spatial variability into context for the period 2010-2020. Finally, the manuscripts report the future evolution of two scenarios by presenting mean statistics and illustrating the spatial differences.

The manuscript has a comprehensible structure, makes use of valid methods and provides mostly well-documented and clear explanations of their assumptions and limitations. I consider the manuscript to be suited for publication after the following revisions are addressed.

Major comments

Identification of clear-sky situations

The overall analysis is clearly described, but not so much is discussed on other implemented methods that could have been used at the mentioned sites. It is not expected to reformulate the methodology but to better justify the selection of the methodology of Long and Ackerman (2020). More discussion

is needed. The authors can refer to the following references for example, or any other the authors might see suitable:

M.J. Reno, C.W. Hansen, Identification of periods of clear sky irradiance in time series of GHI measurements, Renew. Energy 90 (2016) 520–531, https://doi.org/10.1016/j.renene.2015.12.031.

Elias, T., Ferlay, N., Chesnoiu, G., Chiapello, I., and Moulana, M.: Regional validation of the solar irradiance tool SolaRes in clear-sky conditions, with a focus on the aerosol module, Atmos. Meas. Tech., 17, 4041–4063, https://doi.org/10.5194/amt-17-4041-2024, 2024.

Al Asmar, L.; Musson Genon, L.; Eric, D.; Dupont, J.C.; Sartelet, K. Improvement of solar irradiance modelling during cloudy sky days using measurements. Sol. Energy 2021, 230, 1175–1188.

The referee is right, the selection of the Long and Ackerman method needs more discussion since numerous methods exist in the literature for the identification of clear-sky conditions.

Our choice is based on the method's limited number of input parameters (solar zenith angle, global and diffuse SSR) and high adaptability, as it automatically adjusts to the specific conditions of any observational station. This means its application could easily be extended to additional stations where only global and diffuse SSR measurements are available.

In addition, our choice is based on the results of the comparative study of Gueymard et al. (2019), which showed its high precision for the identification of clear-sky conditions. The method notably achieved the second lowest "false positive" score (i.e. percentage of cloudy situations identified as clear-sky) of 7.25%, despite not depending on collocated photometric measurements or clear-sky simulations.

The introductory paragraph of Section 3.1.1 (see lines 279-287 of the revised manuscript) has been modified as follows (changes are in bold text):

"**Although numerous methods have been described in the literature (Reno and Hansen, 2016; Gueymard et al., 2019; Al Asmar et al., 2021), our study relies on** the well-established method of Long and Ackerman (2000) to distinguish clear and cloudy situations based on high frequency (3 minutes or less) ground measurements of global and diffuse surface solar radiation. **This method, which has been used for numerous studies (e.g. Elias et al. (2024)), was chosen for its limited number of input parameters (solar zenith angle, global and diffuse SSR) and high versatility, as it automatically adapts to the specific conditions of any observational station equipped with measurements of both global and diffuse**

horizontal irradiances. Our choice is also based on the results of the comparative study of Gueymard et al. (2019), which showed its high precision for the identification of clear-sky conditions. The method notably achieved the second lowest "false positive" score (i.e. percentage of cloudy situations identified as clear-sky) of 7.25%, despite not depending on collocated photometric measurements or clear-sky simulations."

Reduction of ammonia

In the paper it was mentioned a reduction of 25 % applied to all monthly ammonia emissions. Despite this reduction, nitrate aerosol concentration remain overestimated by the model. Is this due to a parameterization or an assumption within the model?

As mentioned lines 181-182 of the submitted manuscript, the reduction of 25% of ammonia emissions represents a compromise between reducing the overestimation of the AOD in spring and maintaining realistic nitrate concentrations throughout the rest of the year. It is thus to be expected that the model continues to overestimate the contribution of nitrates in spring.

This overestimation could be linked to the emission inventories used for the simulations, which, despite important efforts from the community, still feature significant uncertainties in ammonia emissions, especially at the local scale (Hoesly et al., 2018).

The overestimation could also be linked to the simplified chemical representation of nitrates within the model described in detail by Drugé et al. (2019). In particular, the variability of nitric acid (HNO_3, precursor of nitrate aerosols) defined in ALADIN is based on a fixed monthly climatology taken from CAMS Reanalysis data over 2003-2007, and does not account for the inter-annual variability of the species. Furthermore, due to the low vapour pressure of sulfuric acid (precursor of sulfate aerosols), the formation of ammonium sulfate takes priority over ammonium nitrate formation. Nitrate aerosol concentrations are thus dependent on the variability and uncertainty of sulfuric acid, as the nitric acid can only interact with the ammonia that remains after formation of sulfate aerosols.

References:

Hoesly, R. M., Smith, S. J., Feng, L., Klimont, Z., Janssens-Maenhout, G., Pitkanen, T., Seibert, J. J., Vu, L., Andres, R. J., Bolt, R. M., Bond, T. C., Dawidowski, L., Kholod, N., Kurokawa, J.-I., Li, M., Liu, L., Lu, Z., Moura, M. C. P., O'Rourke, P. R., and Zhang, Q.: Historical (1750–2014) anthropogenic emissions of reactive gases and aerosols from the Community Emissions Data System (CEDS), Geoscientific Model Development, 11, 369–408, https://doi.org/10.5194/gmd-11-369-2018, 2018.

Drugé, T., Nabat, P., Mallet, M., and Somot, S.: Model simulation of ammonium and nitrate aerosols distribution in the Euro-Mediterranean region and their radiative and climatic effects over 1979–2016, Atmospheric Chemistry and Physics, 19, 3707–3731,https://doi.org/10.5194/acp-19-3707-2019, 2019.

Can the authors comment how future work should consider similar/higher reduction?

The chosen reduction factor is strongly influenced by the selected emission inventories, the specific parametrization defined in the model for the formation of nitrate aerosols, and the specific location of the simulations. The Benelux/North of France region is largely impacted by agricultural activities and corresponding ammonia emissions, and the influence of these parameters can significantly fluctuate between regions of the world. Hence, we cannot certify that the reduction used in the present study can be generalized.

We recommend that future studies involving other models, regions or study areas carefully assess nitrate aerosol simulations, which can have a decisive impact on the accuracy of other simulated parameters such as the AOD and SSR.

Was a sensitivity analysis made varying the concentration of ammonia? Or should this be recommended?

In our study, the reduction factor has been the subject of an extensive sensitivity analysis in which different reduction factors as well as several emission inventories have been tested.

This sensitivity analysis has shown that overall a reduction factor of 50% enables a better description of the mean AOD in spring. However, it also highlighted that such a reduction factor leads to an important underestimation of the AOD in summer. In this context, we chose a more reasonable reduction factor of 25%, which gives the best results over the year, as it represents a compromise between reducing the overestimation of the AOD in spring and maintaining realistic nitrate concentrations throughout the rest of the year.

Note that while a specific 50% reduction factor could potentially be applied only to springtime ammonia emissions for optimal results, this approach is deemed precarious. Therefore, we opted for a consistent correction factor across all months.

Clarifications on the choice of the reduction factor have been added to the text (see lines 197-201 of the revised manuscript). The changes are summarized below (in bold text):

“The choice of the reduction factor has been the subject of an extensive sensitivity analysis. The retained adjustment factor **of 25\%, specific to our study,** represents a compromise between reducing the overestimation in spring and maintaining realistic nitrate concentrations throughout the rest of the year. It can be emphasized that such corrections are consistent with current uncertainties in ammonia emissions, which remain significant, especially at the local scale (Hoesly et al., 2018).”

Spatial variability

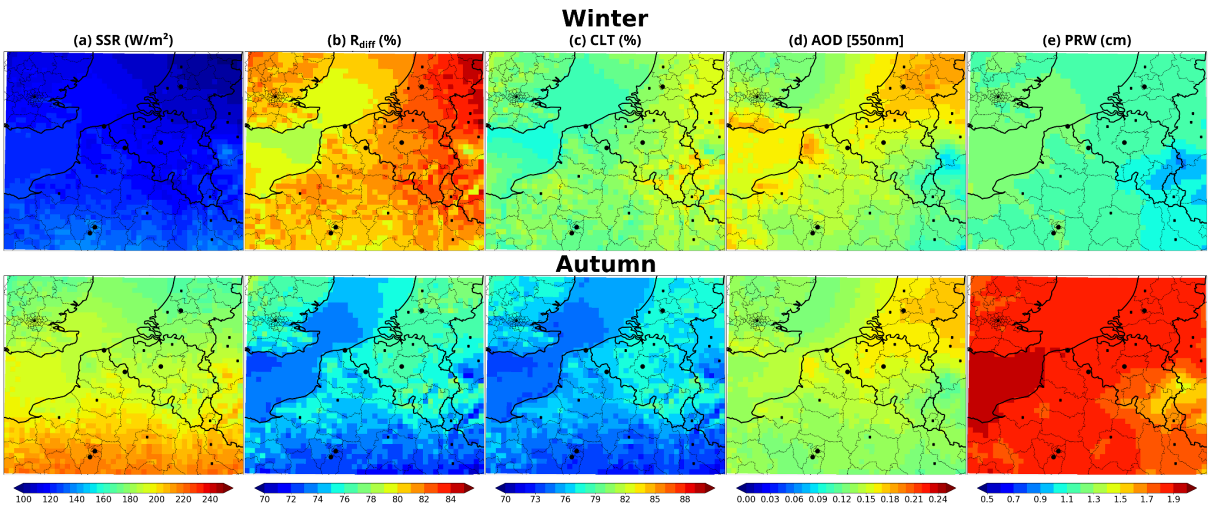
It is understandable the need to deepen the analysis for Spring and Summer seasons for the spatial variability analysis. However, including the analysis illustrated in Figure 7 for Autumn and Winter will enrich the overall analysis and further interpretation with Figure 3 and Figure 4a.

In response to the referee’s comment the following figure has been added to the supplements (Fig. S7 of the revised manuscript).

In addition, a few comments have been included in the main text of Section 4.1 (see lines 483-489) to highlight similarities and discrepancies between spring/summer seasons (previously Fig. 7, now Fig. 8) and winter/autumn (Fig. S7).

The added comments are transcribed below:

“Corresponding simulations of the spatial variability of SSR and associated atmospheric parameters for winter (i.e. December-January-February) and autumn (i.e. September-October-November) seasons are reported in the supplements (Fig. S7). For these two seasons, the spatial patterns are similar to those observed in spring and summer (Fig. 8). However, as expected, AOD ranges are significantly reduced over most of the BNF region, together with increased simulated CLT levels, from 72% to more than 80%. Simulated SSR are largely reduced, below 150 $W.m^{-2}$ in winter, and 220 $W.m^{-2}$ in autumn. Thus, in order to evaluate the impacts of contrasted anthropogenic aerosol future emissions on the high-end range of SSR, we will focus on spring and summer seasons.”



Technical/Minor comments:

Follow ACP guidelines to refer figures in the text. For instance, change (Figure 1) to (Fig. 1).

All references of figures inside parentheses in the text have been changed according to the ACP guidelines.

Correct units. They should be written exponentially in the text, tables and figures.

Units are now written exponentially in the text, tables and figures.

Homogenize how to address chemical species. For example, nitrate (NO3) is defined more than one time.

Some chemical species were indeed defined several times.

Definitions of all acronyms, including for chemical species, have been checked and redundancies have been addressed.

|| = line

|| 1 … change spatio-temporal to spatiotemporal as it was done later in the text

Done.

|| 34 ... increases in photovoltaic

Following a comment from the other referee, the sentence has been changed as follows (see line 36 of the revised manuscript, changes are in bold text):

"In the context of climate change that requires **an** increase of photovoltaic **energy production (relevant for the energy transition)**,[...]"

|| 40 correct sentence… assessment of aerosols’ future evolutions in time or assessment of the future evolution of aerosols in time

The sentence has been changed as follows (see line 53 of the revised manuscript, changes are in bold text with a green background):

“[...] assessment of aerosols**’** future evolutions in time,[...]”

|| 77 ... climate model and the two kinds of

Following the referee’s comment, the sentence line 70 has been changed from “[...] climate model and of the two kinds of [...]” to “[...] climate model and the two kinds of [...]” (see line 83 of the revised manuscript).

|| 88 … Close parenthesis SURFEX (SURFace EXternalisée, Masson et al. (2013))

Done.

|| 148 … A first dataset → The first dataset

Done.

|| 168 remove double parenthesis after van Marle et al. (2017)

Done.

|| 170 … scenarios are

There was an extra “s”. The appropriate spelling should read “the SSP1-1.9 scenario is [...]”. The text has been modified accordingly (see line 187 of the revised text) and the rest of the manuscript has been double checked following comments from both referees.

|| 171 … greenhouse gases emissions → greenhouse gas emissions

Done.

|| 187 BNF region cf Figure 1 → BNF region Fig. 1

Done.

|| 210 Could you add a reference in line 210? or specify that to the best of your knowledge you decide to go for those uncertainties.

The cited uncertainties refer to the results of Vuilleumier et al. (2014) mentioned line 208. However, as the referee pointed out, it wasn’t clear.

For clarity, the reference has been added at the end of line 227 of the revised manuscript.

|| 224 include space ... 550nm → 550 nm

Done.

|| 269 CLT already defined in line 247

Following a previous comment of the referee, definitions of all acronyms have been homogenized. The iteration of “CLT” line 269 has been removed (see line 297 of the revised manuscript).

|| 388 The comparisons shown in Figure 6b

The preposition “in” has been added as advised by the referee (see line 416 of the revised manuscript).

|| 388:390 Improve clarity. The sentence is too long. ‘The comparisons shown in Figure 6b highlight that the underestimation of organic and black carbon aerosols is partially offset in spring and summer by a coincident overestimation of nitrate aerosol concentrations, especially in March and April (around +2 µg/m3), despite the application of a 25% correction factor on ammonia emissions, the main precursor of nitrate aerosols.’

The sentence was indeed too long. It has been changed as follows (see lines 416-419 of the revised manuscript, changes are highlighted in bold text):

“The comparisons shown **in** Figure 6b highlight that the underestimation of organic and black carbon aerosols is partially offset in spring and summer by a coincident overestimation of nitrate aerosol concentrations**, despite the application of a 25% correction factor on ammonia emissions, the main precursor of nitrate aerosols. This offset is especially significant in March and April with differences in total concentrations of around +2 µg/m3 between the model and the measurements.**”

|| 410:411 Correct description. Panel (d) is AOD and panel (c) is CLT

Indeed, the description of the panels was not correct. It has been corrected (see line 452 of the revised manuscript).

|| 467 Is it BC or equivalent BC?. Keep it consistent along the entire manuscript.

The species mentioned line 467 is BC. The term “equivalent BC” is generally used in the literature to refer to the concentrations derived from the aethalometer, such as the one used for the evaluation of ALADIN HINDCAST simulations in Section 3.3.

This is why the term “equivalent BC, or eBC”, has been initially defined in section 2.2.2 (line 236 of the submitted manuscript). However, although it was not properly stated, the term “eBC” was replaced with more simply “BC” for consistency with ALADIN simulations.

To avoid confusions, this change in terminology is now clearly stated in the revised manuscript (see lines 255-257, Section 2.2.2) as follows:

“For consistency with the terminology of ALADIN simulations, equivalent concentrations of black carbon (i.e. eBC) derived by the aethalometer are hereafter referred to simply as BC.”

|| *534 change spatio-temporal to spatiotemporal*

Done.

|| *545 AOD already defined in line 219*

To make the article easier to read, we find it preferable to redefine all acronyms within the conclusions, even though they were defined earlier in the text.

Although it might be obvious, clarify which months the authors consider for their Spring and Summer comparison.

Spring refers to March-April-May (MAM) and summer to June-July-August (JJA).

Both seasons are now defined in the introduction (line 89 of the revised manuscript).

Comments on Figures

Figure 3, Figure S1, Figure S2 and Figure S3

While the lines can be differentiated, the description says green line, but to me it looks blue. Could you change the color of ‘Estimate from ALADIN simulations’?

The color of “Estimate from ALADIN simulations" has been changed to a more noticeable shade of green in Figures 3 (page 12 of the revised manuscript), S1, S2 and S3 (pages 1 and 2 of the revised supplements). In addition, following a comment of the other referee, the monthly mean difference between ALADIN simulations and ground measurements has been added to Figures 3, S1, S2 and S3. For consistency, the color of ALADIN simulations in Figure 6 (page 17) has also been changed.

Figure 8, 9, 10, 11 Long term evolution for SSP3-7.0 Is it possible to include a separated colorbar for the lowermost right-side panel? In case it messes up the structure of the figure, perhaps include an adequate colorbar for these parameters in the appendix?

Following the referee's advice, a distinct figure has been added to the revised supplements (see Figure S10, page 7 of the revised supplements). The figure includes a modified version of the lowermost right-side panels of Figures 8-11 (now Figs. 9-12) with a specific colorbar.