Atmos. Chem. Phys. Discuss., 15, C1440–C1449, 2015
www.atmos-chem-phys-discuss.net/15/C1440/2015/

Atmospheric Chemistry and Physics Discussions
Open Access

ACPD
15, C1440–C1449, 2015

Interactive Comment

Interactive comment on "On the radiative impact of aerosols on photolysis rates: comparison of simulations and observations in the Lampedusa island during the ChArMEx/ADRIMED campaign" *by* S. Mailler et al.

Anonymous Referee #1

Received and published: 8 April 2015

In the paper entitled "On the radiative impact of aerosols on photolysis rates : comparison of simulations and observations in the Lampedusa island during the ChArMEx/ADRIMED campaign", the authors perform a CTM simulation for 1.5 month over the Mediterranean basin using WRF-CHIMERE to compute the radiative effects of aerosols on photolysis rates and on photochemistry, namely the production/destruction of ozone. The model 's results are compared to a variety of satellite and ground-based observations in terms of meteorological parameters, AOD, ozone total columns and actinic fluxes in the vicinity of Lampedusa island. This study is within the scope of

ACP journal and of sufficient originality to merit publication. However there are several weaknesses in the paper that should be resolved before publication.

In particular, the performance of the model is not very good : significant biases are observed on temperature (5 degrees in average), on total O3 column (60 Dobson Units) and on AOD (0.1 to 0.2 in the visible channel during dusty events). And the conclusions of the paper are based on maximum discrepancies on JNO2 and O3 mixing ratios of about 0.001 s-1 and 2 ppbv, respectively. I have a concern that the bias on temperature, ozone or AOD might be too large so that the final results presented without any uncertainty are not robust enough. The authors nevertheless performed an interesting sensitivity test on O3, giving more meaningful results. Adding sensitivity tests on temperature and on AOD would have been very much appreciated to give more insight to the paper and enable an assessment of the uncertainties. For example, what is the influence of the cold bias on ozone production/destruction, and thus on actinic fluxes ? More specific comments are given below.

Specific comments

p 7588, l 11 : give the coordinates of Lampedusa. The site is only described p 7595, but a lot of references to this site occur previously in the manuscript. The description of the location of the site should come earlier.

Sect 2.1.1. : I understand that the WRF-CHIMERE simulation has already been described in a previous paper (Menut et al., 2015). But some of the key features should be reminded in this paper. Do the authors use spectral nudging ? Is it applied also in the planetary boundary layer ? Which meteorological fields have been nudged towards GFS ? Is there any spin-up period before June, 1st? Given the size of the domain, some data at the beginning of the simulation should be discarded. On p 7602, l 23-25, the authors identify the period from 1 to 15 June to examine the impact of aerosols on photochemistry in CHIMERE. This is relevant if this period does not include spin-up.

P 7591, l 13 : The authors mention the model horizontal resolution of 60 km to explain

ACPD
15, C1440–C1449, 2015

to discrepancy observed on the temperature. This can indeed partly explain the lack of a daily cycle in WRF, but not the significant low bias. The differences between modeled and observed temperature are particularly important after 15 June. Does it correspond to a period with different air masses origins? To a denser cloud cover ? Does the 1 June – 15 June period belong to the spin-up time? This poor performance is particularly surprising as this region is well covered by observations assimilated in GFS. Is the bias already present in GFS meteorological fields ? Have the authors performed some sensitivity tests about the physical parameterizations in WRF simulation, in particular the PBL and radiative schemes?

Fig 1 : It is also relevant to compare wind direction, as it can enable to validate the transport patterns that will be computed through the backtrajectories analysis later in the paper.

P 7593 l 15 : The authors use Mischenko's code to compute the aerosol optical properties for dust. This code includes scattering for a variety of non-spherical particles. How is it taken into account in CHIMERE ? What are the assumptions on particles' shapes? A lot of studies have pointed out that the assumption of spherical aerosols in a Mie code can alter the scattering phase function, hence the radiative properties and the photolysis rates, relevant for this study. If the authors assumed only spheres, they should add a sensitivity study to assess the influence of this hypothesis on the photolysis rates affected by the presence of dust particles.

P 7593, l 17 : What is the reason of neglecting the influence of RH on the optical properties ? Are the observed particles totally hydrophobic ? Do the measurements highlight periods with high RH values ? It may be interesting to conduct a short sensitivity analysis to assess the influence of neglecting the scattering growth factor due to humidity on aerosol optical properties.

P 7594, l 16 : The discrepancy on ozone total columns is extremely high, but is not really commented in the paper. Is this due to a significant bias in CHIMERE or is

ACPD
15, C1440–C1449, 2015

the climatology used above 300 hPa poorly constrained ? The sensitivity analysis conducted by the authors on ozone concentrations at the end of the paper is very relevant to understand the impact of such a discrepancy.

Sect. 2.2 : The authors have written their own backplume model to identify the air masses origins. This very simple model seems to have been developed specifically for this study, since there is not any reference to a previous potential work based on this module. What is the reason of developing a new simplistic model for backtrajectories instead of using more common and sophisticated tools, such as HYPLIT, FLEXTRA, FLEXPART ? The model should at least be carefully described and validated : is the backplume model included online in WRF to account for the full dynamics and benefit from the dynamical time step of the simulation, or do the authors run this simple model as a post-processing step where meteorological fields are only available at the chosen output frequency ? What is the random function used to simulate the vertical mixing in the PBL ? Is is physically related to any of the WRF variables representative of turbulence (turbulent kinetic energy, vertical diffusion coefficient,...) ? The authors should also detail the numerical method implemented to take into account the advection.

P 7595, l 18 : The site of Lampedusa is well described here, but is mentioned several times in the previous sections. This should be reorganized.

P 7596, l 10-11 : Did the authors perform an intercomparison between the AOD measurements derived from MFRSR and AERONET when observations are available at the same time ?

P 7596, l 13 : AOD is known to vary with wavelength as the extinction coefficient does, i.e. a power law. There is not any physical reason to interpolate linearly the AOD. It is better to calculate the Angström exponent between two available wavelengths, and then derive the AOD at 400 nm.

Sect 3.1 : The title of this subsection has not been very well chosen since subsection 3.2 also describes a comparison between model outputs and observations. This should

ACPD

15, C1440–C1449, 2015

be reorganized.

Sect. 3.1.1 : AOD derived from CHIMERE is computed at 400 nm and 600 nm. To allow a fair comparison between model and observations, the authors could calculate the modeled AOD at 550 nm using the Angström exponent between 400 and 600 nm.

P 7599, l 10 : The AOD are averaged from 1 June to 15 July. If this period includes model spin-up, it should be reduced.

p 7600 : Over Europe, one can notice a factor 2 to 3 between CHIMERE-derived AOD and AOD retrieved from MODIS. Does it indicate a poor representation of anthropogenic pollution in the simulation ? Or is it mostly due to the discrepancy related a higher RH, and thus the scattering growth factor neglected in this study ?

p 7602, l 9 : A significant peak in the AOD is missed by the model. What is causing this peak ?

P 7602, l 14 : The reader would expect here a more quantitative comparison using some statistical scores (correlation coefficient, RMSE, bias,...), rather than a presentation in a rather qualitative style ("AOD values simulated by CHIMERE [...] compare well to observations").

Sect. 3.1.3 : I don't really understand the purpose of the comparison of the measured concentrations to the second model level. WRF model is terrain-following, suggesting that Lampedusa site is located in the first model layer. Are the altitudes given in this section in meters a.g.l or a.m.s.l. ? This is confusing.

p 7605, l 2-5 : Where does the number of 5 ug/m3 come from ? Figure 7 indicates an mean overestimation of 25 ug/m3 at the surface.

p 7605, l 14-25 : The authors need to be careful in their conclusions. Although the relation between aerosol mass concentrations and AOD have been shown by various studies to be almost linear for the different components of the aerosol taken individually, it is not always true for the bulk mass of aerosols, as its chemical composition may vary.

ACPD
15, C1440–C1449, 2015

A fairly good agreement in AOD does not necessarily lead to a good agreement in mass concentrations. Given that PM10 are strongly overestimated close to the surface (about 25 ug/m3), whereas AOD quite well reproduced, does it suggest an underestimation of dust transport in the free troposphere that could counterbalance the total aerosol column ? The authors also partly ascribe the surface overestimation to numerical diffusion (p7604, l 20). Does it indicate that numerical diffusion is better above the surface ?

P 7605, l 26 – p 7606, l 15 : Why are there different backtrajectories in Fig. 9 ? This has not been described earlier. Do they correspond to different tracers injected in a small volume around the release point ? If yes, the authors should give more details on the methodology used in Sect. 2.2 : how many tracers ? What are the boundaries of the volume where air parcels are released from ?

p 7608, l 2-3 : The bias looks indeed larger for the simulation without aerosols. Is this in agreement with Fig. 5 showing a slight overestimation of the AOD from CHIMERE in comparison to MFRSR/AERONET ? Higher aerosol loads should reduce the radiative fluxes available for NO2 photodissociation.

P 7608, l 8-9 : The good correlation between modeled and observed JNO2 values can hardly be linked to the optical properties of aerosols. In Fig. 11a for instance, the impact of including or not the aerosols is very weak because the AOD itself is not significant ($\sim$ 0.1). Aerosols have only a noticeable impact on JNO2 when the aerosol loads are important. The only variations on the back dashed line in Fig. 10 are correlated with the high aerosol optical depths. The authors may want to infer from their simulations the threshold for AOD that should be reached to have a noticeable influence on JNO2.

Sect. 3.2.3 : According to CHIMERE speciation, the authors could also identify in Fig. 13 the points mainly related to dust events and the points where the contribution of dust in the AOD is rather weak. They could therefore also plot the regression lines JNO2 =

ACPD
15, C1440–C1449, 2015

f(AOD) and JO1D=f(AOD) for their own dataset, which would give more insight to the paper and would enable them to properly compare their results with previous studies (Casasanta et al., 2011 ; Gerasopoulos et al., 2012).

p 7614, 4-7 : The maximum difference on O3 is 2 ppbv. What is the associated uncertainty ? Is this result robust according to the various assumptions made in the simulation : neglecting scattering growth factor, assuming spherical particles ? The discrepancy on temperature shown in Fig. 1 is about 5 degrees. What is the impact of this cold bias on ozone production or destruction ? The maximum difference between a run with aerosol forcing and a run without aerosol forcing is 0.001 s-1 (Fig. 11b) for a dust event. The authors must check how robust is this result, since most of the main conclusions are linked to such small discrepancies. A sensitivity test on temperature on JNO2, JO1D, O3 mixing ratios would have been very much appreciated.

p 7615, l 6-12 : this sensitivity test is relevant for the paper, but its description should come earlier in the manuscript, when the different simulations are presented (p 7590). It should be also mentioned at the beginning of the conclusion, together with the description of the REF and NA simulations.

p 7616, l 10-13 : The authors do not provide any explanation for this counter intuitive result. Does it highlight a compensation effect in the REF simulation with a too low stratospheric O3 associated to a too high AOD during dusty events ? It would be interesting to set up a simulation including both a 18% increase in ozone (as in O3+ simulation) and a decrease in aerosol emissions to fit the measured AOD in Fig. 5.

p 7616 : the conclusion should include the overestimation of AOD in CHIMERE during dust events.

Technical comments

In the text, JO1D is used everywhere. But in figures, it is often called JO3 or JO3(1D). Please use JO1D everywhere in the manuscript and in the figures for consistency.

P 7590, l 8Âă: 1 June to 15 July

A lot of acronyms have not been defined in the text, e.g. WRF, NCEP, GFS, MEL-CHOIR, HTAP, EDGAR, LMDZ-INCA, GOCART, ADIENT, AERONET...

P 7591, l 11-13 : please reformulate. “here” should be avoided. Use rather “shown in Fig. 1”.

P 7594 l 4 : what do the authors call the “online” ozone concentrations ?

P 7598, l 15 : anthropogenic

p 7598, l 22: Saharan

P 7599, l 11 : CHIMERE realistically reproduces

P 7599, l 24 : Capo Verde islands

P 7600, l 25 : Replace “thick aerosols” by “high aerosol loads”

p 7601, l 19 : steadily decreases

P 7603, l 6 : The authors must choose only one acronym : Lidar or LIDAR and keep it along the whole paper.

P 7603, l 14-17 : this sentence is very long and hard to read. Please reformulate.

P 7603, l 18 : display a very similar structure

P 7604, l 18 : overestimation

P 7605, l 3 : boundary

p 7605, l 10 : total

p 7605, l 11 : “This is the case”... “and primary anthropogenic”

P 7606, l 11 : most

p 7606, l 16 : “as a balance” does not mean “as a summary”

p 7608, l 1 : remove “that”

P 7609, l 15-22 : a reference to Fig. 10 and Fig. 11 is missing here.

P 7610, l 15 : different

p 7610, l 24 : we examine

p 7611, l 6 : at local noon

p 7614, l 1 : The ozone concentrations

p 7615, l 8 : a sensitivity simulation identical

p 7616, l 9 : to be biased

P 7617, l 29 : the REF simulation

p 7618, l 11 : with in situ measurements

Table 1 caption : Sectional bins

Fig 4 : the subfigures are very small and difficult to read.

Fig. 5 caption : evolution of modeled AOD...

Fig 6 : For sake of clarity, it would be better to use the dateticks already used in Fig. 5, 7, 10, and 14 for consistency between figures.

Fig 6 : the caption is wrong since (a) and (b) have been inverted

Fig 15 : The subfigures are too small and difficult to read. It might be better to display the subfigures as a 2x2 matrix. A subfigure showing NOX could be added for the discussion about the regions with higher/lower ozone.

List of authors : first names should be switched to abbr.

ACPD
15, C1440–C1449, 2015

Interactive comment on Atmos. Chem. Phys. Discuss., 15, 7585, 2015.

ACPD

15, C1440–C1449, 2015

Interactive Comment