Anonymous Referee #1.

General comments:

Rev. #1: From my understanding, the paper "Assessment of the radiative effects of aerosol in an on-line coupled model over the Iberian Peninsula" presents results of a numerical study focused on the sensitivity of atmospheric aerosol particles optical properties over Iberian Peninsula (IP), nominally Aerosol Optical Depth (AOD), Ångström Exponent (AE), and backscattering vertical profile, to the feedbacks induced by the aerosol direct and indirect radiative effects. […] The study is focused on two distinct aerosol scenarios over IP, one consists of an episode of Saharan dust transport toward the southern region of IP, and the second of a biomass burning event that occurred in the north region of Portugal. Although the scientific goal of the paper is of significant relevance, the paper exposure needs improvements.

A: First, we would like to thank the anonymous referee #1 for their valuable comments in the interactive comment on "Assessment of the radiative effects of aerosols in an on-line coupled model over the Iberian Peninsula" by Laura Palacios-Peña et al. (No. acp-2016-473). The manuscript has been revised after reviewer's comments in order to correct errors and to introduce the reviewer' suggestions for improving the quality of the paper. Please see below our point-by-point replies:

Rev. #1: Beginning from the title, in my view, the authors did not assess the aerosols radiative effects as the title suggest. So far, their major focus has been on the response of aerosol optical properties field over IP, mainly AOD and EA, to the on-line coupling of the aerosol radiative effects in the model. Therefore, I think there is a need to adequate the title in order to accurately express the paper goal and content.

A: As suggested by the Rev. #1, the title has been changed by "Evaluating the representation of aerosol optical properties by an on-line coupled model over the Iberian Peninsula".

Rev. #1: For the sake of clarity, the authors should make clear the distinction between what they refer as radiative feedbacks and the aerosol radiative effects.

A: An effort has been made in the abstract and the introduction to define radiative effects and radiative feedbacks. Aerosol radiative effects refer to direct and semi-direct effects, produced by the aerosol-radiation interactions (ARI); and indirect effects, produced by aerosol-cloud interactions (ACI) (as described in the submitted manuscript). These radiative effects produce feedbacks to meteorology/emissions, which are called aerosol radiative feedbacks.

Rev. #1: Given the role of the numerical experiments on the manuscript goals and conclusions, there is not much discussion on the model system and simulations configurations, physical and chemical modules, leaving it to references. Further details are needed, especially as regard to the model aerosol microphysical and optical modules, which have relevant impact on the variables analysed, and indirect and direct radiative effects parametrization. Also, there is not much discussion about the mechanisms that drive the feedbacks induced by the online simulation of the aerosol direct or indirect radiative effects on the aerosol field over IP.

A detailed description of the aerosol, microphysical and optical modules as well as the previous description of the representation of aerosol-radiation-clouds interactions has been included in the revised version of the manuscript:

"The most important configuration to bear in mind for this work is the aerosol module. This aerosol module is based on the modal aerosol MADE (Modal Aerosol Dynamics Model) (Ackermann et al., 1998) which is a modification of the Regional Particulate Model (Binkowski and Shankar, 1995). Here aerosol particles are represented by three log-normal size distributions, corresponding to an Aitken mode (nucleation mode 0.1 µm diameter), and accumulation mode (0.1 – 2 µm), and a coarse mode (> 2 µm) (Forkel et al. 2012). SOA have been incorporated into MADE in the SORGAM (Secondary Organic Aerosol Model) module (Schell et al., 2001).

Aerosol chemical properties and sizes are used to determine aerosol optical properties as a function of wavelength using the method outlined in Fast et al. (2006) and Barnard et al. (2010). In brief, each chemical constituent of the aerosol is associated with a complex index of refraction. The overall refractive index for a given size bin is determined by volume averaging, with Mie theory and summation over all size bins used to determine composite aerosol optical properties. Wet particle diameters are used in the calculations (Chapman et al. 2009).

The microphysical module consist of the Lin scheme based on Lin et al. (1983) and Rutledge and Hobbs (1984), is a single moment scheme including some modifications, as saturation adjustment following Tao et al. (1989) and ice sedimentation, which is related to the sedimentation of small ice crystal (Mitchell et al., 2008). It includes six classes of hydrometeors: water vapour, cloud water, rain, cloud ice, snow, and graupel (Baró el al. 2015). WRF-Chem model allows transforming the single into a double moment scheme of the Lin microphysic scheme. This implementation is described in Chapman et al. (2009). Following Ghan et al. (1997), a prognostic treatment of cloud droplet number was added, 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 and follows 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 (Baró et al. 2015).

Finally, the effect of aerosols on incoming solar radiation within WRF-Chem is determined by transferring relevant parameters to the shortwave radiation scheme, representing radiative feedbacks due to aerosol-radiation interactions. The interactions of clouds and incoming solar radiation have been implemented by linking simulated cloud droplet number with the shortwave radiation scheme and with Lin microphysics (Skamarock et al., 2005). Therefore, droplet number will affect both the calculated droplet mean radius and cloud optical depth when using shortwave radiation scheme, representing radiative feedbacks due to aerosol-clouds interactions."

References have been included in the revised version:

Barnard, J. C., Fast, J. D., Paredes-Miranda, G., Arnott, W., and Laskin, A.: Technical note: evaluation of the WRF-Chem" aerosol chemical to aerosol optical properties" module using data from the MILAGRO campaign. Atmos. Chem. Phys., 10 (15), 7325-7340, 2010.

Baró, R., Jiménez-Guerrero, P., Balzarini, A., Curci, G., Forkel, R., Grell, G., Hirtl, M., Honzak, L., Langer, M., Pérez, J.L., Pirovano, G., San José, R., Tuccella, P., Werhahn, J. and Žabkar, R.: Sensitivity analysis of the microphysics scheme in WRF-Chem contributions to AQMEII phase 2. Atmos. Environ., 115, 620-629, 2015.

Chapman, E. G., Gustafson Jr., W. I., Easter, R. C., Barnard, J. C., Ghan, S. J., Pekour, M. S., and Fast, J. D.: Coupling aerosol-cloud-radiative processes in the WRF-Chem model: Investigating the radiative impact of elevated point sources, Atmos. Chem. Phys., 9, 945-964, doi:10.5194/acp-9-945-2009, 2009.
Fast, J. D., Gustafson, Jr., W. I., Easter, R. C., Zaveri, R. A., Barnard, J. C., Chapman, E. G., Grell, G. A., and Peckham, S. E.: Evolution of Ozone, Particulates and Aerosol Direct Radiative Forcing in the Vicinity of Houston Using a Fully Coupled Meteorology-Chemistry-Aerosol Model, J. Geophys. Res., 111, D21305, doi:10.1029/2005JD006721, 2006.
Forkel, R., Werhahn, J., Hansen, A. B., McKeen, S., Peckham, S., Grell, G., and Suppan, P.: Effect of aerosol-radiation feedback on regional air quality–a case study with WRF/Chem. Atmos. Environ., 53, 202-211, 2012.
Ghan, S. J., Leung, L. R., Easter, R. C. and Abdul-Razzak, H.: Prediction of Droplet Number in a General Circulation Model, J. Geophys. Res., 102, 21 777–21 794, 1997.
Liu, Y., Daum, P. H., and McGraw, R. L.: Size Truncation Effect, Threshold Behavior, and a New Type of Autoconversion Parameterization, Geophys. Res. Lett., 32, L11811, doi:10.1029/2005GL022636, 2005.
Lin, Y.-L., Farley, R. D., and Orville, H. D.: Bulk Parameterization of the Snow Field in a Cloud Model, J. Climate Appl. Meteor., 22, 1065–1092, 1983.
Mitchell, D.L., Rasch, P., Ivanova, D., McFarquhar, G., Nousiainen, T.: Impact of small ice crystal assumptions on ice sedimentation rates in cirrus clouds and GCM simulations. Geophys. Res. Lett., 35, 2008.
Rutledge, S. A. and Hobbs, P. V.: The Mesoscale and Microscale Structure and Organization of Clouds and Precipitation in Midlatitude Cyclones. XII: A Diagnostic Modeling Study of Precipitation Development in Narrow Cold-Frontal Rainbands, J. Atmos. Sci., 20, 2949-2972, 1984.
Skamarock, W. C., Klemp, J. B., Dudhia, J., Gill, D. O., Barker, D. M., Wang, W., and Powers, J. G.: A Description of the Advanced Research WRF Version 2. NCAR Technical Note, NCAR/TN-468+STR, 88 pp, National Center for Atmospheric Research, Boulder, Colorado, USA, 2005, available at: http: //wrf-model.org/wrfadmin/publications.php.
Tao, W.K., Simpson, J., McCumber, M.: An ice-water saturation adjustment. Mon. Weather Rev., 117, 231–235, 1989.

Rev. #1: Results and discussions are essentially describing the discrepancies between simulations results and observation without further discussion on the potential drivers.

A: For AOD, discrepancies between simulations and observations can be ascribed to errors in the model estimation of the aerosol dry mass, the fraction of particles for a given mass or the water associated with aerosols. On the other hand, the known errors from observations have to be considered (as indicated in the manuscript, page 8: "we should bear in mind that this fact may be conditioned by the MODIS underestimation of AOD_{550} *levels for high loads of this type of particles, which has been reported by Chu et al., 2002; Levy et al., 2005 and Remer et al., 2005, among others"). For AE, discrepancies can be ascribed to an underestimation on the variability of particles size.*

The aforementioned comment has been incorporated into the Conclusions section (pages 11-12).

Specific comments:

Rev. #1: Page 2, Line 09: "Light-absorbing aerosols such as biomass burning exert a warming influence. . ." That may be true for black carbon aerosol particles, however biomass burning aerosol plumes are not only composed by black carbon. Biomass burning plume as a whole may have a cooling effects (references example: Schafer et al., 2002, Observed reductions of total solar irradiance by biomass-burning aerosols in the Brazilian Amazon and Zambian Savanna, GRL, Volume: 29 Issue: 17; Procopio et al., 2004, Multiyear analysis of amazonian biomass burning smoke radiative forcing of climate, GRL, Volume: 31 Issue: 3).

A: The sentence has been rephrased in order to take into account the reviewer comment. E.g. "Light-absorbing aerosols such as black carbon, which are a component of biomass burning, exert a warming influence (e.g. Jacobson, 2001)."

Rev. #1: Page 3, Line 14: Please, include specifically which modelling output are you refering to.

A: Modelling outputs are aerosol optical properties (aerosol optical depth, AOD and Angstrom exponent, AE). This has been clarified in the revised manuscript.

Rev. #1: Page 5: Emissions sources are discussed here, however nothing is said about the dust emission, one of the aerosol type focus of the study.

A: WRF-Chem predicts online dust emission as a function of the land usage information and the simulated meteorological fields. In this work and following Shaw et al. (2008), dust emission flux (G) depends on: an empirical proportionality constant estimated based on regional specific data (C); the vegetation mask accounting for vegetation type (α); the friction velocity (u_*); the threshold friction velocity below which dust emission does not occur ($u_{*t} = 20\ cm\ s^{-1}$ following Shaw et al., 2008); and the soil wetness factor accounting for soil moisture (f_w).

$$G = \alpha C u_*^4 \left(1 - \frac{f_w u_{*t}}{u_*}\right)$$

This has been clarified in the revised manuscript.

Shaw, W., Allwine, K. J., Fritz, B. G., Rutz, F. C., Rishel, J. P., and Chapman, E. G.: An evaluation of the wind erosion module in DUSTRAN, Atmos. Environ., 42, 1907–1921, 2008.

Rev. #1: Page 5, Line 17: ". . . aerosol particles are represented by two lognormal size distributions, corresponding to an Aitken mode and an accumulation mode. . .": Considering that an event of Saharan dust outbreak is analysed, a coarse mode consideration wouldn't be relevant? The absence of a coarse mode aerosol in the model parametrization certainly helps to explain the discussed model difficulty to simulate Angstrom Exponent variability.

A: The model includes coarse aerosols in the model parameterization. This point has been clarified in the revised version of the manuscript. E.g. "Here aerosol particles are represented by three log-normal size distributions, corresponding to an Aitken mode (nucleation mode 0.1 μm diameter), and accumulation mode (0.1 – 2 μm), and a coarse mode (> 2 μm) (Forkel et al. 2012)."

Forkel, R., Werhahn, J., Hansen, A. B., McKeen, S., Peckham, S., Grell, G., & Suppan, P. (2012). Effect of aerosol-radiation feedback on regional air quality–a case study with WRF/Chem. Atmospheric environment, 53, 202-211.

Rev. #1: Page 6, Line 18: MODIS Angstrom Exponent is only available for ocean region? If yes, so the analysis was not restricted to Iberian Peninsula, but also over the surround sea and ocean.

A: As the reviewer indicates this area covers the Iberian Peninsula and the surrounding sea and ocean. For this reason, a better description of the study area has been done in the revised version of the manuscript.

Rev. #1: Page 8, Line 1- 5: Certainly MODIS retrievals have issues, but also it would be important to discuss the modelling issues that can contribute to the discrepancies.

A: Generally a too high predicted AOD by the model can be explained by either too much aerosol dry mass present in the model, too large fraction of small particles for a given mass, *or due to an excess of water associated with the aerosols (Chapman et al. 2009). This has been clarified in the revised manuscript (Page 8).*

Chapman, E. G., Gustafson Jr., W. I., Easter, R. C., Barnard, J. C., Ghan, S. J., Pekour, M. S., and Fast, J. D.: Coupling aerosol-cloud-radiative processes in the WRF-Chem model: Investigating the radiative impact of elevated point sources, Atmos. Chem. Phys., 9, 945-964, doi:10.5194/acp-9-945-2009, 2009.

Rev. #1: Page 8, Line 1 – 2: Is the correlation coefficient obtained from model simulation comparison with MODIS data distinct from that calculated for the comparison between model simulation against AERONET? If so, why is correlation coefficients for model x MODIS much higher than correlation coefficients for model x AERONET (Table 3)? How does MODIS AOD compare with AERONET stations AOD?

A: Correlation coefficients for model x AERONET are obtained from a comparison between a point (AERONET) and a cell (model outputs) covering the corresponding station coordinates following a nearest neighbour approach. In spite of the use of this approach, small errors on the spatial distribution of the model representation of the evaluated variables can appear, producing lower correlation coefficient values than the comparison with MODIS data, where the comparison is done cell (MODIS) vs. cell (model) with approximately the same resolutions. This comment has been introduced in the paper (Page 10)

Regarding the second question (MODIS AOD vs. AERONET AOD), the comparison between MODIS AOD and AERONET stations AOD has been done by using the revised protocol developed by Petrenko et al. (2012), where satellite and sun photometer are compared within a spatial radius of ±25 km and a temporal interval of ±30 min. A valid collocation is one where there are at least three MODIS pixels and two sun photometer measurements within the spatial/temporal window. For Collection 6 (C6), the correlation is R = 0.86, and that 69.4 % of MODIS AOD fall within expected uncertainty of ± (0.05 + 15 %) (Levy et al. 2013).

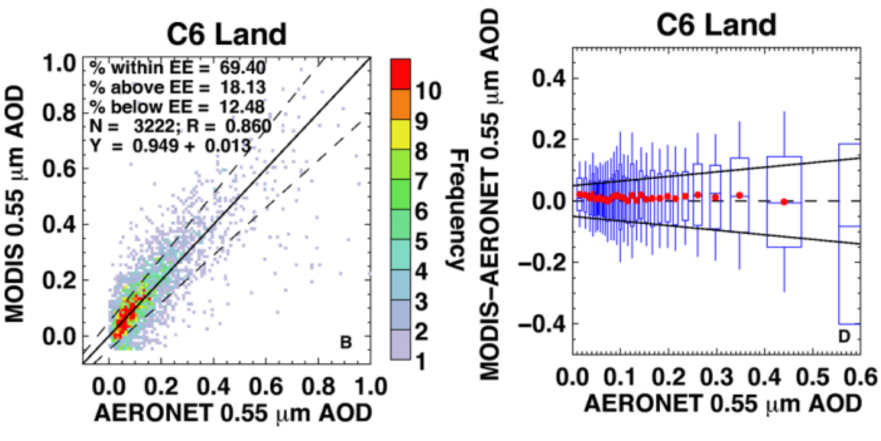


Figure 1. Left: frequency scatter plots for AOD at 0.55 μm over dark-land compared to AERONET, plotted from 6 months of Aqua (January and July; 2003, 2008 and 2010), computed with C6 algorithm (b). One-one lines and EE envelopes ± (0.05 + 15 %) are plotted as solid and dashed lines. Collocation statistics are presented in each panel. Right: the same information plotted as AOD error (MODIS-AERONET) versus AERONET, broken into equal number bins of AERONET AOD (d). One-one line (zero error) is dashed and EE envelopes are solid. For each box-whisker, its properties and what they represent include: width is 1-σ of the AOD bin, whereas height, whiskers, middle line and red dots are the 1-σ, 2-σ, mean and median of the AOD error, respectively

Petrenko, M., Ichoku, C., and Leptoukh, G.: Multi-sensor Aerosol Products Sampling System (MAPSS), Atmos. Meas. Tech., 5, 913–926, doi:10.5194/amt-5-913-2012, 2012.

Levy, R. C., Mattoo, S., Munchak, L. A., Remer, L. A., Sayer, A. M., Patadia, F., and Hsu, N. C.: The Collection 6 MODIS aerosol products over land and ocean, Atmos. Meas. Tech., 6, 2989-3034, doi:10.5194/amt-6-2989-2013, 2013.

Rev. #1: Page 10, Line 4 – 5: The inclusion of more days in the analysis may provide a better analysis from the statistical perspective.

A: The reviewer is right in his/her appreciation; however, bearing in mind the high computational costs and the framework of this work (related to EuMetChem Cost Action ES1004), just two episodes for the year 2010 have been included. In EuMetChem, the objective is to evaluate two important aerosol episodes differing in the type of aerosol (biomass burning vs. dust). The inclusion of more days in the analysis would imply a high computational cost and would not represent such extreme events as the ones considered in this work.

Rev. #1: Page 18, Table 1: A map of the distribution of the AERONET sites and the EARLINET station in Granada would be helpful to the readers to follow the discussions. For example, that can be done in one of the AOD field map from the simulation.

A: Table 1 with stations coordinates has been changed by a map of the distribution of AERONET and EARLINET stations in the revised version of the manuscript.

Technical corrections:

Rev. #1: Although the comprehension of the manuscript is not affected, I would recommend that the authors make use of an editing service, so that the writing can be improved. There are many sentences that need improvements; here I list some of them:

A: Please find below the list of recommendations of the Reviewer and the corrections made.

Page 1, Abstract first line: ". . .over the Earth's climate..." to ". . . on the Earth's climate. . ." *(Done)*
Page 2, Line 3: ". . .cause changes are: (1) scattering and absorption of solar radiation. . ." to ". . .cause changes are: (1) scattering and absorbing solar and terrestrial radiation. . ." Dust aerosol in particular may affect terrestrial radiation. *(Done)*
Page 2, Line 14: The sentence "The large uncertainty quantifying these . . ." read better as "The uncertainty quantification of these aerosol effects on the Earth radiative budget is much higher. . ." *(Done)*
Page 3, Line 22: ". . .altering the global budget indirectly. . ." to ". . .altering the global energy budget indirectly. . ." *(Done)*
Page 3, Line 27: "The grid size is 6000 cells. . ." to "The grid size consists of 6000 cells. . ." *(Done)*
Page 7, Line 18: "We can then state then that the changes. . ." to "We can then state that the changes. . ." Page 9, Line 16: replace ". . .10 (a) & (c). . ." to ". . .10 (a) and (c). . ." *(Done)*
Page 9, Line 21: " Sagres stations . . ." to "Sagres station. . ." *(Done)*
Page 10, Line 3: "Several specific days . . ." to "Two specific day. . ." *(Done)*
Page 19, Table 1 and 2: Part of the table at the right side is missing. *(Revised)*
Recommendation for the figures legends: Include the period over which mean field AOD and AE are calculated and avoid abbreviations such as S.L (significant level) *(Revised)*

Anonymous Referee #2.

Rev. #2: The objective of this paper is to quantify the aerosol radiative feedback for the Iberian Peninsula for some pollution episodes. [...] The subject of this study is relevant for publication in ACP. It is crucial to accurately estimate feedback of aerosols from different sources to radiation budget over the region.

A: As for the anonymous referee #1, we would like to thank to anonymous referee #2 for their valuable comments in the interactive comment on "Assessment of the radiative effects of aerosols in an on-line coupled model over the Iberian Peninsula" by Laura Palacios-Peña et al. (No. acp-2016-473). The manuscript has been revised after reviewer's comments in order to correct errors and to introduce the reviewer' suggestions for improving the quality of the paper. Please see below our point-by-point replies:

Rev. #2: The section 2.1 provides limited information about the WRF-Chem model setup used in the study. Which gas chemistry, microphysics etc. options were used in the model?

A: This point has been clarified in the revised version of the manuscript. "The following physics options were applied for both simulations, including (or not) aerosol radiative feedbacks: Rapid Radiative Transfer Method for Global (RRTMG) longwave and shortwave radiation scheme; the Yonsei University (YSU) PBL scheme, the NOAH land-surface model, the Lin microphysics scheme and the updated version of the Grell-Devenyi scheme with radiative feedbacks. Further description of the physics can be found in Grell et al. (2005). According to chemistry options, the followings were applied: MADE/SORGAM aerosol scheme; the RADM2 gas phase mechanism and the Fast-J photolysis scheme."

This description has been introduced in the revised version of the manuscript.

Rev. #2: Why did the authors choose the SORGAM module? It's well known that the SORGAM drastically underestimates secondary organic aerosol (SOA) concentrations, consequently total aerosol concentrations. There are versions of the MADE aerosol scheme coupled to new SOA schemes in WRF-Chem (e.g. Tuccella et al., 2015).

A: As the review indicates the SORGAM module underestimates simulated PM2.5 mass, mainly attributable to SOA (Grell at al., 2005; McKeen et al., 2007 and Tuccella et al., 2012). As reported by Tuccella et al., (2012), one of the most probable reasons for OM underestimation is that the RADM2 chemical mechanism (also used in this work) does not include the oxidation of biogenic monoterpenes and has a limited treatment of anthropogenic VOC oxidation (McKeen et al., 2007).

We agree with the reviewer that the election of SORGAM may bring underestimation in SOA levels. However, it is really hard to establish the cause of SORGAM's underestimation, especially because the AOD levels are overestimated during the biomass burning episode.

The aforementioned authors also point to other causes of PM negative bias. Finally, another potential source of the PM2.5 bias is the simulation of the meteorological fields, as temperature or wind speed. It could be linked to unspeciated PM2.5 due to underestimation of its emissions. So, the a priori selection of a SOA mechanism is hard to establish.

Moreover, the election of SORGAM comes conditioned by the participation of our group in EuMetChem Cost Action ES1004 and AQMEII initiative. Our configuration, which uses the MADE/SORGAM aerosols and the RADM2 gas-phase mechanisms, was established within this Cost Action, where other groups used different configurations of SOA (e.g. VBS) so we may have information about the sensitivity of the WRF-Chem model to the election of several *physico-chemical options (such as the election of the SOA mechanism). That's the main cause for the election of the SORGAM module.*

Grell, G. A., Peckham, S. E., Schmitz, R., McKeen, S. A., Frost, G., Skamarock, W. C., and Eder, B.: Fully coupled "online" chemistry within the WRF model, Atmos. Environ., 39, 6957-6975, 2005.

McKeen, S., Chung, S. H., Wilczak, J., Grell, G., Djalalova, I., Peckham, S., Gong, W., Bouchet, V., Moffet, R., Tang, Y., Carmichael, G. R., Mathur, R., and Yu, S.: Evaluation of several PM2.5 forecast models using data collected during the ICARTT/NEAQS 2004 field study, J. Geophys. Res., 112, D10S20, doi: 10.1029/2006JD007608., 2007.

Tuccella, P., Curci, G., Visconti, G., Bessagnet, B., Menut, L., and Park, R. J.: Modeling of gas and aerosol with WRF/Chem over Europe: Evaluation and sensitivity study, J. Geophys. Res., 117, D03303, doi: 10.1029/2011JD016302, 2012.

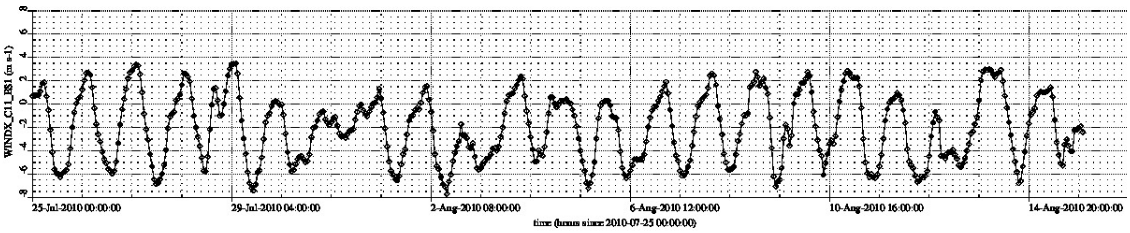
Tuccella, P., G. Curci, G. A. Grell, G. Visconti, S. Crumeyrolle, A. Schwarzenboeck and A. A. Mensah.: A new chemistry option in WRF-Chem v. 3.4 for the simulation of direct and indirect aerosol effects using VBS: evaluation against IMPACT-EUCAARI data, Geosci. Model Dev., 8(9), 2749-2776, 2015.

Rev. #2: The authors need to provide more details on how the aerosol-radiation and aerosol-cloud interactions are parameterized in their version of WRF-Chem. These details could help to better interpret the model-observation discrepancies.

A: This fact is also highlighted by the Anonymous Referee #1, so we refer to the answer above where a detailed description of the aerosol, microphysical and optical modules as well as the previous description of the representation of aerosol-radiation-clouds interactions has been done. This has been included in the revised version of the manuscript.

Rev. #2: The model was run on 23km resolution. This is a relatively coarse model grid. It doesn't allow simulating land-sea breeze and other mesoscale circulations. Moreover, in such resolution there are more parameterized (by cumulus parameterization) clouds in the model. Since the model doesn't treat aerosol-cloud feedback in cumulus parameterization, the overall ACI effect can't be captured by these model settings.

A: In spite of the relatively coarse model grid, the model allows the representation of land-sea breezes. The next figure represents the time series of wind on a point in the east coast of the Iberian Peninsula. In this figure we can see the daily cycle due to the land-sea breeze.



WINDX_C11_BS1 from COST ES1004 Russian forest fire case, no feedbacks

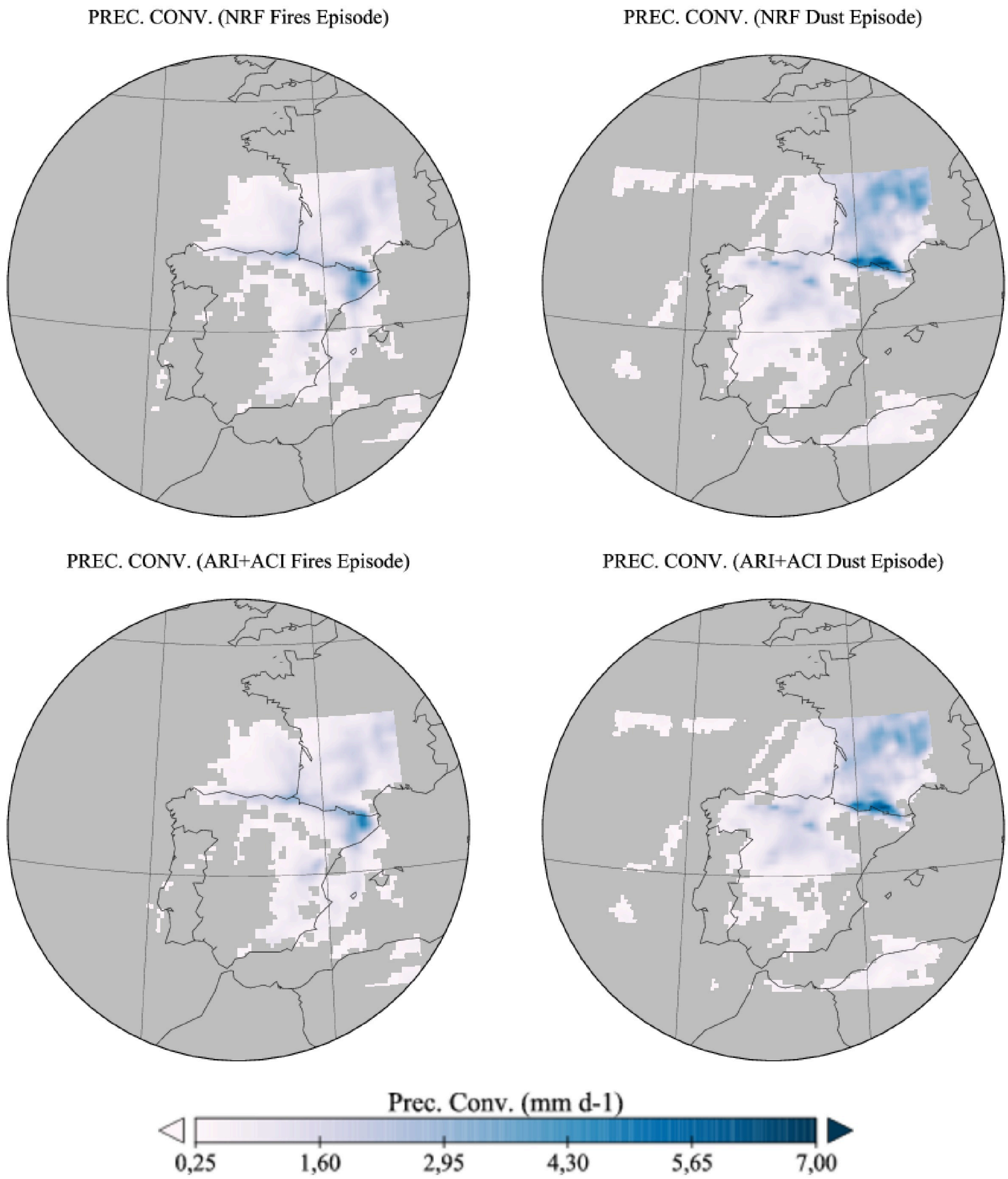
According to the treatment of aerosol-cloud feedbacks, as the reviewer indicated and as reported by Archer-Nicholls et al., (2016), WRF-Chem has a limitation to assess aerosol-cloud interactions because the couplings are not computed in convective clouds simulated by the cumulus parameterisation (Chapman et al., 2009; Yang et al., 2011). We are well aware that the limitation of the model but a WRF-Chem state of the art version has been implemented. In spite of this, thanks to the reviewer's comment, we have evaluated the cumulus presence in the episodes study. The next figure shows the mean accumulated convective precipitation as a representation of the cumulus presence. A threshold of 0.25 mm day^{-1} is considered, being values under this threshold negligible. The figure shows that the highest values, around 5 mm day^{-1}, are found over the north-east of the domain (over the Pyrenees mountains). So, we understand that during both episodes the cumulus presence is limited. Moreover and according to the AOD values, shown in the initial version of the manuscript, the area with the *highest values of convective precipitation is not strongly affected by the high aerosol loads study in this work.*

In spite of this, a comment about the limitation of the model due to the aerosol-cumulus interactions has been done in the revised version of the manuscript.

Archer-Nicholls, S., Lowe, D., Schultz, D. M., and McFiggans, G.: Aerosol–radiation–cloud interactions in a regional coupled model: the effects of convective parameterisation and resolution, Atmos. Chem. Phys., 16, 5573-5594, doi:10.5194/acp-16-5573-2016, 2016.

Chapman, E. G., Gustafson Jr., W. I., Easter, R. C., Barnard, J. C., Ghan, S. J., Pekour, M. S., and Fast, J. D.: Coupling aerosol-cloud-radiative processes in the WRF-Chem model: Investigating the radiative impact of elevated point sources, Atmos. Chem. Phys., 9, 945-964, doi:10.5194/acp-9-945-2009, 2009.

Yang, Q., W. I. Gustafson Jr., Fast, J. D., Wang, H., Easter, R. C., Morrison, H., Lee, Y.-N., Chapman, E. G., Spak, S. N., and Mena-Carrasco, M. A.: Assessing regional scale predictions of aerosols, marine stratocumulus, and their interactions during VOCALS-REx using WRF-Chem, Atmos. Chem. Phys., 11, 11951–11975, doi:10.5194/acp-11-11951-2011, 2011.



Rev. #2: Another uncertainty stems from using ECMWF analysis fields for the meteorological initial and boundary conditions in the regional WRF-Chem modeling. The ECMWF model assimilates met. observations, which might be already affected by those dust and fire aerosols. Hence, the base WRF-Chem model case implicitly may already include some of the aerosol feedback. I understand that it's hard to set up a "perfect" regional modeling framework to study the aerosol-meteorology interactions, however this issue needs to be mentioned in the paper.

A: The reviewer is right and this issue has been mentioned in the revised version of the manuscript. Page 7: "At this point, it should be mentioned that the use of ECMWF operational archive for meteorological initial and boundary conditions can produce that some of the aerosol feedback may already take into account in the base case (NRF) because of the model assimilation of meteorological observations of the ECMWF."

Rev. #2: I don't see much discussions of the simulated ACI effect in the paper. For clarity it'd better to show three model cases - w/o any aerosol feedback, with aerosol feedback on radiation and with aerosol feedback on radiation+clouds, and discuss them more thoroughly.

A: We appreciate the reviewer's suggestion. However, for the sake of brevity and in order not to increase the length of the manuscript (what would affect readability of the paper) we decided to show only the accumulated effects of NRF versus ARI+ACI cases.

Rev. #2: Another missing piece in this paper is lack of evaluations of the simulated aerosol concentrations. Thus, it's hard to interpret AOD comparisons given the lack of information about the model's skill to simulate aerosol mass concentrations in dust and smoke plumes.

A: An evaluation of the simulated aerosol concentrations is presented by Im et al., (2015), where PM simulations for the year 2010 in the context of AQMEII2 are evaluated. The simulations evaluated in the manuscript on revision present the same configuration of the ES1 simulation from Im et al., (2015) and therefore the evaluation results have been mentioned in the revised version of the manuscript citing the work of Im et al. (2015).

This point has been clarified in the revised version of the manuscript.

Im, U., Bianconi, R., Solazzo, E., Kioutsioukis, I., Badia, A., Balzarini, A., Baró, R., Bellasio, R., Brunner, D., Chemel, C., Curci, G., Denier van der Gon, H., Flemming, J., Forkel, R., Giordano, L., Jiménez-Guerrero, P., Hirtl, M., Hodzic, A., Honzak, L., Jorba, O., Knote, C., Makar, P.A., Manders-Groot, A., Neal, L., Pérez, J.L., Pirovano, G., Pouliot, G., San Jose, R., Savage, N., Schroder, W., Sokhi, R.S., Syrakov, D., Torian, A., Tuccella, P., Wang, K., Werhahn, J., Wolke, R., Zabkar, R., Zhang, Y., Zhang, J., Hogrefe,C. and Galmarini, S.: Evaluation of operational online-coupled regional air quality models over Europe and North America in the context of AQMEII phase2. Part II: particulate matter, Atmos. Environ., 115, 421–441, 2015.

Minor comments:

Rev. #2: Authors use many abbreviations in the text. I suggest adding a table showing all of them in one place.

A: A table showing all of abbreviations had been included as Apendix in the revised version of the manuscript.

Apendix. List of acronyms.

ACI	*Aerosol-cloud interactions*
AE	*Angström Exponent*

AERONET	*AErosol Robotic NETwork*
AOD	*Aerosol Optical Depth*
ARI	*Aerosol-radiation interactions*
BSCAT	*Backscatter*
DB	*Deep Blue*
DT	*Dark Target*
EARLINET	*European Aerosol Research Lidar Network*
ECMWF	*European Centre for Medium-Range Weather Forecasts*
EuMetChem	*European framework for online integrated air quality and meteorology modelling*
IFS-MOZART	*Integrated Forecasting System - Model for ozone and related tracers*
IP	*Iberian Peninsula*
IPCC	*Intergovernmental Panel on Climate Change*
IS4FIRES	*Integrated monitoring and modelling system for wild-land fires*
MACC-II	*Monitoring Atmospheric Composition and Climate-Interim Implementation*
MAE	*Mean Absolute Error*
MBE	*Mean Bias Error*
MEGAN	*Model of Emissions of Gases and Aerosols from Nature*
MODIS	*Moderate Resolution Imaging Spectroratiometer*
NRF	*No radiative feedbacks*
r	*Correlation Coefficient*
RF	*Radiative feedbacks*
RRTMG	*Rapid Radiative Transfer Method for Global*
S.L.	*Significance Level*
TNO	*Netherlands Organization for Applied Scientific Research*
YSU PBL	*Yonsei University Planetary Boundary scheme*
WRF-Chem	*Weather Research and Forecasting model coupled with Chemistry*

page 9: correct "values shows" *(Done)*
page 11: correct "fires particles" *(Done)*
References section: The paper by Iacono et al. is entered twice. *(Revised and corrected)*

Evaluating the representation of aerosol optical properties by an on-line coupled model over the Iberian PeninsulaAssessment of the radiative effects of aerosols in an on-line coupled model over the Iberian Peninsula

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Abstract. The effects of atmospheric aerosol particles on the Earth's climate over the Earth's climate mainly depend on their optical, microphysical and chemical properties, which modify the Earth radiative budget. The aerosol radiative effects can be divided into direct and semi-direct effects, produced by the aerosol-radiation interactions (ARI); and indirect effects, produced by aerosol-cloud interactions (ACI). In this sense the objective of this work is to assess whether the inclusion of aerosol radiative feedbacks in the on-line coupled WRF-Chem model improves the modelling outputs over the Iberian Peninsula (IP) and surrounding water areas. For that purpose, the methodology bases on the evaluation of modelled aerosol optical properties under different simulation scenarios. The evaluated data come from two WRF-Chem simulations for the IP differing in the inclusion/no-inclusion of ARI and ACI (NRF/RF simulations). The case studies cover two episodes with different aerosol types over the IP in 2010, namely a Saharan desert dust outbreak and a forest fire episode. The evaluation uses observational data from AERONET stations and MODIS sensor, including aerosol optical depth (AOD) and Angström exponent (AE). Experimental data of aerosol vertical distribution from the EARLINET Granada station are used for checking the models. The results indicate that for the spatial distribution the best-represented variable is AOD and the largest improvements of including the aerosol radiative feedbacks are found for the vertical distribution. In the case of the dust outbreak, a slight improvement(worsening) is produced over the areas with medium(high/low) levels of AOD (-9%/+12% of improvement) when including the aerosol radiative feedbacks. For the wildfires episode, improvements of AOD representation (up to 11%) over areas further away from emission sources are estimated, which compensates the computational effort of including aerosol feedbacks in the simulations. No evident improvement is observed for the AE representation, whose variability is largely underpredicted by both simulations.

1 Introduction

It is nowadays widely recognised that aerosol particles exert a substantial influence on Earth's climate changing the radiative budget (Charlson et al., 1992; Hansen et al., 1997; Ramanathan and Feng, 2009; Boucher et al., 2013, IPCC, 2013, among many others). The principal mechanisms by which aerosols cause these changes are: (1) scattering and absorption ~~of~~ solar and terrestrial radiation (aerosol-radiation interactions, ARI) (e.g. Ruckstuhl et al., 2008) and: (2) modification of clouds and precipitation, thereby affecting both radiation and hydrology, or increasing the reflectivity of clouds (aerosol-cloud interactions, ACI) (e.g. Twomey, 1974; Albrecht, 1989; Twomey, 1991). In the first case, light scattering by aerosol particles such as sea salt or desert dust increases the solar radiation reflected by the planet, producing a cooling influence. Light-absorbing aerosols such as ~~-~~black carbon, which are a component of biomass burning, exert a warming influence ~~biomass burning exert a warming influence~~ (e.g. Jacobson, 2001). These radiative influences are quantified as forcings (in W m^{-2}), defined as the perturbation to the energy balance of the Atmosphere-Earth system. A warming influence is denoted a positive forcing, and a cooling influence, negative (IPCC, 2013). Generally, modelling tools and observations indicate that anthropogenic aerosols have had a cooling influence on Earth since preindustrial time, with a total ARI+ACI medium-confidence radiative forcing (excluding the effect of absorbing aerosol on snow and ice) of -0.9 (-1.9 to -0.1) W m^{-2} (Boucher et al., 2013). The uncertainty quantification of these aerosol effects on the Earth radiative budget is much higher ~~The large uncertainty quantifying these aerosol effects on the Earth radiative budget are much higher~~ than for any other climate-forcing agent (IPCC, 2013). This happens because the physical, chemical and optical aerosol properties are highly variable in space and time scales due to the aerosol particles short-lived and non-uniform emissions (Forster et al., 2007).

In order to reduce this uncertainty, the use of models is one of the most powerful tools to understand the different processes affecting the climate system. As aerosol may strongly drive the Earth's climate on global and regional scales, fully-coupled meteorology-climate and chemistry models allow for accounting the climate-chemistry-aerosol-cloud-radiation feedbacks mechanisms between simulated aerosol concentrations and meteorological variables. It is also a promising way to go for future atmospheric simulation systems, leading to a new generation of models for improved meteorological, environmental and chemical weather forecasting (Baklanov et al., 2014).

Europe may be one of the most climatically sensitive world regions (Giorgi, 2006). Within the target domain, the role of aerosol particles may then be even more crucial over such regions as the Mediterranean basin, a crossroad that fuels the mixing of particles from different sources (Papadimas et al., 2012). The Iberian Peninsula (IP), as a good example within the Mediterranean basin, can be affected by high aerosols concentration of different aerosol types. Due to its closeness to the Sahara Desert, the IP is frequently affected by dust outbreaks with large aerosol loads that modulate the aerosol climatology in different areas of this region, especially in Southern Spain (e.g. Toledano et al., 2007; Guerrero-Rascado et al., 2008, 2009; Córdoba-Jabonero et al., 2011; Antón et al., 2012; Pereira et al., 2014) and Portugal (e.g. Wagner et al., 2009; Preißler et al., 2011). On the other hand, the Mediterranean climate, with high summer temperatures and dry soil-air conditions, encourage forest fires episodes over this region (Alados-Arboledas et al., 2010). Both types of emissions give major contributions to particle concentration in the atmosphere, particularly in the warmer season (Elias et al., 2006).

There is a large number of studies assessing the aerosol feedbacks effects over the IP using different remote sensing measurement methods, using devices such as sun photometers (Lyamani et al., 2005, 2006; Toledano et al., 2007; Cachorro et al. 2008; Obregón el at., 2012), nephelometers (Pereira et al., 2008, 2011), lidars (Guerrero-Rascado et al., 2008) or a combination of these (Elias et al., 2006; Córdoba-Jabonero et al., 2011). Other studies using these instruments join satellite measurements to carry out this assessment (Cachorro et al., 2006; Guerrero-Rascado et al., 2009). Even, there are studies that use these different measurements to estimate by a radiative transfer model the aerosol radiative forcing over some regions (Santos et al., 2008; Guerrero-Rascado et al., 2009; Valenzuela et al., 2012) or over the whole IP (Mateos et al. 2014). On the other hand, a number of studies (e.g. Myhre et al., 2007; Myhre et al., 2009) have tried to assess the aerosol feedbacks effects on a global scale, while other works (e.g. Péré et al., 2010; Meij et al., 2012; Curci et al., 2014, among others) have a more regional approach. However, no modelling studies of the aerosol radiative effects have ever been carried out for the IP.

According to Randall et al. (2007), the responses of the climate system to aerosols and their effects on the radiative budget of the Earth are the most uncertain climate feedbacks.

Therefore, the objectives of this work are (i) to assess whether the inclusion of aerosol radiative feedbacks in the on-line coupled WRF-Chem model improves the modelling outputs of aerosol optical properties (aerosol optical depth, AOD and Angstrom exponent, AE) over the IP and surrounding water areas (seas and ocean) and (ii) to evaluate the representation of aerosol optical properties by this model over the target domain.

2 Methodology

In this paper we evaluate the AOD and AE output of different simulations carried out by the WRF-Chem model (Grell et al., 2005) by using observational data provided by several instruments: two ground-based data networks (AERONET and EARLINET) and a sensor on-board a satellite (MODIS). The results of the evaluation of the simulations presented here concerning particulate matter concentrations can be found in Im et al. (2015). Therefore, in this contribution we will focus on the evaluation of aerosol properties. Two different setups of the model have been considered, including/not including aerosol radiative feedbacks in the simulation. According to Boucher et al. (2013), the inclusion of these feedbacks involves a change on the internal energy flows to the Earth system, affecting cloud cover or other components of the climate system such as aerosol particles, and, thereby, altering the global energy budget indirectly.

The evaluation has been performed by using classical statistics according to Willmott et al. (1985), Weil et al. (1992) and Willmott and Matsuura (2005). The individual model-prediction error or bias (e_i), the mean bias error (MBE), mean absolute error (MAE) and the correlation coefficient (r) have been calculated. All data needs have pre-processed and bilinearly interpolated to a common working grid. This has a resolution of 0.~~1º~~ 2º and covers between 35º and 47º north and -15 and 5º east. The grid size ~~is~~ consist of 6000 cells and the grid type is a regular lon-lat grid. After the interpolation, modelled data are evaluated against MODIS. The data to compare with AERONET and EARLINET are extracted from the model cell covering the corresponding station coordinates (Table Fig. 1) following a nearest neighbour approach.

First, in order to evaluate whether the inclusion of aerosol radiative feedbacks in the on-line coupled WRF-Chem model produces significant changes on the studied variables (or changes are just mere signal noise), a surrogate variable, associated to the significance level of the changes modelled (S.L.), is defined (Eq.1). Therefore, high values of S.L. indicate whether the changes between simulations including (and not including) aerosol radiative feedbacks are noticeable with respect to the variability of the signal or not, and therefore, their significance:

$$S.L. = \frac{\frac{1}{n}\sum_{i=1}^{n}\left|x_{i_{NRF}} - x_{i_{RF}}\right|}{S_{NRF}^{2}}\, x100 \,, \qquad (1)$$

where S.L. is the significance level, x_i is the value of the studied variable and S_{NRF}^2 is the associated variance for the case not taking into account any aerosol radiative feedbacks (NRF). Moreover, NRF represents the base case and RF is the aerosol radiative feedbacks simulation that includes the ARI+ACI.

Second, to evaluate whether the inclusion of the aerosol radiative feedbacks in the simulations leads to an improvement of the error of the model, the variable Improvement of the MAE is used (Eq. 2):

$$Improvement\ of\ MAE = \frac{1}{n}\sum_{i=1}^{n}|e_i|_{NRF} - \frac{1}{n}\sum_{i=1}^{n}|e_i|_{RF} \,, \qquad (2)$$

where $|e_i|$ is the absolute error of the simulations.

Finally, to estimate whether the inclusion of the aerosol radiative feedbacks in the simulations produces an improvement of the vertical distribution of aerosols, the normalized improvement of MAE has been calculated (Eq. 3):

$$Nor.Improv.of\ MAE\ (\%) = \frac{1}{n}\left(\frac{\sum_{i=1}^{n}|e_i|_{NRF} - \sum_{i=1}^{n}|e_i|_{RF}}{\sum_{i=1}^{n}|e_i|_{NRF}}\right) x100 \,, \qquad (3)$$

2.1 Modelling data: WRF-Chem

The evaluated data comes from regional air quality-climate simulations performed using the WRF-Chem online-coupled meteorology and chemistry model (Grell et al., 2005), version 3.4.1, under the umbrella of the EuMetChem Cost Action ES1004. A detailed description of the simulations can be found in Forkel et al. (2015). A brief description of the modelling methodology taken from the aforementioned work is described below.

The following physics options were applied for both simulations, including (or not) aerosol radiative feedbacks: Rapid Radiative Transfer Method for Global (RRTMG) longwave and shortwave radiation scheme; the Yonsei University (YSU) PBL scheme, the NOAH land-surface model, the Lin microphysics scheme and the updated version of the Grell-Devenyi scheme with radiative feedbacks. Further description of the physics can be found in Grell et al. (2005). According to chemistry options, the followings were applied: the Modal Aerosol Dynamics Model for Europe with the Secondary Organic Aerosol Model (MADE/SORGAM) aerosol scheme; the Regional Acid Deposition Model version 2 (RADM2) gas phase mechanism and the Fast-J photolysis scheme.The following physics options were applied for both simulations, including (or not) radiative feedbacks: Rapid Radiative Transfer Method for Global (RRTMG) longwave and shortwave radiation scheme; the Yonsei University (YSU) PBL scheme, the NOAH land-surface model and the updated version of the Grell-Devenyi scheme with radiative feedbacks. Further description of the physics can be found in Grell et al. (2005).

For all simulations discussed in this paper the native modelling grid spacing is 23 km (270 by 225 grid cells, Lambert Conformal Conic projection with center at 50N and 12E). The modelling domain covers Europe and a portion of Northern Africa and as well as large areas affected by the Russian forest fires. However, because of the scope of the paper is the IP, only data for a domain covering the IP and surrounding seas and ocean -has been used (Fig. 21). In the vertical direction, the atmosphere up 50 hPa is resolved into 33 layers with a higher resolution close to the surface.

Initial and boundary conditions for the meteorological variables were obtained from 3-hourly data with 0.25° resolution (analysis at 00 and 12 UTC and respective forecasts 3/6/9 hours) from the ECMWF operational archive. 3-hourly chemistry boundary conditions for the main trace gases and particulate matter concentrations were available from the ECMWF IFS-MOZART model run from the MACC-II project (Monitoring Atmospheric Composition and Climate-Interim Implementation, Inness et al., 2013) 1.125° spatial resolution.

Anthropogenic emissions for the EU domain provided by the TNO (Netherlands Organization for Applied Scientific Research) from a recent update of the TNO MACC emissions inventory (http://www.gmes-atmosphere.eu/; Pouliot et al., 2012, 2014; Kuenen et al., 2014) were applied.

Biomass burning emission data have been calculated from global fire emission data that have been supplied from the integrated monitoring and modelling system for wild-land fires (IS4FIRES) project (Sofiev et al., 2009) with 0.1º x 0.1º spatial resolution. Day and night vertical injection profiles were also provided. WRF-Chem emission species have been calculated by speciation following Andreae and Merlet (2001) and Wiedinmyer et al. (2011). However, no heat release due to the fires was taken into account.

Biogenic emissions are based on the Model of Emissions of Gases and Aerosols from Nature (MEGAN) model (Guenther et al. 2006). MEGAN is on-line coupled with WRF-Chem and makes use of simulated temperature and solar radiation. Moreover,

WRF-Chem predicts online dust emission as a function of the land usage information and the simulated meteorological fields. In this work and following Shaw et al. (2008), dust emission flux (G) depends on: an empirical proportionality constant estimated based on regional specific data (C); the vegetation mask accounting for vegetation type (α); the friction velocity (u_*); the threshold friction velocity below which dust emission does not occur (u_{*t} = 20 cm s^{-1}) following Shaw et al., 2008); and the soil wetness factor accounting for soil moisture (f_w).

$$G = \alpha C u_*^4 \left(1 - \frac{f_w u_{*t}}{u_*}\right) \quad (4)$$

WRF-Chem predicts dust emission as a function of the wind and is estimated online from the meteorological parameters

The most important feature to bear in mind for this work is the aerosol module. This aerosol module is based on the modal aerosol MADE (Modal Aerosol Dynamics Model) (Ackermann et al., 1998) which is a modification of the Regional Particulate Model (Binkowski and Shankar, 1995). Here aerosol particles are represented by three log-normal size distributions, corresponding to an Aitken mode (nucleation mode 0.1 µm diameter), and accumulation mode (0.1 – 2 µm), and a coarse mode (> 2 µm) (Forkel et al. 2012).~~Here aerosol particles are represented by two lognormal size distributions, corresponding to an Aitken mode and an accumulation mode. They both describe submicrometer-diameter particles and micrometer particles~~. SOA have been incorporated into MADE in the SORGAM (Secondary Organic Aerosol Model) module (Schell et al., 2001).

Aerosol chemical properties and sizes are used to determine aerosol optical properties as a function of wavelength using the method outlined in Fast et al. (2006) and Barnard et al. (2010). In brief, each chemical constituent of the aerosol is associated with a complex index of refraction. The overall refractive index for a given size bin is determined by volume averaging, with Mie theory and summation over all size bins used to determine composite aerosol optical properties. Wet particle diameters are used in the calculations (Chapman et al. 2009).

The microphysical module consist of the Lin scheme based on Lin et al. (1983) and Rutledge and Hobbs (1984), is a single moment scheme including some modifications, as saturation adjustment following Tao et al. (1989) and ice sedimentation, which is related to the sedimentation of small ice crystal (Mitchell et al., 2008). It includes six classes of hydrometeors: water vapour, cloud water, rain, cloud ice, snow, and graupel (Baró el al. 2015). WRF-Chem model allows to transform the single into a double moment scheme of the Lin microphysic scheme. This implementation is described in Chapman et al. (2009). Following Ghan et al. (1997), a prognostic treatment of cloud droplet number was added, 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 and follows 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 (Baró et al. 2015).

Finally, the effect of aerosols on incoming solar radiation within WRF-Chem is determined by transferring relevant parameters to the shortwave radiation scheme, representing radiative feedbacks due to aerosol-radiation interactions. The interactions of clouds and incoming solar radiation have been implemented by linking simulated cloud droplet number with the shortwave radiation scheme and with Lin microphysics (Skamarock et al., 2005). Therefore, droplet number will affect both the calculated droplet mean radius and cloud optical depth when using shortwave radiation scheme, representing radiative feedbacks due to aerosol-clouds interactions. A limitation of WRF-Chem in the treatment of aerosol-cloud interactions is that this couplings are not computed in convective clouds simulated by the cumulus parameterisation (Chapman et al., 2009; Yang et al., 2011; Archer-Nicholls et al., 2016).

Although the modelling domain covers all Europe, for the purpose of this work data from the IP and surrounding areas with a resolution of 0.2° has been extracted for two important aerosol episodes in 2010. One of these episodes consists of a Saharan desert dust outbreak (from 28 June to 12 July) and a forest fires episode (from 25 July to 7 August). These episodes are selected because they represent two situations with a high load of atmospherics aerosol particles, when the radiative budget can be strongly affected. No volcanic emissions were considered in spite of the Eyjafjallajökull eruption in spring 2010. However, the volcanic plume reached all the IP only in May 2010 (Sicard et al., 2012; Navas-Guzmán et al., 2013), which is out of the scope of these case studies.

The simulations are run for two different configurations differing in the inclusion/no-inclusion of aerosol radiative feedbacks (ARI+ACI). The base case or NRF simulation, does not take into account any aerosol feedbacks and the RF simulation adds the ARI and ACI to the previous modelling setup. At this point, it should be mentioned that the use of ECMWF operational archive for meteorological initial and boundary conditions can produce that some of the aerosol feedback may already take into account in the base case (NRF) because of the model assimilation of meteorological observations of the ECMWF.

2.2 Observational data

2.2.1 Moderate Resolution Image Spectrometer (MODIS)

The satellite data chosen to evaluate the WRF-Chem simulations comes from MODIS (Levy et al., 2005) Level-2 Atmospheric Aerosol Product (MXD04_L2), collection or version 6 (C6) (Levy et al., 2013). The MODIS Aerosol Products monitor the ambient aerosol optical thickness over the oceans globally and over a portion of the continents. Daily Level 2 data have a spatial resolution of a 10x10 km. Two MODIS Aerosol data product files have been selected: MOD04_L2, containing data collected from the Terra platform; and MYD04_L2, containing data collected from the Aqua platform. In this case, the MXD04_L2 provides full global coverage of aerosol properties from the Dark Target (DT) aerosol retrieval algorithm, which is applied over ocean and dark land (e.g., vegetation) (Levy et al., 2013).

The variables used from MODIS are Aerosol Optical Depth (AOD) and Angström Exponent (AE).

AOD corresponds with AOD at a wavelength of 550 nm (AOD_{550}) for both ocean (best) and land (corrected) with best quality data (Quality Assurance Confidence = 3). The valid range of data is -0.05 to 5.0; that means a permission of small negative AOD values in order to avoid an arbitrary negative bias at the low AOD_{550} end in long-term statistics. This is because MODIS does not have sensitivity over land to retrieve aerosol to better than ±0.05 + 15 % under very clean conditions. Negative values of AOD_{550} have been considered as zero in this study. Over ocean the estimated error is -0.02 - 10%, +0.04 + 10% (Levy et al., 2013).

AE stands for AE for wavelengths between 550 and 860 nm ($AE_{550/860}$) over the ocean. The valid range for this variable is -1.0 to 5.0. In Collection 6, the preliminary estimated error for $AE_{550/860}$ is 0.45; pixels with an AOD_{550} > 0.2 are expected to have a more accurate $AE_{550/860}$ representation (Levy et al., 2013).

2.2.2 Aerosol Robotic Network (AERONET)

The Aerosol Robotic Network (AERONET) collaboration (Holben et al., 1998) provides globally distributed observations of spectral AOD, inversion products, and precipitable water in diverse aerosol regimes. The highest quality data can be found in Version 2, Level 2.0 (cloud-screened and quality-assured) data products.

The data used from AERONET in this work comes from level 2.0 of AOD at different wavelengths (AOD_{440}, AOD_{675}, AOD_{870} and AOD_{1020}) and AE ($AE_{440/870}$) from stations covering the IP available for the episodes studied (~~Table~~ Fig. 1). Typically the total uncertainty for AOD data under cloud-free conditions is <±0.01 for λ>440 nm and <±0.02 for shorter wavelengths (Holben et al., 1998).

2.2.3 European Aerosol Research lidar Network (EARLINET)

EARLINET (Pappalardo et al., 2014) is the first aerosol lidar network, established in 2000, with the main goal to provide a comprehensive, quantitative, and statistically significant database for the aerosol distribution on a continental scale. EARLINET data include particle backscatter and extinction coefficient profiles at 355, 532 and 1064 nm. EARLINET data used include backscatter profiles (BSCAT) at 355 and 532 nm (for the dates and times selected, no information is available at 1064 nm). The only station with available data for the studies cases in the IP during the year 2010 is Granada, and therefore is the only station included in this study.

3 Results and discussion

3.1 Significance level of simulated changes

First, to assess the effect of the inclusion of aerosol radiative feedbacks in the on-line coupled WRF-Chem model on the studied variables, the significance study described in Section 2 has been carried out. During the dust episode (Fig. 2~~1~~), the inclusion of aerosol radiative feedbacks produces differences with a significance level (defined as the ratio for the NRF-RF differences and the associated variance for the case not taking into account any aerosol radiative feedbacks) for AOD_{550} higher than 60% over the south-western IP. The rest of the domain presents S.L. ratios > 100 % in spite of the high AOD_{550} variance values (above 0.05). In the case of $AE_{440/870}$, the entire domain shows significance levels higher than 100%.

The inclusion of aerosol radiative feedbacks during the simulated fire episode (Fig. ~~2~~3) produces differences with a S.L. > 100%) for AOD_{550} over most of the domain. Over the area of fire particles emissions, S.L. ranges between 50 and 100% due to the higher absolute changes (> 0.2) than variance values (> 0.05). Similarly, for the dust episode the $AE_{440/870}$ over the entire domain shows significance levels > 100%.

Hence, over most of the domain the changes or differences due to the inclusion of aerosol radiative feedbacks have a high S.L., usually higher than 100%, and therefore the changes modelled are significant with respect to the variability of the studied variables. We can then state ~~then~~ that the changes discussed below are caused by the inclusion of the aerosol radiative feedbacks in our simulations, and not to the mere signal noise.

3.2 Model output vs. Terra-MODIS data

The results of the comparison between model outputs with MODIS data from Terra platform are shown in Figs. ~~3~~4-~~6~~7. The results from Aqua Platform are similar to Terra, and are therefore not shown here (but included in the Supplementary Material). Fig. ~~3~~ 4 (a) shows the mean values of AOD_{550} from MODIS for the dust outbreak. In this episode, high levels (above 0.4) over the south and the east of the domain are found, due to the shape of the dust outbreak. On the other hand, for the fires episode (Fig. ~~4~~ 5 (a)), the highest levels of MODIS AOD_{550} (> 0.25) are shown over the north of Portugal due to the presence of black carbon coming from wildfires, and over the south of the domain where a dust intrusion occurred at the end of this episode (> 0.3).

For AOD_{550} over the entire domain, the model outputs present low values of the MBE (represented by Figs. ~~3~~ 4 and ~~4~~5, (c) and (d)) for both NRF and RF simulations. During the dust episode the model underestimates MODIS AOD_{550} (MBE minimum values for NRF and RF simulations, respectively, -0.31 and -0.36) over the locations with important dust loads (high AOD_{550}) and overestimates (MBE maximum values 0.32 and 0.31) the low levels of AOD_{550}. Although the bias is generally lower during the fires episode, a peak of positive bias (0.47 for both simulations) is evaluated over the Portugal fire area, thus the model overestimates AOD_{550} for biomass burning particles for both model configurations, including or not aerosol radiative feedbacks. However, we should bear in mind that this fact may be conditioned by the MODIS underestimation of AOD_{550} levels for high loads of this type of particles, which has been reported by Chu et al., 2002; Levy et al., 2005 and Remer et al., 2005, among others. On the other hand, generally a too high predicted AOD by the model can be explained by either too much aerosol dry mass present in the model, too large fraction of small particles for a given mass, or due to an excess of water associated with the aerosols (Chapman et al. 2009). The estimation of the MAE (Table ~~2~~1) shows a slight increase for the RF simulation in both episodes for maximum and minimum values of this statistical figure.

With respect to the correlation coefficients (Figs. ~~3~~ 4 (e) and ~~3~~ 4 (f) for the dust episode and ~~4~~ 5 (e) and ~~4~~ 5 (f) for the fires episode), both simulations show high levels (around 0.9) of this statistical figure during the dust episode, except for those areas with high levels of AOD_{550}, where the correlations are lower (even with negative correlations values close to -0.5). Conversely, for the fire episode, correlation values are close to 1 both for both cases (NRF and RF) over the entire domain, especially over the areas with high values of AOD_{550}.

When considering the improvement (or not) of the AOD_{550} when including aerosol radiative feedbacks in the simulations, the difference in the MAE of the simulations between NRF and RF is estimated as defined in Eq. (2). For the dust episode (Fig ~~3~~ 4 (b)), a slight improvement (worsening) is produced over the areas with medium (high/low) levels of AOD_{550}, taking these changes values between -0.09 and +0.12. For the fire episode (Fig ~~4~~ 5 (b)), a worsening of MAE (difference NRF-RF of -0.02) is simulated close to the source of biomass burning aerosols. However, an improvement (up to +0.11) over areas further away from this source is estimated, which compensates the importance of including aerosol feedbacks in the simulations when assessing the improvement of worsening of simulations.

In the case of the $AE_{550/860}$ from MODIS, the results are analogous for both episodes. Low values (< 0.45, shown in Fig. 5 6 (a)) of this variable over the southeast of the domain are found. This, together with the high levels of AOD_{550} (Fig. 3 4 (a)), is a clear indication that natural dust aerosols coming from the Saharan desert govern the AOD_{550} levels here. On the other hand, for the fire episode (Fig. 6 7 (a)), the highest levels (around 1.6) are found over the north of Portugal, coincident with the fires areas, representing thus the emissions of biomass burning particles. Generally, for both simulations in both episodes, the model underestimates the high values of $AE_{550/860}$ and overestimates the low values.

For the dust episode, the MBE (Fig 65 (c) and (d)) minimum values are found of -0.65 and -0.62 for NRF and RF simulations (underestimation) and the maximum MBE takes values of 0.77 and 0.78, respectively (overestimation). Concerning the correlation coefficient (Fig. 5 6 (e) and (f)), also for both simulations the value of this statistic is lower than for AOD_{550}. Over most of the domain negative values are found (around -0.7) and positive values found are low (< 0.3).

On the other hand, during the fires episode (Fig. 67.) MBE minimum values (underestimation) are found around -0.61 and -0.65 for NRF and RF simulation, respectively, and maximum MBE values around 0.68 and 0.66 for NRF and RF simulations. With respect to the correlation coefficient, just for the dust episode, positive correlations (> 0.5) are located over the most of the domain, while negative correlations are estimated over the emission areas of biomass burning particles (with values around -0.8). However, for both episodes, a slight decrease for maximum and minimum MAE values (Table 12) are observed when the aerosol radiative feedbacks are taken into account.

At the same time, there is a slight improvement for RF simulations for the dust episode over the areas where the $AE_{550/860}$ is overestimated (reaching values of improvement of MAE of 0.13) and a slight worsening (values of improvement of MAE around -0.09) over the areas where this variable is underestimated (Fig 5 (b)). For the fire episode, a slight improvement (values of improvement of MAE of 0.16) is found over the south-eastern part of the domain and a slight worsening (around -0.18) over the rest of the IP (Fig 6 (b)).

3.3 Model output vs. AERONET data

This section shows the results of the comparison between model output and AERONET data. First, a linear regression is estimated (Figs. 78, 98 and 109) and the correlation coefficients are calculated for the daily averages (Table 23). For AOD at different wavelengths during the dust episode, the results indicate that the stations where the model show higher skills are Barcelona and Sagres (maximum correlation coefficient 0.72) and, in for the fire episode, Caceres and Evora (maximum correlation coefficient 0.9 and 0.85, respectively). For $AE_{440/870}$, during the dust episode, the best-represented stations are Caceres and Sagres (maximum correlation coefficient 0.62 and 0.57, respectively) and, for the fire episode, Autilla (0.75) and Evora (0.66). At this point, it is important to note that this comparison is obtained between a point (AERONET) and a cell (model outputs) covering the corresponding station coordinates following a nearest neighbour approach. In spite of the use of this approach, small errors on the spatial distribution of the model representation of the evaluated variables can appears, producing lower correlation coefficient values than the comparison with MODIS data, where the comparison is done cell (MODIS) ~~by~~versus cell (model output) with approximately the same resolution. Results do not indicate a clear improvement or worsening for both variables in both episodes when including the aerosol radiative feedbacks in our modelling configuration.

High levels of AERONET AOD are found between 2-10 of July 2010 due to the dust outbreak in Barcelona and Sagres (Fig. 1~~0~~1 (a) & (c)). The time series of these stations have been selected as representative among all AERONET stations in the IP affected by the Saharan dust outbreak (see Supplementary Material for information on the rest of AERONET stations over the IP). Maximum values of AERONET AOD occur between 7 and 10 July 2010. For AOD_{1020}, AOD_{870} and AOD_{675}, the model underestimates the highest levels of AOD, represented by the minimum bias values (Table ~~3~~2). On the other hand, between 2 and 6 of July 2010 (medium levels of AOD) the model overestimates the values of this parameter (Table ~~3~~2). When an underestimation (overestimation) is produced, the bias is lower (higher) for lower wavelengths. Sagres station~~s~~ lacks of AOD_{675} and AOD_{440} data. Finally, the behaviour of AOD_{440} in Barcelona is different from the other wavelengths due to the location of the station, close to a main street of the city where fine particles are emitted because of the road traffic.

For the fire episode, the shown stations are Caceres and Evora (see Supplementary Material for the rest of stations). In both stations, AOD presents the highest levels from 28 to 30 of July due to the wildfires occurred in Portugal (Table ~~3~~2). Except for the first two days, the model tends to underestimate the AOD values. For all wavelengths the bias or error, in both stations, increases when the wavelength decreases.

Regarding the $AE_{440/870}$, for the dust episode the AERONET values show~~s~~ low values corresponding to large particles (generally between 0 and 1) in Sagres station, indicating the dust origin of the particles at this site. For the fires episode, values generally range between 1.5 and 2.5 at Evora station, revealing the small size of the biomass burning particles. Generally for all stations in both episodes, the model overestimates (underestimates) $AE_{440/870}$ values when there are low (high) values of this variable. Hence, the model strongly undepredicts the variability of this variable for the two configurations.

3.4. Model output vs. EARLINET data

Finally, the results of the comparison between model output and EARLINET data are shown. In this section, only the dust episode is studied because the only station with available data for both the study episodes in the IP during the year 2010 is Granada. At this site, dust has an important contribution to aerosol loads. ~~Several~~ Two specific days (6 and 12 July 2010) are shown for the sake of brevity, but this discussion is valid for other days of this episode. It is important to notice the differences between both discrete profile resolutions: the model profile with 33 levels from the ground to approx. 20 km; and the profile measurement, which much higher vertical resolution (7.5 m). So the results below should be considered mainly from a qualitative perspective. However, in order to provide a more quantitative approach, the MAE of the model versus lidar observations is estimated.

As for the particle backscatter (BSCAT) at 532nm for 6 July ~~–~~2010 (Fig. 12~~1~~ (a)), the lidar detects a peak between 1.5 and 2 $x10^{-6}$ $m^{-1}sr^{-1}$ around 3250 m above sea level caused by a dust layer. Although the model outputs overestimate the BSCAT values, simulations capture the profile of BSCAT. Although NRF and RF model configurations perform similarly, there is a slight improvement in the MAE of the vertical profile (estimated after Eq. 3) when the aerosol radiative feedbacks are taken into account (Fig. 1~~1~~2 (a)). Average MAE is 6.37 $x10^{-7}$ and 6.22 $x10^{-7}$ $m^{-1}sr^{-1}$ for NRF and RF simulations, respectively. Henceforth, the normalized MAE is improved by 2.4% when aerosol radiative feedbacks are included in WRF-Chem simulations.

For the BSCAT for 12 July 2010 (Fig. 1~~1~~2 (b)), the model overestimates the BSCAT values of the vertical profile, as aforementioned. However, the shape of the vertical profile is correctly reproduced. Mean MAE is 3.14 $x10^{-7}$ and 3.12 $x10^{-7}$ $m^{-1}sr^{-1}$ at 355nm for NRF and RF simulations, respectively; and 4.1 x10-7 $m^{-1}sr^{-1}$ at 532nm for both cases. Here, the improvement when including aerosol radiative feedbacks is very limited, and estimated as 0.63% and 0.14% at 355 and 532nm, respectively.

4 Conclusions

The use of on-line coupled models is one of the most powerful tools to understand the different processes influencing the climate system. In particular, for the study of atmospheric aerosol particles realistic simulation of the combined ARI and ACI are needed, irrespective of the aerosols source, where the interactions of aerosols, meteorology, radiation, and chemistry are coupled in a fully interactive manner. The use of modelling tools requires the observational study of physical, chemical and optical properties of aerosol particles to establish its behaviour and to assess how good these properties are represented in on-line coupled models.

In this study, two configurations including/not including (NRF/RF simulations) the aerosol radiative feedbacks have been assessed against a number of remote sensing observations for two episodes characterized by dust and biomass burning aerosols, respectively.

For the comparison between model output and MODIS data, the best-represented variable is AOD, with low values of mean bias and high values of correlation coefficient both for NRF and RF simulations. Discrepancies between simulations and observations can be ascribed to errors in the model estimation of the aerosol dry mass, the fraction of particles for a given mass or the water associated with aerosols. On the other hand, we should bear in mind the known errors from observations. The inclusion of the aerosol radiative feedbacks produces a slight improvement in the model representation for medium values of this variable and a worsening for the lowest and highest values. At the same time, the model output of AE representation leads to underestimate the variability of this variable. This occurred for both episodes and may be related to the fact that the size distribution of the aerosol function within WRF-Chem considers a medium size of particles, smaller for dust and larger for fire~~s~~ particles. The inclusion of aerosol feedbacks does not produce a clear benefit, taking into account the expensive computational cost required for including the ARI and ACI in the model.

As well as for MODIS, for the comparison between model output and AERONET data, the results indicate that the best-represented variable is AOD. Generally, for both episodes, the model underestimates the levels of AOD, but the highest levels of this variable for dust episode are underestimated. It is important to note that the bias is usually higher for low wavelengths. In both episodes, the AE is overestimated for low levels and underestimated for high levels, since the modelled variability is strongly underestimated. For both variables, there is not a clear improvement of the model outputs for the aerosol radiative feedbacks simulation for any station in both episodes.

For the comparison between model output and EARLINET data, the results show a general slight improvement in the representation of vertical aerosol profiles when the aerosol radiative feedbacks are taken into account for all studied wavelength.

It is important to take into account these considerations to improve the time-efficiency when running the simulations, because the inclusion of aerosol radiative feedbacks in the simulations has a notable increase of the computational time. The improvements observed, in particular related to the vertical distribution of aerosols, justify the inclusion of aerosol radiative feedbacks in WRF-Chem on-line coupled model and the much higher time devoted to running the simulations.

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Apendix. List of acronyms.

ACI	*Aerosol-cloud interactions*
AE	*Angström Exponent*
AERONET	*AErosol Robotic NETwork*
AOD	*Aerosol Optical Depth*
ARI	*Aerosol-radiation interactions*
BSCAT	*Backscatter*
DB	*Deep Blue*

DT	*Dark Target*
EARLINET	*European Aerosol Research Lidar Network*
ECMWF	*European Centre for Medium-Range Weather Forecasts*
EuMetChem	*European framework for online integrated air quality and meteorology modelling*
IFS-MOZART	*Integrated Forecasting System - Model for ozone and related tracers*
IP	*Iberian Peninsula*
IPCC	*Intergovermental Panel on Climate Change*
IS4FIRES	*Integrated monitoring and modelling system for wild-land fires*
MACC-II	*Monitoring Atmospheric Composition and Climate-Interim Implementation*
MAE	*Mean Absolute Error*
MBE	*Mean Bias Error*
MEGAN	*Model of Emissions of Gases and Aerosols from Nature*
MODIS	*Moderate Resoultion Imaging Spectroratiometer*
NRF	*No radiative feedbacks*
r	*Correlation Coefficient*
RF	*Radiative feedbacks*
RRTMG	*Rapid Radiative Transfer Method for Global*
S.L.	*Significance Level*
TNO	*Netherlands Organization for Applied Scientific Research*
YSU PBL	*Yonsei University Planetary Boundary scheme*
WRF-Chem	*Weather Research and Forecasting model coupled with Chemistry*

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Table 1: Stations with available information in the IP for the 2010 episodes selected.

Station	Coordinates (Lat ,Lon)	Data network
Autilla	41,997, -4.603	AERONET
Barcelona	41.386, 2.117	AERONET
Burjassot	39.508, -0.418	AERONET
Cabo da Roca	38.783, -9.500	AERONET
Caceres	39.479, -6.343	AERONET
Evora	38.568, -7.912	AERONET
Granada	37.164, -3.605	AERONET & EARLINET
Huelva	37.016, -6.569	AERONET
Malaga	36.715, -4.478	AERONET
Sagres	37.048, -8.874	AERONET

~~**Table 2: Maximum AERONET AOD at 1020, 870, 675 and 440 nm; maximum and minimum error or bias values for both episodes at different representative stations. Values in italics represents a better bias for the RF than for the NRF simulations.**~~

Table 1: Maximum AERONET AOD at 1020, 870, 675 and 440 nm; maximum and minimum error or bias values for both episodes at different representative stations. Values in italics represents a better bias for the RF than for the NRF simulations.

Dust Episode																
	Barcelona Station								**Sagres Station**							
	1020nm		*870nm*		*675nm*		*440nm*		*1020nm*		*870nm*		*675nm*		*440nm*	
Max AERONET AOD value	0.43		0.45		0.47		0.55		0.39		0.42					
	NRF	RF	NRF	RF	NRF	RF	NRF	RF	NRF	RF	NRF	RF	NRF	RF	NRF	RF
Minimum bias value	**-0.22**	**-0.27**	**-0.20**	**-0.25**	**-0.20**	**-0.22**	**-0.34**	**-0.31**	**-0.19**	**-0.22**	**-0.17**	**-0.20**				
Maximum bias value	**0.11**	***0.09***	**0.12**	***0.11***	**0.16**	***0.14***	**0.24**	***0.19***	**0.23**	***0.17***	**0.27**	***0.21***				
Fire Episode																
	Caceres Station								**Evora Station**							
	1020nm		*870nm*		*675nm*		*440nm*		*1020nm*		*870nm*		*675nm*		*440nm*	
Max AERONET AOD value	0.23		0.30		0.38		0.67		0.35		0.37		0.41		0.61	
	NRF	RF	NRF	RF	NRF	RF	NRF	RF	NRF	RF	NRF	RF	NRF	RF	NRF	RF
Minimum bias value	**-0.13**	***-0.12***	**-0.17**	***-0.16***	**-0.21**	***-0.19***	**-0.40**	***-0.38***	**-0.24**	**-0.26**	**-0.25**	**-0.26**	**-0.24**	**-0.27**	**-0.33**	**-0.34**
Maximum bias value	**0.09**	**0.13**	**0.09**	**0.14**	**0.13**	**0.20**	**0.18**	**0.26**	**0.12**	**0.12**	**0.14**	**0.14**	**0.19**	**0.19**	**0.31**	***0.29***

Table 2: Values of correlation coefficient for the linear regression between AERONET and simulation daily means. Values in italic indicate the highest correlation coefficient among the different AOD/AE.

		Autilla		**Barcelona**		**Burjassot**		**Caceres**		**Evora**		**Granada**		**Huelva**		**Malaga**		**Sagres**	
		NRF	RF	NRF	RF	NRF	NRF	RF	NRF	RF	NRF	NRF	RF	NRF	RF	NRF	RF	NRF	RF
Dust Episode	*AOD_{1020}*	**0.17**	**0.19**	**0.59**	**0.50**	***0.15***	**0.04**	**0.39**	**0.33**	***0.58***	**0.53**	**0.35**	**0.32**	**0.41**	**0.35**	**0.22**	**0.18**	**0.71**	**0.68**
	AOD_{870}	**0.20**	**0.21**	**0.62**	**0.54**	**0.14**	**0.03**	***0.39***	**0.33**	**0.55**	**0.51**	**0.35**	**0.32**	**0.41**	**0.36**	**0.24**	**0.20**	***0.72***	**0.68**
	AOD_{675}	**0.21**	***0.22***	**0.70**	**0.64**	**0.15**	**0.04**	**0.38**	**0.33**	**0.49**	**0.45**	**0.37**	**0.33**	**0.43**	**0.39**	**0.26**	**0.22**		
	AOD_{440}	**0.18**	**0.18**	**0.72**	***0.72***	**0.12**	**0.03**	**0.30**	**0.27**	**0.28**	**0.27**	***0.38***	**0.34**	***0.48***	**0.44**	***0.29***	**0.25**		
	$AE_{440/870}$	***0.09***	**0.08**	***0.16***	**0.04**	***0.15***	**-0.05**	***0.62***	**0.51**	***0.38***	**0.36**	**-0.23**	***-0.32***	***0.22***	**0.08**	**0.20**	***-0.26***	**0.35**	***0.57***
Fire Episode	*AOD_{1020}*	**0.66**	**0.51**	**0.78**	**0.72**	**0.29**	**0.14**	**0.83**	**0.78**	**0.84**	**0.82**	**0.60**	**0.48**	**0.45**	**0.40**	**0.03**	**0.08**	**0.43**	**0.55**
	AOD_{870}	**0.68**	**0.52**	***0.80***	**0.76**	**0.35**	**0.18**	**0.86**	**0.82**	***0.85***	**0.82**	**0.61**	**0.51**	**0.47**	**0.42**	**0.03**	**0.08**	**0.44**	***0.56***
	AOD_{675}	**0.68**	**0.53**	**0.80**	**0.79**	**0.40**	**0.22**	**0.90**	**0.87**	**0.85**	**0.82**	***0.63***	**0.56**	**0.50**	**0.45**	**0.03**	***0.09***		
	AOD_{440}	***0.69***	**0.54**	**0.76**	**0.77**	***0.44***	**0.25**	***0.90***	**0.89**	**0.79**	**0.77**	**0.62**	**0.61**	***0.53***	**0.48**	**0.04**	**0.09**		
	$AE_{440/870}$	***0.75***	**0.71**	***0.50***	**0.46**	**0.06**	***0.18***	**-0.03**	***0.05***	***0.66***	**0.56**	***0.27***	**0.16**	***0.25***	**0.20**	***0.3***	**0.24**	**0.28**	***0.28***

~~***Dust Episode***~~																
	~~**Barcelona Station**~~								~~**Sagres Station**~~							
	~~*1020nm*~~		~~*870nm*~~		~~*675nm*~~		~~*440nm*~~		~~*1020nm*~~		~~*870nm*~~		~~*675nm*~~		~~*440nm*~~	
~~*Max AERONET AOD value*~~	~~0.43~~		~~0.45~~		~~0.47~~		~~0.55~~		~~0.39~~		~~0.42~~					
	~~NRF~~	~~RF~~	~~NRF~~	~~RF~~	~~NRF~~	~~RF~~	~~NRF~~	~~RF~~	~~NRF~~	~~RF~~	~~NRF~~	~~RF~~	~~NRF~~	~~RF~~	~~NRF~~	~~RF~~
~~*Minimum bias value*~~	~~**-0.22**~~	~~**-0.27**~~	~~**-0.20**~~	~~**-0.25**~~	~~**-0.20**~~	~~**-0.22**~~	~~**-0.34**~~	~~**-0.31**~~	~~**-0.19**~~	~~**-0.22**~~	~~**-0.17**~~	~~**-0.20**~~				
~~*Maximum bias value*~~	~~**0.11**~~	~~***0.09***~~	~~**0.12**~~	~~***0.11***~~	~~**0.16**~~	~~***0.14***~~	~~**0.24**~~	~~***0.19***~~	~~**0.23**~~	~~***0.17***~~	~~**0.27**~~	~~***0.21***~~				
~~***Fire Episode***~~																

	Caceres Station								Evora Station							
	1020nm		*870nm*		*675nm*		*440nm*		*1020nm*		*870nm*		*675nm*		*440nm*	
Max AERONET AOD value	0.23		0.30		0.38		0.67		0.35		0.37		0.41		0.61	
	NRF	RF	NRF	RF	NRF	RF	NRF	RF	NRF	RF	NRF	RF	NRF	RF	NRF	RF
Minimum bias value	**-0.13**	***-0.12***	**-0.17**	***-0.16***	**-0.21**	***-0.19***	**-0.40**	***-0.38***	**-0.24**	**-0.26**	**-0.25**	**-0.26**	**-0.24**	**-0.27**	**-0.33**	**-0.34**
Maximum bias value	**0.09**	**0.13**	**0.09**	**0.14**	**0.13**	**0.20**	**0.18**	**0.26**	**0.12**	**0.12**	**0.14**	**0.14**	**0.19**	**0.19**	**0.31**	***0.29***

Table 3: Values of correlation coefficient for the linear regression between AERONET and simulation daily means. Values in italic indicate the highest correlation coefficient among the different AOD/AE.

		Autilla		Barcelona		Burjassot		Caceres		Evora		Granada		Huelva		Malaga		Sagres	
		NRF	RF	NRF	RF	NRF	NRF	RF	NRF	RF	NRF	NRF	RF	NRF	RF	NRF	RF	NRF	RF
Dust Episode	*AOD 1020*	0.17	0.19	0.59	0.50	*0.15*	0.04	0.39	0.33	*0.58*	0.53	0.35	0.32	0.41	0.35	0.22	0.18	0.71	0.68
	AOD 870	0.20	0.21	0.62	0.54	0.14	0.03	*0.39*	0.33	0.55	0.51	0.35	0.32	0.41	0.36	0.24	0.20	*0.72*	0.68
	AOD 675	0.21	*0.22*	0.70	0.64	0.15	0.04	0.38	0.33	0.49	0.45	0.37	0.33	0.43	0.39	0.26	0.22		
	AOD 440	0.18	0.18	0.72	*0.72*	0.12	0.03	0.30	0.27	0.28	0.27	*0.38*	0.34	*0.48*	0.44	*0.29*	0.25		
	AE440/870	*0.09*	0.08	*0.16*	0.04	*0.15*	-0.05	*0.62*	0.51	*0.38*	0.36	-0.23	*-0.32*	*0.22*	0.08	0.20	*-0.26*	0.35	*0.57*
Fire Episode	*AOD 1020*	0.66	0.51	0.78	0.72	0.29	0.14	0.83	0.78	0.84	0.82	0.60	0.48	0.45	0.40	0.03	0.08	0.43	0.55
	AOD 870	0.68	0.52	*0.80*	0.76	0.35	0.18	0.86	0.82	*0.85*	0.82	0.61	0.51	0.47	0.42	0.03	0.08	0.44	*0.56*
	AOD 675	0.68	0.53	0.80	0.79	0.40	0.22	0.90	0.87	0.85	0.82	*0.63*	0.56	0.50	0.45	0.03	*0.09*		
	AOD 440	*0.69*	0.54	0.76	0.77	*0.44*	0.25	*0.90*	0.89	0.79	0.77	0.62	0.61	*0.53*	0.48	0.04	0.09		
	AE440/870	*0.75*	0.71	*0.50*	0.46	0.06	*0.18*	-0.03	*0.05*	*0.66*	0.56	*0.27*	0.16	*0.25*	0.20	*0.3*	0.24	0.28	*0.28*

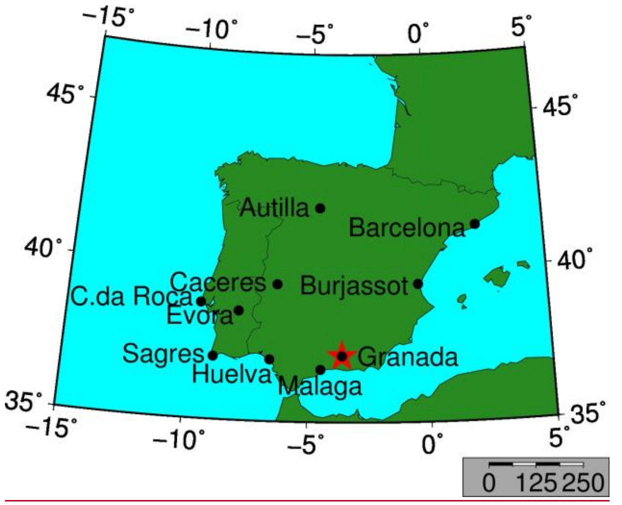

Figure 1: Map of the distribution of the AERONET (points) and the EARLINET (star) stations.

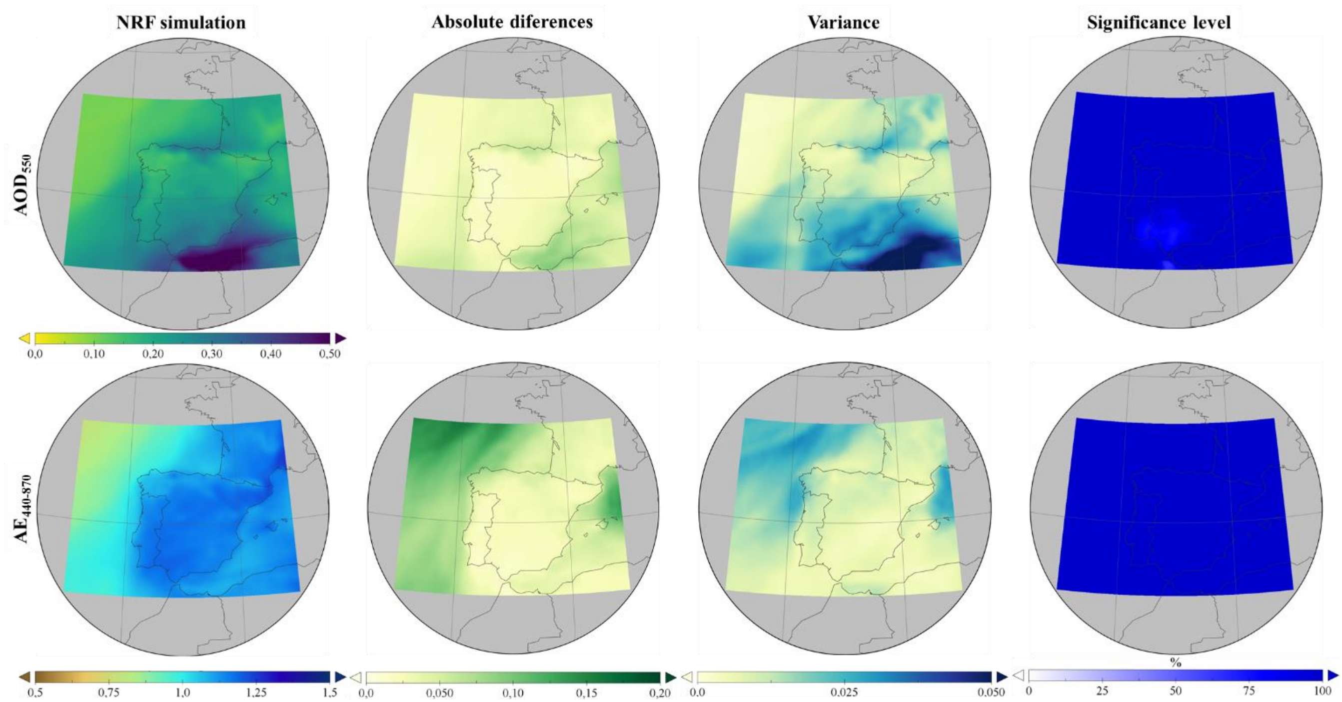

Figure 21: Dust episode (temporal mean from 28 June to 12 July). (tTop: AOD_{550}; bottom: $AE_{440/870}$). From left to right: (a) modelled value of the variable, (b) value of the absolute differences between NRF-RF simulations, (c)variance value of NRF simulation and (d) Significance .level (S.L.)L. values.

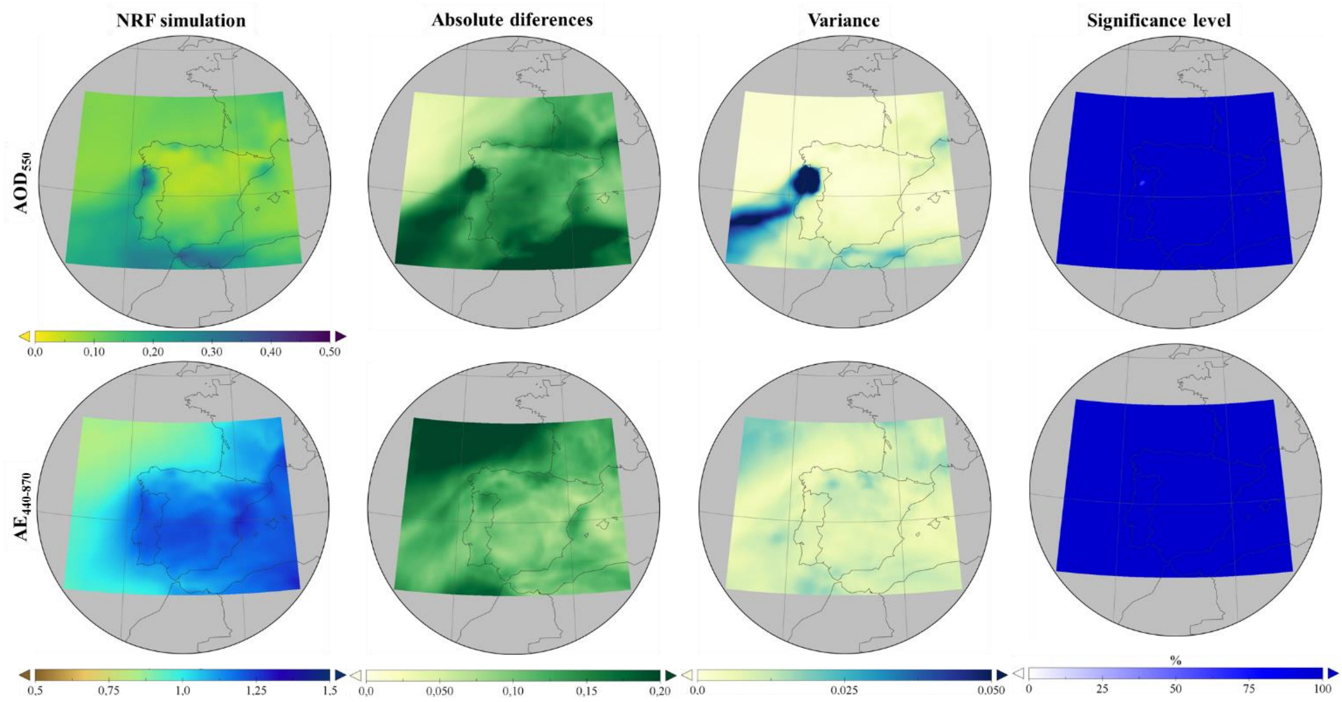

Figure ~~2~~3: Id. Fig. ~~1~~ 2 for the fires episode, temporal mean from 25 July to 7 August.

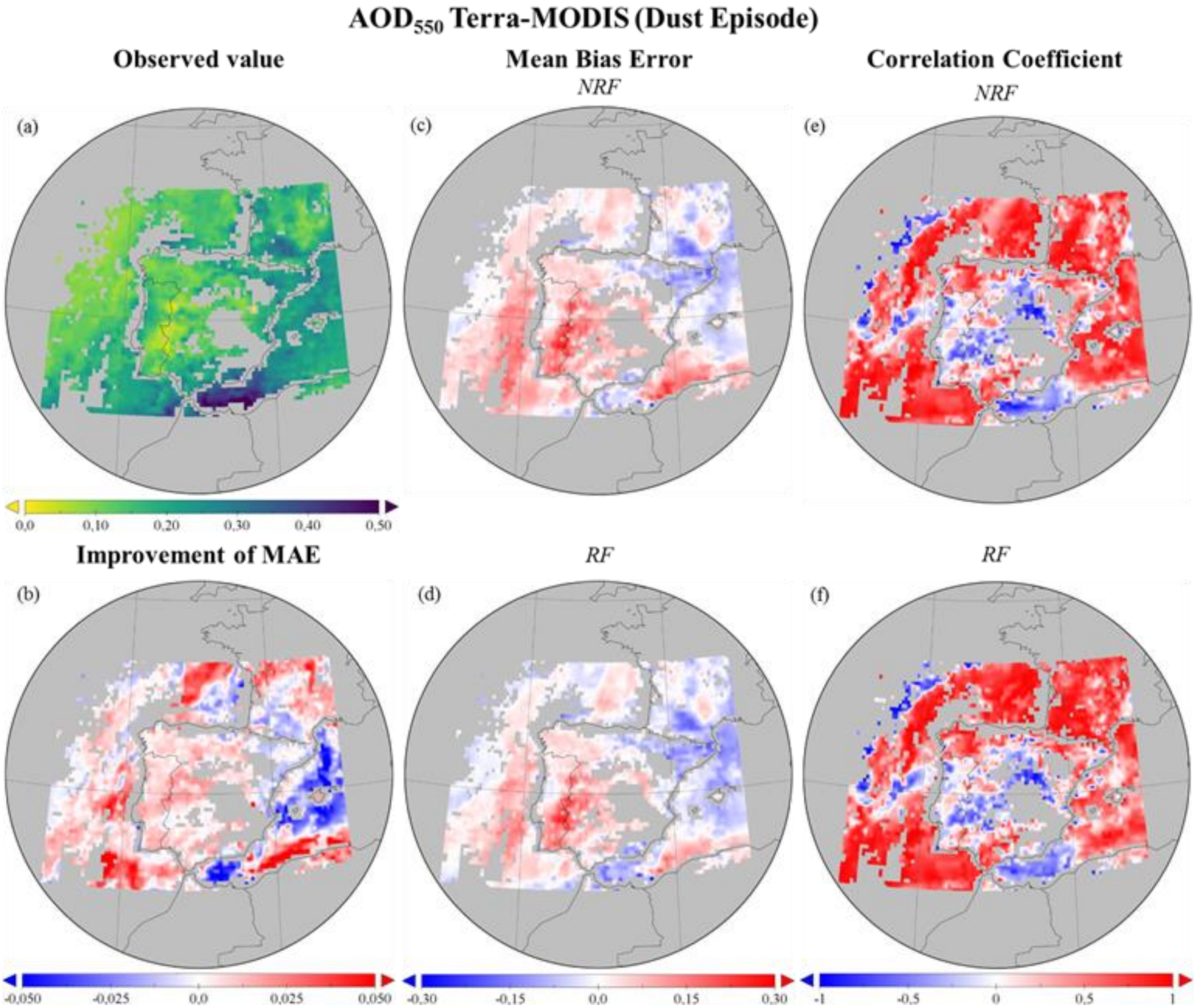

Figure ~~3~~4: Comparison of AOD_{550} model output vs. AOD_{550} from MODIS data for the dust episode (temporal mean from 28 June to 12 July). (a) AOD MODIS values. (b) Improvement of MAE due to the inclusion of RF (MAE in RF-NRF simulations). (c) and (d) MBE for NRF and RF simulations, respectively. (e) and (f) correlation coefficient for NRF and RF simulations.

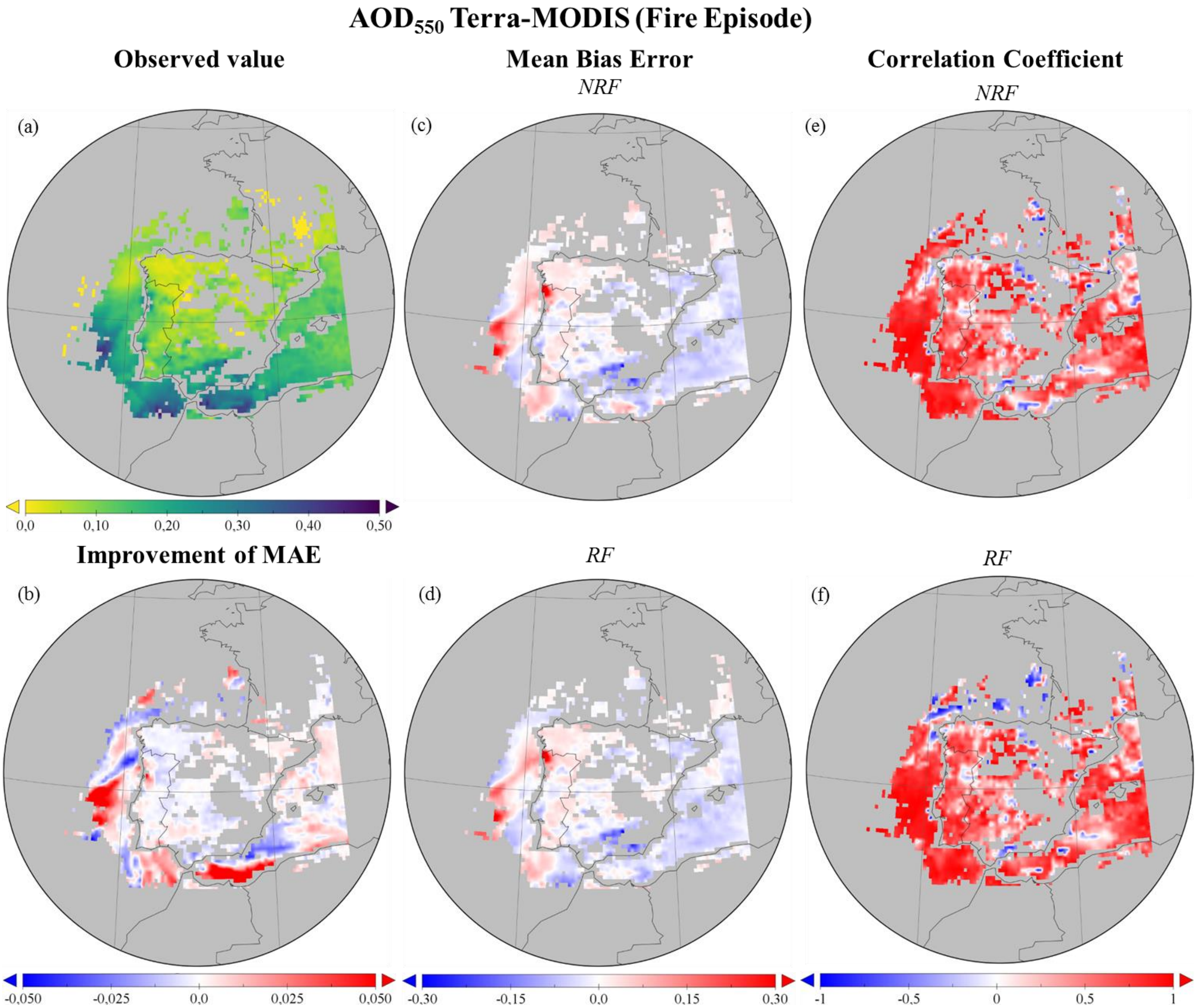

Figure 54: As Fig. 4 for the fires episode (temporal mean from 25 July to 7 August).

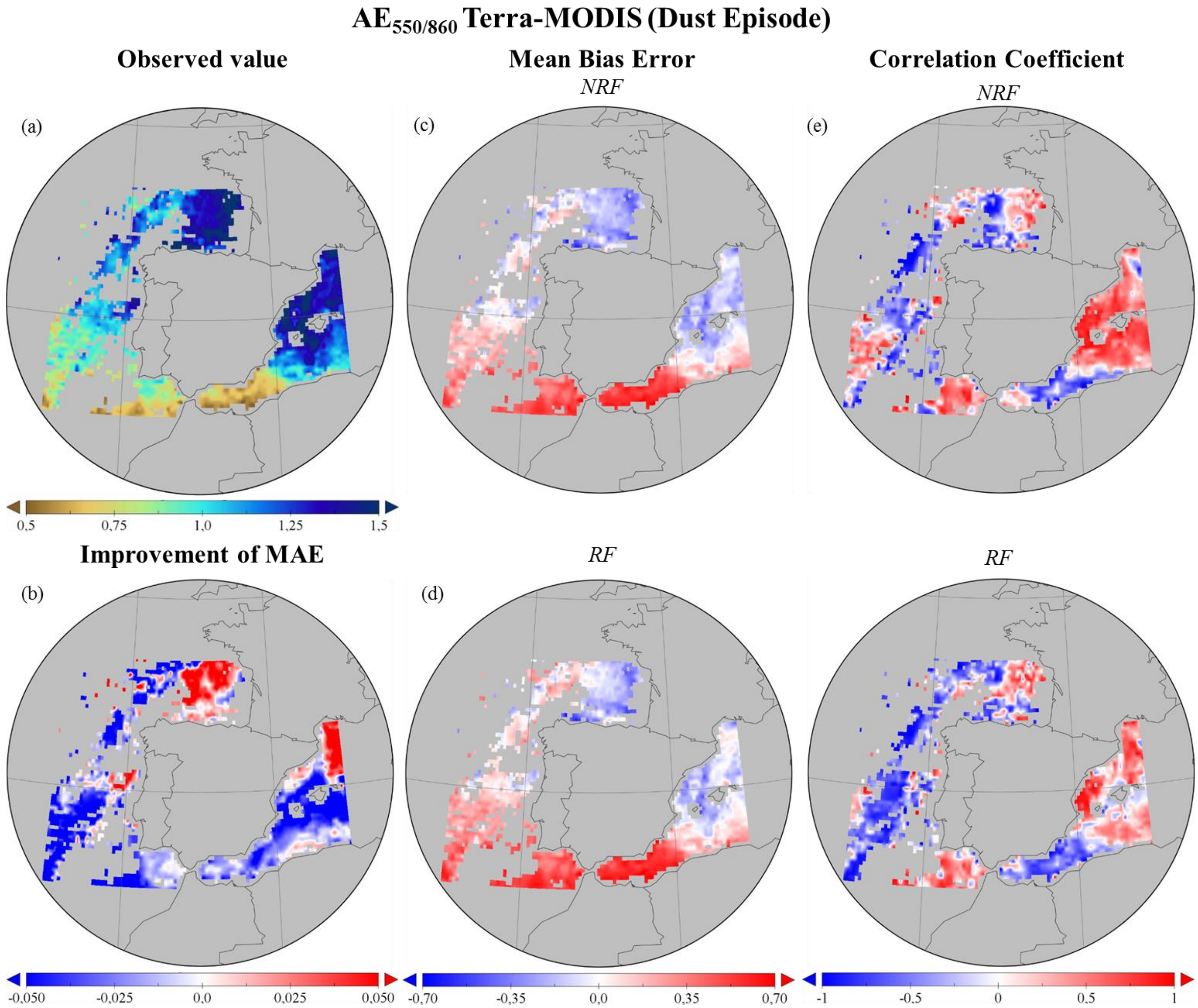

Figure 6~~5~~: Comparison of $AE_{550/860}$ model output vs. AE at same wavelength from MODIS data for the dust episode (temporal mean from 28 June to 12 July). (a) AE MODIS values. (b) Improvement of MAE due to the inclusion of RF (MAE in RF-NRF simulations). (c) and (d) MBE for NRF and RF simulations, respectively. (e) and (f) Correlation coefficient for NRF and RF simulations.

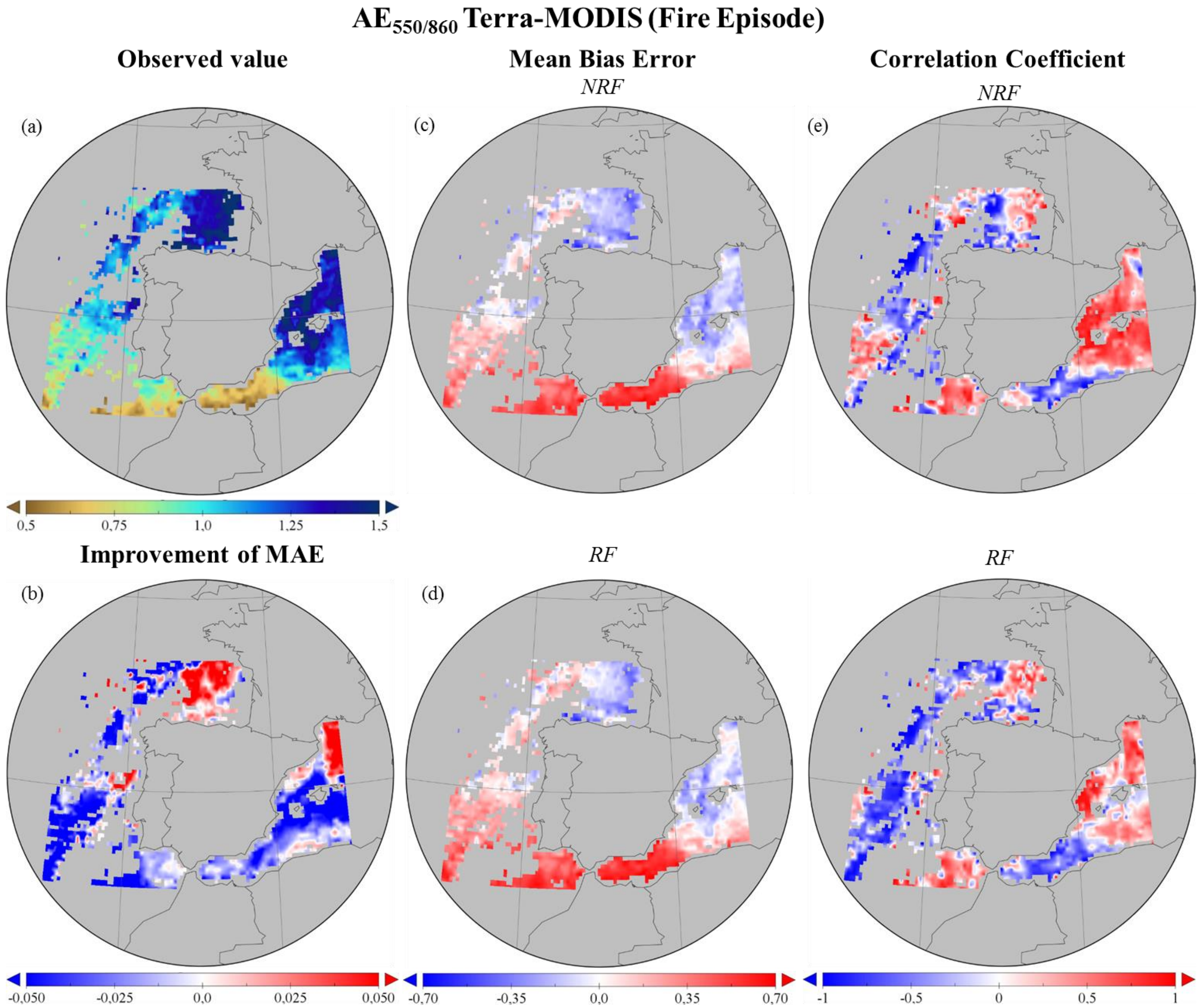

Figure ~~6~~7: As Fig. ~~5~~6 for the fires episode (temporal mean from 25 July to 7 August).

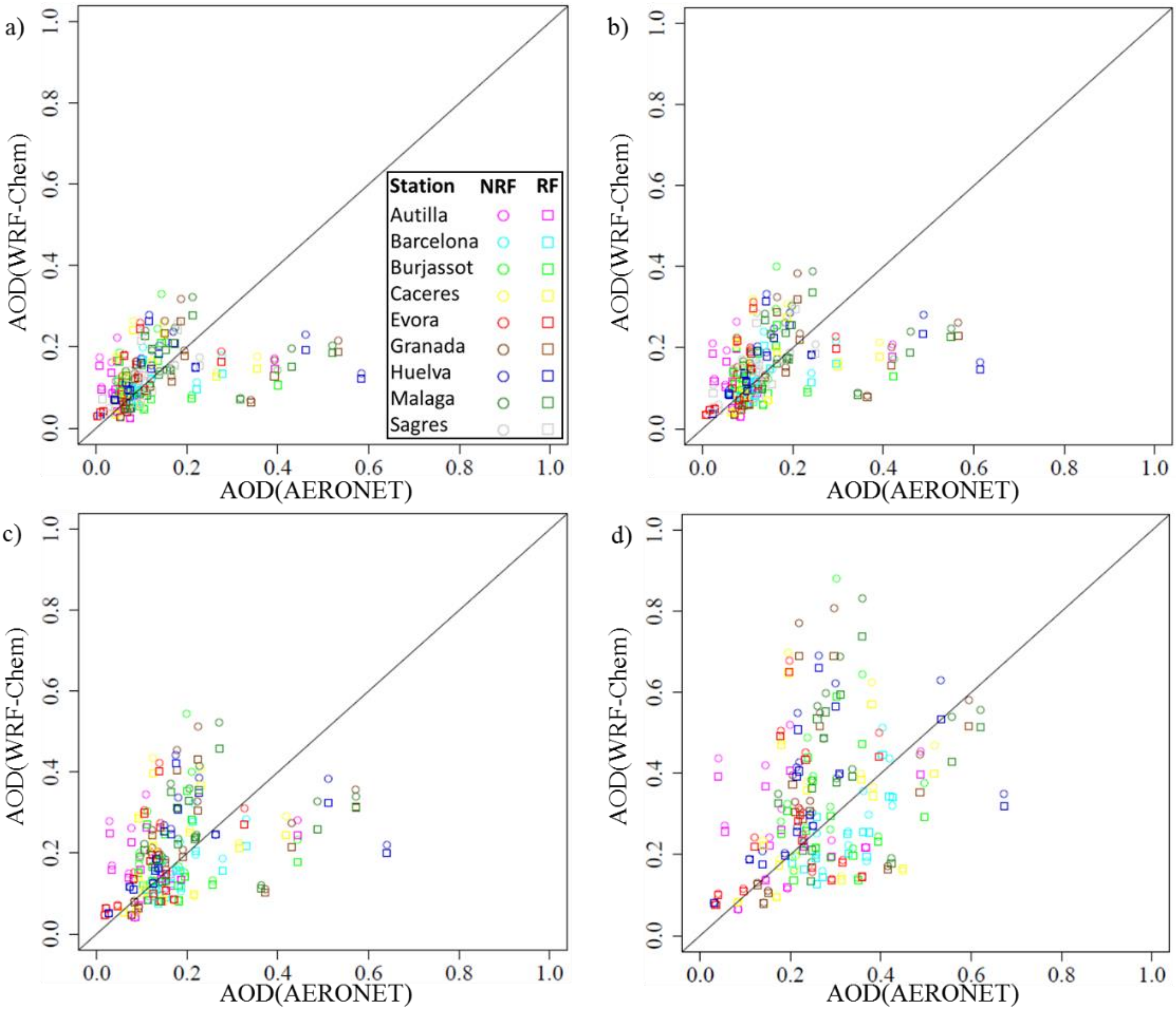

Figure 8~~7~~: Linear regression between AERONET (x) and simulations daily data (y; NRF in circles and RF in squares) for the dust episode (from 28 June to 12 July): (a) AOD$_{1020}$ (b) AOD$_{870}$ (c) AOD$_{675}$ and (d) AOD$_{440}$.

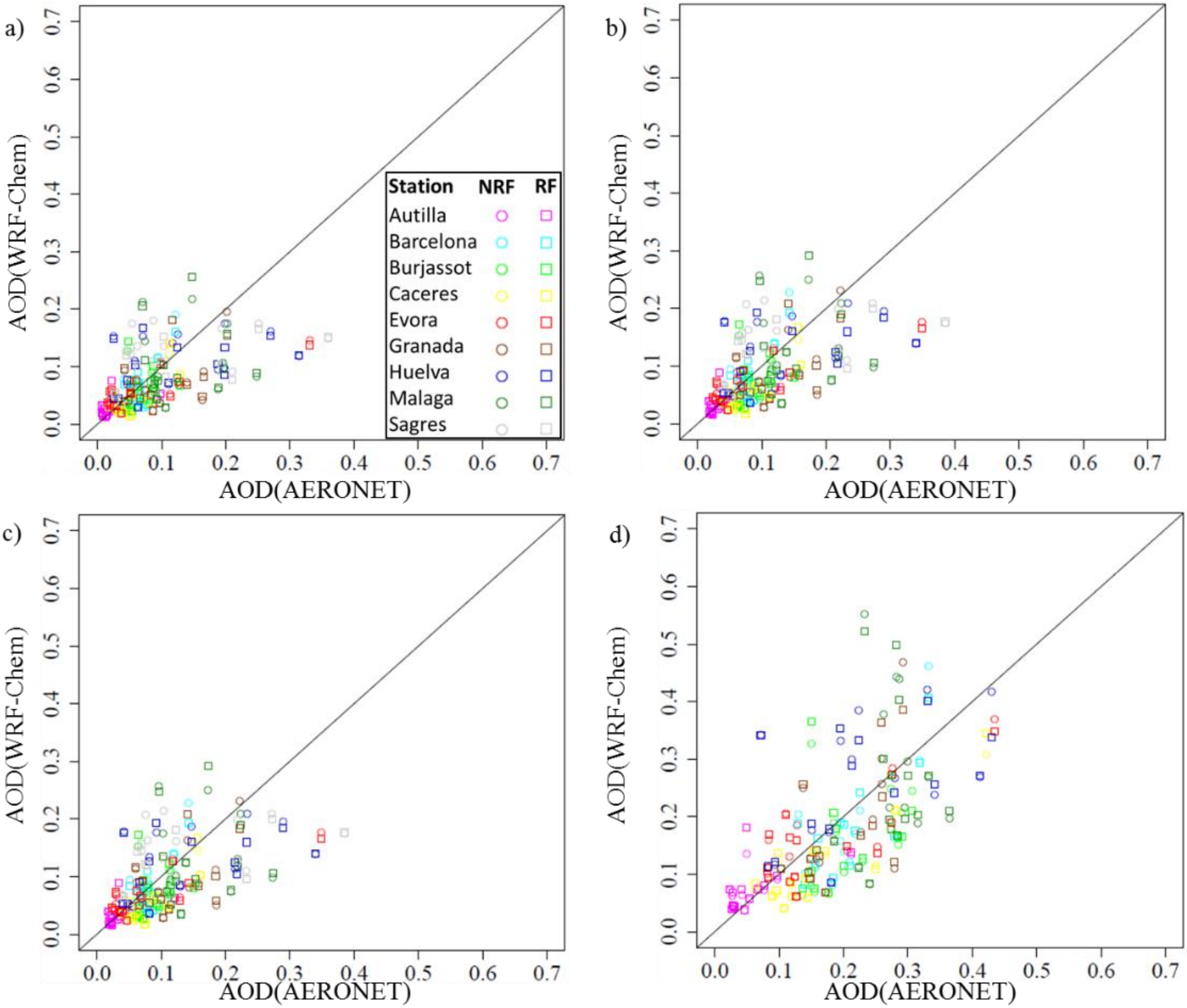

Figure 98: As Fig. 87 for the fires episode (from 25 July to 7 August).

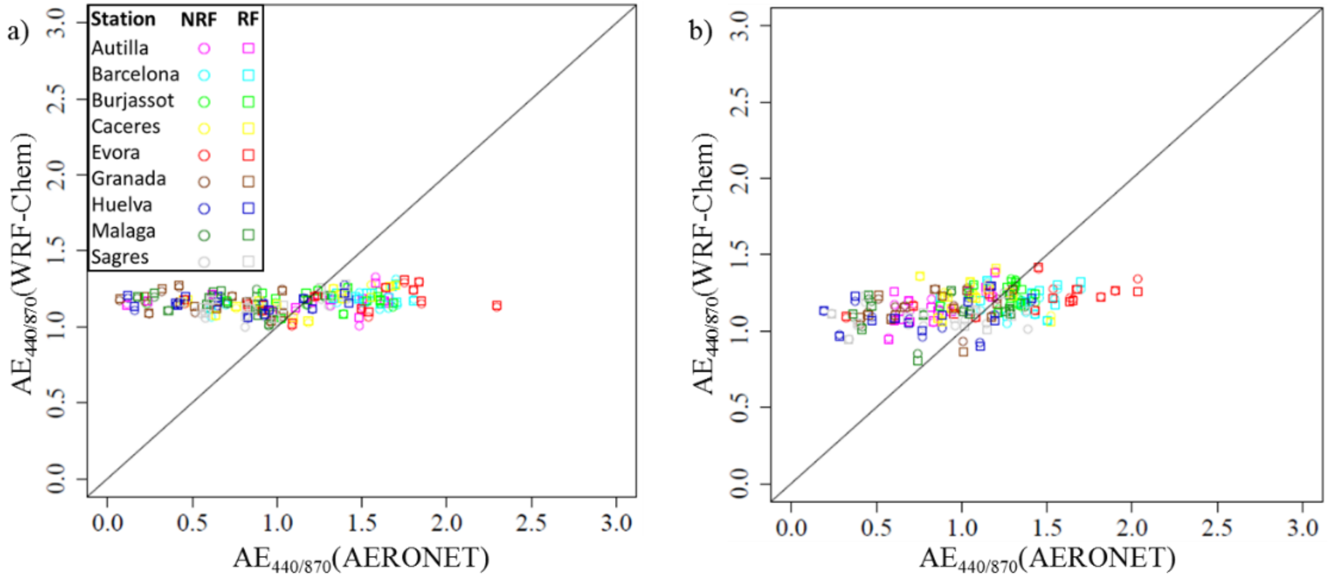

Figure 109: Linear regression between AERONET (x) and simulations daily data (y, NRF in circles and RF in squares) of AE$_{440/870}$: (a) dust episode (from 28 June to 12 July); (b) fire episode (from 25 July to 7 August).

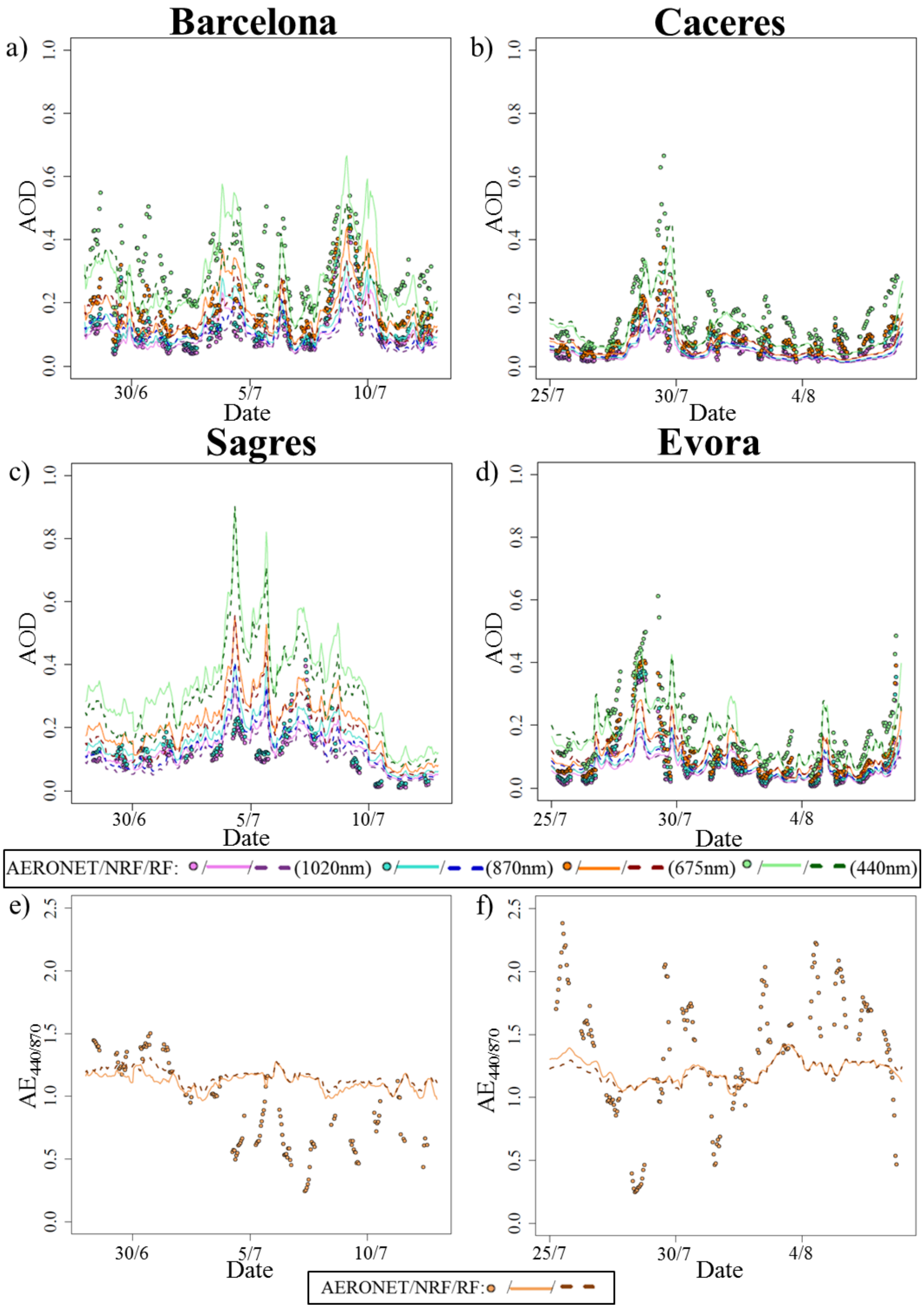

Figure 11~~0~~: AERONET (dots), NRF (line) and RF simulations (dashed line). AOD at different AERONET wavelengths: (a) and (c) Barcelona and Sagres stations for the dust episode (from 28 June to 12 July) and (b) and (d) Caceres and Evora stations for the fire episode (from 25 July to 7 August). (e) $AE_{440/870}$ in the Sagres station for the dust episode (from 28 June to 12 July) and for the (f) Caceres station for the fire episode (from 25 July to 7 August).

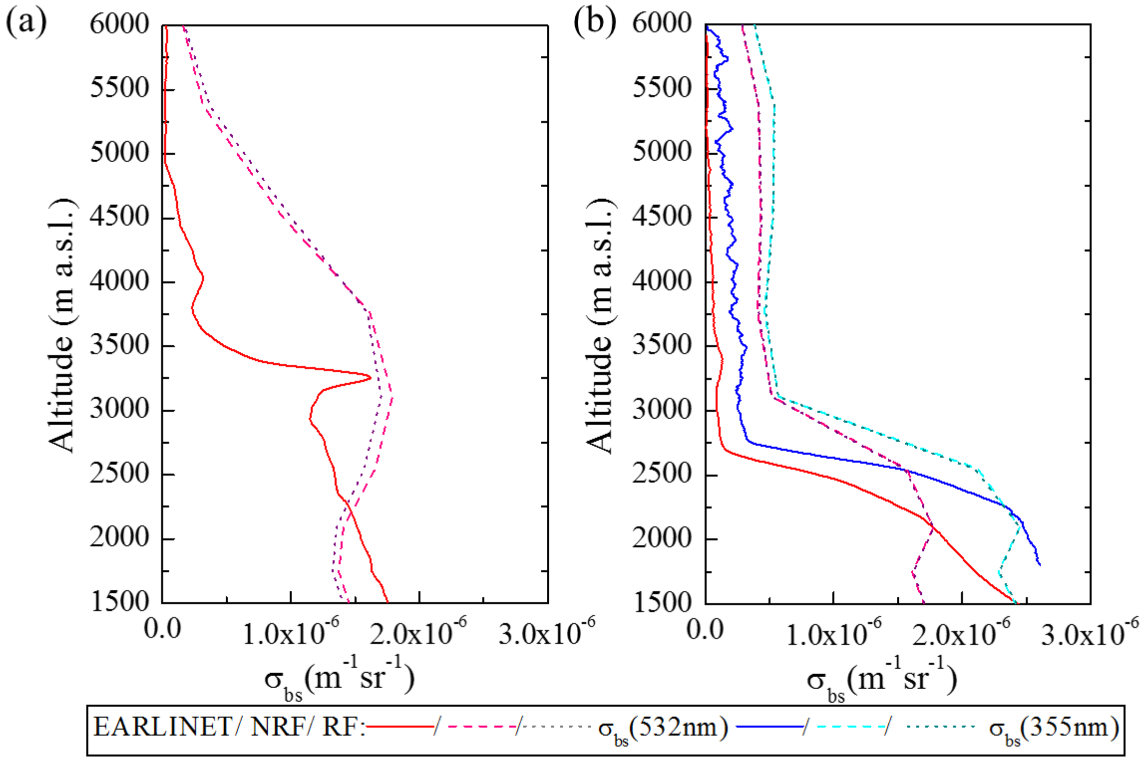

Figure 1~~1~~2: EARLINET (line), NRF (dashed line) and RF simulations (dotted line) of the backscatter coefficiente at 532nm and 355 nm. (a) For July 6, 2010 at 0200 UTC and (b) for July 12, 2010 at 1300 UTC.