

## Answer to Anonymous Referee # 1

We would like to thank reviewer #1 for his very careful reading of our manuscript, yielding to scientific questions that needed to be raised, as well as the correction of many grammatical and typographic mistakes. Our answers are given below : in black bold fonts are the Reviewer comments, in blue our answers, and in green the description of the corresponding changes in the manuscript.

***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 robust enough. The authors nevertheless performed an interesting sensitivity test on O3, giving more meaningful results. Adding sensitivity tests on temperature and AOD would have been very much appreciated to give more insight to the paper and enable an assessment of the uncertainties. For example, what if the influence of the cold bias on ozone production/destruction, and thus on actinic fluxes ?***

We understand the concern of the Reviewer, particularly regarding the uncertainty on temperature. While  $j(\text{NO}_2)$  is very robust and has only a small dependence on temperature and on ozone column (Dickerson et al., 1982), this is not the case of  $j(\text{O}_1\text{d})$ .

$J(\text{O}_1\text{d})$  dependence on temperature is significant (see Fig. 9 of Dickerson et al. 1982). An decrease of the temperature from 297 to 292 K, which is the typical underestimation of temperature at Lampedusa in our study during daytime, corresponds to an decrease of about 10% of  $J(\text{O}_1\text{d})$ . Therefore, the error on temperature in our model's meteorological inputs may cause an underestimation of  $j(\text{O}_1\text{d})$  photolysis rates of about 10%.

However, it is worth noting that the three runs (REF run with the reference configuration, NA run without AOD, and O3+ with enhanced ozone column) work exactly with the same temperature and meteorological conditions, so that the effect of temperature does not prevent us from comparing the REF and NA runs (in order to retrieve the effect of the AOD), nor from comparing the REF and O3+ run (in order to retrieve the impact of the ozone column). The O3+ run already gives the possible magnitude of the uncertainties on the modelled  $j(\text{O}_1\text{d})$  values (on the order of 20%, see Fig. 10).

As the impact of temperature of ozone and nitrogen dioxide photolysis rates is already well-known, we did not perform a new sensitivity test on temperature. However, we added a paragraph commenting the effects from this temperature bias, based on the Dickerson et al. (1982) results.

***p. 7588, l. 11 : give the coordinates of Lampedusa/ the site in 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***

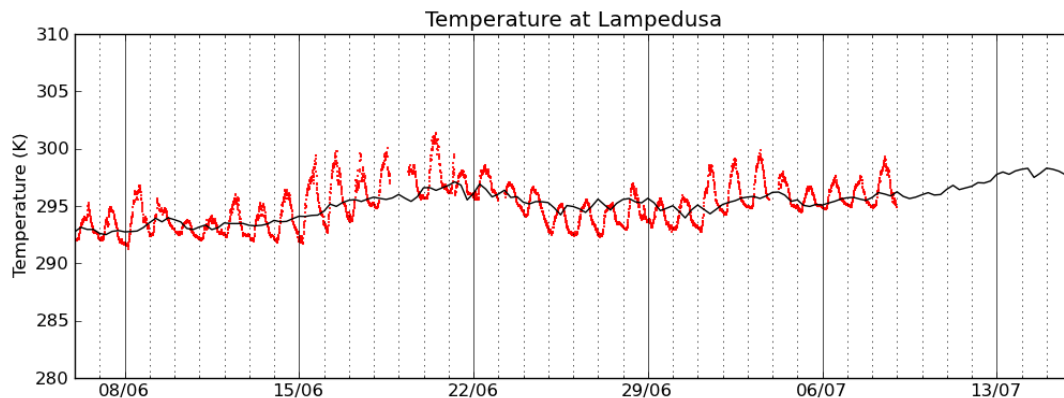
The following words have been added at this point :

Lampedusa, a small island located off the coasts of Sicily and Tunisia, hosts a Station for Climate observation run by the ENEA on its North-Eastern Coast (35.5°N, 12.6°E)

***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.***

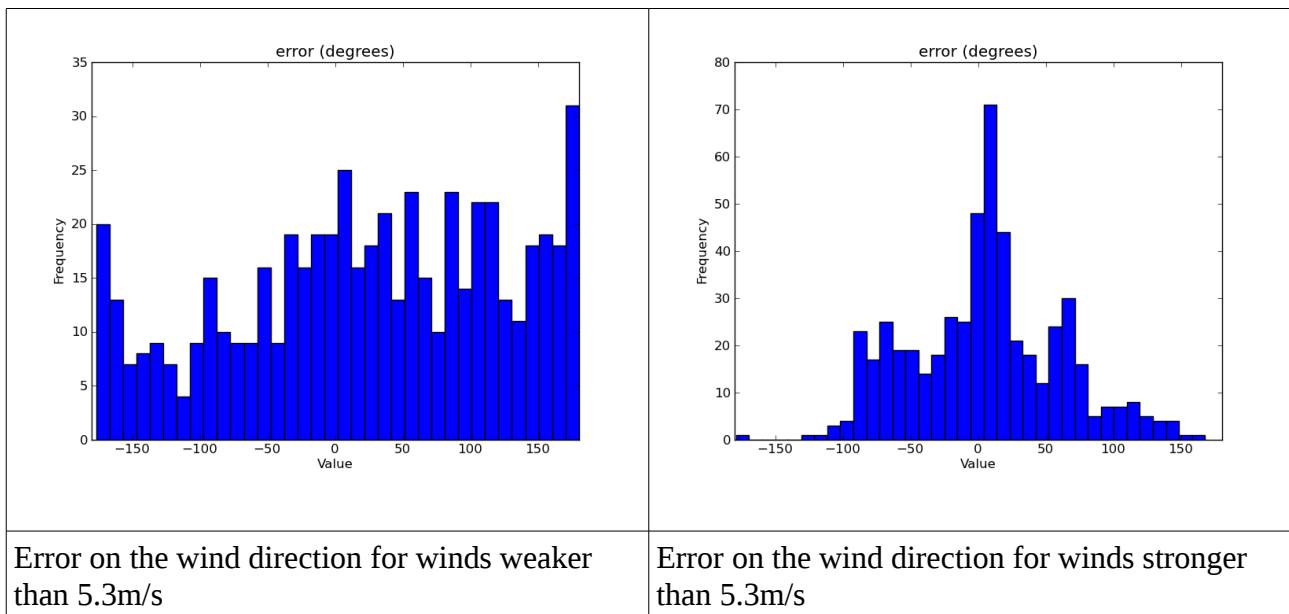
This has been done by including a short description of the meteorological model configuration, in two short paragraphs at the beginning of Section 2.1.1.

**p. 7591, l. 13 : The authors mention the model horizontal resolution of 60 km to explain the discrepancy observed on temperature. This can indeed partly explain the lack of a daily cycle in WRF but not the significant low biases (...) 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 ?**



We performed this test, and found that the bias in temperature at the first model level relative to the measured values (red dots) is not present in the GFS data (black line) : no significant bias appears between the GFS data and the measured values at Lampedusa. This changes the interpretation of the temperature bias as presented in the initial version. This bias can now be attributed to problems in the WRF simulation itself, rather than resolution issues, since the GFS data interpolated at the same resolution as the simulation doesn't have such a bias. It is now explicitly stated in the manuscript that the temperature bias comes from the meteorological model itself rather than from the NCEP/GFS analysis. Even though it is problematic to have used simulations with such a bias on temperature, we examine in the corrected version the possible consequences of this bias on the results of the study, using the well-known relationships of  $j(\text{O}1\text{d})$  with temperature. On the other hand, a possible bias on temperature does not affect much the  $j(\text{NO}2)$  photolysis rates, as also mentioned in the revised version.

**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.**



We include here the comparison of the hourly wind direction between the model and observations of the 10m wind measured at the Lampedusa ground station. We observed that the model performs poorly regarding the weak winds, and relatively well for stronger winds. Therefore, we provide here two histograms : one for the winds lower than the median ( $<5.3\text{m/s}$ , 551 measurement points), one for the stronger winds ( $>5.3\text{m/s}$ , 523 measurement points). For weak winds, the direction of the simulated wind is hardly correlated to the simulated one : the wind error is distributed relatively evenly between  $-180^\circ$  and  $+180^\circ$ . For stronger winds, on the contrary, the wind distribution is very peaked around  $0^\circ$ , and for half of the times the error lies between  $-35.5^\circ$  and  $+35.5^\circ$ . In less than 10 % of the cases however, the absolute error on wind direction is above  $90^\circ$ .

The strong errors for weak winds can be explained by the fact that, when the synoptic wind is weak, the wind at the coastal location of the Lampedusa station is dominated by very local effects such as land-breeze and sea-breeze which cannot be represented adequately at the model resolution. On the other hand, for stronger wind velocities, synoptic-scale structures which are better represented by the model dominate, with a still significant error on wind direction, possibly due to local effects as well.

***p. 7593, l. 15 : the authors use Mishenko's code to compute the aerosol 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' shape ? (...) If the authors assumed only spheres, they should add a sensitivity test to assess the influence of this hypothesis on the photolysis rates affected by the presence of dust particles.***

Non-sphericity has not been taken into account in this study because, in our model as in most models, uncertainties related to the size distribution of dust and other aerosols are still large, and need to be fixed before one can examine seriously in such models the possible effect of non-sphericity. Doing such a sensitivity test would give the illusion that non-sphericity of particles is among the main causes of errors on photolytic rates, which is certainly not the case so far.

However, we agree that a specific study on this issue might be relevant at this time to assess exactly what is the possible impact of non-sphericity of aerosols on photolytic rates, among other causes of error such as size distribution and the uncertainty of the mixing state of aerosols. We think that this i, however, beyond the scope of the present paper.

***p. 7593, l. 17 : What is the reason for neglecting the influence of the RH on the optical properties ? (...) 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.***

This sentence was actually misleading and badly written. Actually, water uptake by aerosols are taken into account by CHIMERE using the ISORROPIA module for hygroscopic species such as ammonium, sulphates and nitrates, and the optical effect of this liquid-phase water is explicitly taken into account by the Fast-JX module. This is now explicitly stated in Section 2.1.2.

Regarding the particular case of dust, it is generally considered that water uptake by dust particles has generally too small an effect to significantly affect the particle sizes (Herich et al., 2009).

***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 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.***

A discussion has been introduced on this point, using the Ziemke et al. (2011) ozone climatology. It is concluded that the low bias on ozone column can be attributed mostly or entirely to the poorly-constrained ozone climatology used above 300 hPa (end of Section 2.1.2)

**Sec 2.2 : The authors have written their own backplume model to identify the air mass origin. This very simple model seems to have been developed specifically for this study (...). What is the reason of developing a new simplistic model for backtrajectories instead of using more common and sophisticated tools such as HYSPLIT, FLEXTRA, FLEXPART ? (...) The authors should also detail the numerical method implemented to take into account the advection.**

We actually use this homemade backtrajectory model mostly for historical reasons. We think that it is based on reasonable hypotheses of laminar advection in the free troposphere, and random mixing within the boundary layer. However, we are not able to discuss the advantages and limitations of this model compared to other widespread tools as HYSPLIT, FLEXTRA and FLEXPART.

Additional explications of how the backplume model works have been added to the papers, as well as the following statement explaining our choice : « Even though this backplume model is possibly not comparable to state-of-the-art models such as HYSPLIT or FLEXPART, this model has been chosen for its simplicity of use in a study in which backtrajectories are not a critical part. It does not necessarily imply that such a simplified formulation would be adequate for studies in which accuracy of the backplume simulations is critical. »

***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.***

This has been done by adding the coordinates of Lampedusa and a brief statement on its geographical location at the beginning of the introduction.

**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 ?**

Such a comparison is provided in Di Sarra et al. (2015), already cited. These authors find a mean bias of the MFRSR AOD always smaller than 0.004 for long-term series (1999-2013), and a  $r^2$  correlation coefficient always above 0.97 at all wavelengths. These results are now recalled in the paper : « It was shown in Di Sarra et al. (2015) that the mean bias of the MFRSR AOD relative to the AERONET measurements is always smaller than 0.004 for long-term series (1999-2013), with a  $r^2$  correlation coefficient always above 0.97 at all wavelengths between the AERONET and the MFRSR measurements. The very good correspondance between both time series make it possible to use the MFRSR measurements to complete the AERONET time series, as done in the present study. »

**p. 7596, l. 13 : AOD is known to vary with wavelength as the extinction coefficient does, i.e. A power law. There is no 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.**

This remark is totally correct, and concerns Figs. 3, 4, and 5 of the initial article (Figs. 4, 5, 6 of the revised version). The calculations for these figures and the corresponding statistical scores have been redone, and the text of the article has been modified accordingly. The effect was however not very considerable, due to the fact that the interpolation is done between wavelengths that are relatively close to each other, reducing nonlinearity.

The left panel of Fig. 4 has also been updated by restricting the averaging period in order to drop a 5-day spinup period, and performing the interpolation of the CHIMERE AOD at 550 nm from the AOD values at 400 nm and 600 nm, as requested. In spite of this improved methodology, the resulting figure does not differ much from the initial version. The caption of this figure has been modified accordingly.

The right panel of Fig. 4 has been updated by restricting the averaging period to drop the 5-day spin-up period, consistently with CHIMERE data.

Fig. 5 has been updated by showing the AOD at 550 nm (instead of 600 nm in the initial version) to ensure consistency with the MODIS values (available at 550 nm). The interpolation is performed using the Angstrom power law as requested. The caption has been changed accordingly.

Fig. 6 has been redone by performing the interpolation according to the Angstrom power law for the interpolation of the AERONET and MFRSR values to 400 nm. No significant changes arise due to the fact that the wavelengths for which measurements are available are very close to 400 nm (380 nm and 440 nm in the case of AERONET, 416 nm and 440.6 nm in the case of MFRSR)

Table 3, which is added in the revised version of the article also uses interpolation following the Angstrom power law for the measured values.

***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 be reorganized.***

Title of Sect. 3.1 has been changed as « Representation of the aerosols in the model : comparison to observations »

**Sect. 3.1.1 : AOD derived from CHIMERE is computed at 400 ,m 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..**

This has been done, as described above. Figs. 4 and 5 are updated consistently, with no changes substantial enough to change the discussion.

**p. 7599, l. 10 : The AOD are averaged from 1 June to 15 July. If this period includes spinup, it should be reduced.**

Yes, this period included spinup. In the revised version, in this and all other calculations of the article we remove the five first days in the simulation in order to limit possible artefacts due to the spinup time. Therefore, all results are now presented from Jun. 6 to July 15 instead of June 1 to July 15.

**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 to a higher RH, and thus the scattering growth factor neglected in this study ?**

As indicated above, the initial indication that the growth factor was neglected was a mistake (which has been corrected). Actually, the growth factor is taken into account for the main hygroscopic anthropogenic species (sulphates, nitrates, ammonium). Therefore, we are not able to provide a convincing explanation for this underestimation, which also appears in the AOD comparison with AERONET stations in this area (Mainz, Palaiseau), which are added in the revised version of the paper (Table 3).

The paragraph about water uptake by aerosols is now as follows :

« As in Bian and Zender (2003), we chose to neglect the influence of relative humidity on the optical properties of mineral dust, which has been shown to have a very small effect on the volume of dust particles (Herich et al., 2009). However, water uptake by hygroscopic species such as nitrates, sulphates and ammonium in subsaturated conditions is represented using the ISORROPIA module (Nenes et al, 1998), as described in Bessagnet et al. (2004). The optical effect of the liquid-phase water generated by the hygroscopic growth of these aerosols is taken into account by the Fast-JX module as a separate aerosol species with the optical characteristics of water. »

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

We are not able to interpret this fact. Many reasons exist why a peak should be missed by the model, and we unfortunately do not have the necessary data to interpret 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.**

A quantitative comparison with correlation coefficients, standard deviations in the models and observations, correlation coefficients and their significance (Table 3) has been added in the revised manuscript for 12 stations, giving more quantitiveness and representativeness to the rather qualitative discussion of the initial manuscript. This discussion is included in Subsection 3.1.1 of the revised manuscript.

**Sect. 3.1.3 : I don't really understand the purpose of the comparison to the measured**

*concentrations to the second model level. WRF model is terrain-following, suggesting that the Lampedusa site is located in the first model layer. Are the altitudes given in this section in meters a.g.l. Or in meters a.m.s.l. ? This is confusing.*

In the initial version of the article, the comparison was performed at the first model level, considering that the model is terrain-following. However, since the island of Lampedusa is subgrid-scale in our model configuration, it cannot be considered that the model is terrain-following above the measurement station. Therefore, following a suggestion of the Editor, we chose to perform the comparison here at the second model level. However, we have checked that, since both these model levels are included inside the PBL, differences between these two model levels are small compared to the simulated values (and to their error compared to measured values)

**p. 7605 : Where does the number of 5  $\mu\text{g}/\text{m}^3$  come from ? Figure 7 indicates a mean overestimation of 25  $\mu\text{g}/\text{m}^3$ .**

Yes, this was a typo, we thank the Reviewer for his careful attention in detecting this bad mistake !

**p. 7605 : the authors need to be more 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, this is not always true for the bulk mass of aerosols, as its chemical composition may vary. 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, whereas AOD is 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. Does it indicate that numerical diffusion is better above the surface ?**

We completely agree with the Reviewer that this paragraph is in part overstated, particularly the sentence « The fact that the AOD in Lampedusa as well as other stations is represented in a realistic way by the model (Fig. 5a) is an indication that the total aerosol loads represented by CHIMERE is realistic ». Apart from the reasons listed by the Reviewer, the uncertainty on aerosol size distribution is also a critical factor.

Therefore, we removed the sentence cited above in quotes, which we agree was rather speculative. This speculative sentence is replaced by a quantitative discussion on the statistical scores for comparison between observed and modelled AOD based on Tab. 3.

**p. 7605 : Why are there different backtrajectories in Fig. 9 ? This has not been described earlier (...)**

We added the following paragraph in the revised manuscript, which also states explicitly that the simplified model used here is not necessarily comparable to state-of-the-art model with more complete formulations :

« Particles launched at the same initial position can have distinct evolutions back in time in time : therefore, the initial sample of 100 particles have distinct backtrajectories depending on their random vertical movements inside the convective boundary layer, and their partly random vertical movements within the free troposphere. Even though this backplume model is possibly not comparable to state-of-the-art models such as HYSPLIT or FLEXPART, this model has been chosen for its simplicity of use, for a study in which backtrajectories are not a critical part. It does not necessarily imply that such a simplified formulation would be adequate for studies in which accuracy of the backplume simulations is critical. ». This states explicitly the possible limitations of

the model we used, and explains a bit further the way it works.

**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 NO<sub>2</sub> dissociation.**

The larger bias in the simulation without aerosols (or, better said, without their radiative effects), NA is attributable to the radiative effects of the aerosols. The difference between the bias in the NA simulation (12.3%) and in the REF simulation (8.2%) is attributable to the radiative effect of aerosols because the radiative effect of aerosols is the only difference between these two simulations.

Therefore, the effect of the aerosols explains an average reduction of about 4% in the value of  $j(\text{NO}_2)$  above Lampedusa during the simulated period. This average effect of 4% cannot be linked to the slight overestimation of AOD at Lampedusa : this overestimation of the AOD would only explain a corresponding slight overestimation of the aerosol effect on  $j\text{NO}_2$  in Lampedusa.

In the revised paper (Table 3), statistical scores are provided for the simulated AOD compared to the observed values. The positive bias of the simulated AOD is 17.9%, so that we can estimate that the reduction of 4% in  $j\text{NO}_2$  in the REF simulation compared to the NA simulation is possibly overestimated by  $17.9\% \times 4\% = 0,7\%$  . Therefore, the effect of the error in modelled values of the AOD at Lampedusa is relatively small compared to the total effect of the aerosols.

To clarify the interpretation of these figures, the following words have been added to the manuscript (in green) :

« Two observations can be made from Fig.10a. First, that the values of diurnal maxima of  $j(\text{NO}_2)$  in both simulations are positively biased. This bias is of 12.3% for the simulation without aerosols (NA), and 8.2% in the reference simulation, so that, in average during the simulation period, the direct radiative effect of the aerosol reduced the daily of  $j(\text{NO}_2)$  by about 4%

**p. 7608, l. 8-9 : The good correlation between modeled and observed JNO<sub>2</sub> values can hardly be linked to the optical properties of aerosols. Fin Fig. 11a for instance, the impact of including or not the aerosols is very weak because the AOD itself is not significant (~ 0.1). Aerosols have only a noticeable impact on JNO<sub>2</sub> when the aerosol loads are important. The only variations on the black 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 JNO<sub>2</sub>**

In Fig. 11a, from a qualitative point of view, the effect of the aerosols on  $j\text{NO}_2$  is significant from June 6 to June 10, from June 20 to June 24, and July 3 to July 5, and more weakly from July 13 to 15, corresponding to AOD values exceeding 0.2.

We do not agree with the statement that « The good correlation between modeled and observed JNO<sub>2</sub> values can hardly be linked to the optical properties of aerosols ». The only difference between the NA and the REF simulation is precisely the inclusion of the radiative effect of the aerosols in the latter one. Therefore, the spectacular increase in the correlation coefficient from the NA simulation ( $R=-0.05$ ) to the REF simulation ( $R=0.92$ ) can be attributed exclusively to the radiative effect of the aerosols. It is however true that this effect is significant mostly when the AOD is significant ( $> 0.2$ ), so that this very high correlation rate is essentially due to the effect of (relatively) strong AOD.



To take into account this observation in the revised manuscript without putting too much emphasis on the threshold of qualitative threshold  $AOD \sim 0.2$  (which is rather arbitrary since the relationship between AOD and photolysis rates is essentially linear - Fig 14), we added the following sentence into the manuscript : Comparison between Figs. 12a and 6a shows that this effect is substantial only when the AOD reaches or exceeds values around 0.2.

**Sect. 3.2.3 : According to the 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  $JNO_2=f(AOD)$  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. 2013).**

Our simulation does not include cases of strong AOD due to non-dust aerosols. Therefore, we are not able to perform a useful separation between dust and non-dust cases, unlike Gerasopoulos (2012).

To clarify this point, we added the following sentence into the revised manuscript :

« It is worth noting at this point that, during our simulation period, no significant AOD peaks have been simulated due to non-dust aerosols, so that the scatter plot obtained in the REF simulation (Fig. 13b) shall be compared to the red regression line given by Gerasopoulos et al. (2012) for cases when dust predominates rather than to the blue regression line given for cases when non-dust aerosols predominate. »

**p. 7614, l. 4-7 : The maximum difference on O3 is 2 ppbv. What is the associated uncertainty ? Is this result robust ?**

The differences found on ozone concentrations here, about 2 ppbv in maximum, are not very substantial, and are small compared to many other causes of errors that are common in chemistry-transport models (errors in the emission inventories, in the meteorology, in transport and mixing processes, deposition, etc.). However, this difference map shows the error due to omitting the radiative effect of the aerosols, all other things equal : the errors in these processes are the same in the REF and the NA simulation, so that the residual difference between the concentrations simulated in both simulations, around 2 ppbv, is attributable with certainty to the radiative effect of the aerosols.

This result is not a critical part of the manuscript, these maps are just here to give an estimation of the possible magnitude of the aerosols on ozone concentrations through their radiative effect, and to show that these results are consistent with the results in Bian et al. (2003b).

**(...) The authors should check how robust is this result since most of the main conclusions are linked to such small discrepancies. A sensitivity test on temperature on  $JNO_2$ ,  $JO_1D$ ,  $O_3$  mixing ratios would have been very much appreciated.**

The effect of a temperature bias on ozone mixing ratios is important but we think it is beyond the scope of the present study, which is focused on the effect of aerosols on the photolysis rates of ozone and  $NO_2$ . More critical for the present study is the possible effect of temperature on  $j(NO_2)$  and  $j(O_1d)$ , which we now discuss in a new paragraph in the introduction based on existing bibliography. An estimation of the effect of temperature bias on  $j(O_1d)$  is provided, while it is

known from the literature that the effect of temperature on  $j(\text{NO}_2)$  is small. As these precisions shall be useful for the reader, we included them in the following sentence :

The temperature bias is in average of about 5K for daily temperature maxima and 3K for daily temperature minima. The impact of a 5K underestimation of daytime temperature on  $J(\text{NO}_2)$  and  $J(\text{O}1\text{d})$  photolysis rates can be estimated according to Dickerson et al. (1982). Both  $J(\text{NO}_2)$  and  $J(\text{O}1\text{d})$  values increase with temperature, but the dependancy of  $j(\text{notwo})$  on temperature is much weaker than that of  $J(\text{O}1\text{d})$ . While  $J(\text{O}1\text{d})$  increases by more than 50% when temperature increases from 273K to 307K,  $J(\text{NO}_2)$  does so by less than 5%. Therefore, the impact of a cold bias of 5K on  $J(\text{O}1\text{d})$  can generate an underestimation of 5 to 10% on  $J(\text{O}1\text{d})$ , and only about 1% on  $j(\text{notwo})$ .

**p. 7615, l. 6-12 : this sensitivity test is relevant for the paper, but its description whould 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.**

Description of this additional simulation has been moved up, and this simulation is now introduced at the same time as the two others. The paragraph « Sensitivity to a bias in total ozone column » has been modified accordingly, and the O3+ simulation has been introduced in the conclusion as well.

**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.**

Unfortunately, we are not able to give a better explanation here than in the initial manuscript. We can just state that we observed that the photolytic rates calculated by Fast-JX are closer to reality when we use the ozone climatology recommended by the model developers than when we try to use a « debiased » ozone climatology.

This is not due to a compensation between an overestimated effect of the aerosols and and underestimated ozone column. The effect of the AOD on  $j(\text{O}1\text{d})$  is about  $1 \cdot 10^{-3} \text{ s}^{-1}$  for an AOD of 0.5 (Fig. 13), while the effect of the 18% increase in the ozone column is about  $8 \cdot 10^{-3} \text{ s}^{-1}$  (Fig. 10b), eight times stronger. And this factor of 8 is obtained by examining the ratio of the effect of the **entire** aerosol column to the effect of the increased ozone column.

But the overestimation of AOD by the model is of « only » 17.9% at Lampedusa : so the order of magnitude of the effect of overestimation in AOD at Lampedusa can be estimated at about 20% of the total aerosol effect : this potential effect is then 30 times smaller that the effect of the increased ozone column.

The statement that Fast-JX seems to perform better when left with its original ozone climatology than with a « debiased » climatology is only a preliminar (and uncomfortable) finding, of interest only for the developers of CHIMERE and Fast-JX. However, the comparison between the O3+ and the REF simulation, and the very large differences in  $j(\text{O}1\text{d})$ , suggest that it is of interest for model developers to take into account the real-time variations of the ozone column, which are up to 20% from week to week, and may have a very considerable effect on simulated  $j(\text{O}1\text{d})$  values and therefore on ozone mixing rations in the troposphere.

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

events.

The following quantitative statement has been included in the conclusion :

In the case of Lampedusa, the correlation coefficient between simulated and observed AOD at 400 nm is strong (0.79), with an average positive bias of 0.04 in the simulated AOD (17.9% of the average observed value). These correlation and bias of the simulated vs observed values vary greatly depending on the measurement stations. For stations in north Africa or around the Mediterranean, the bias is generally moderate (-35% to +17,9% in the ten considered stations) and the correlation coefficients vary from -0.14 to 0.79. For the two stations that were considered in northern Europe (Palaiseau, France, and Mainz, Germany), the negative bias in the simulated values is strong (-61.7% and -45.3% respectively), with very weak correlation coefficient. **It is also of interest to note that the peak AOD values at the Lampedusa and Palma de Mallorca stations tend to be overestimated by up to 50% by the CHIMERE model during the simulation period.**

**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.**

Done accordingly, thanks.

**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...**

This has been done, thanks. However, we have not been able to find the meaning of « ADIENT ». CHIMERE is not an acronym but a non-translatable pun in French, ISORROPIA is not either (it seems to mean « equilibrium » in Greek)

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

This and other occurrences of « here have been modified. « Here » has been replaced by « in the present study » in many case throughout the manuscript.

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

Online was intended to mean « simulated within CHIMERE », which was redundant with the following of the sentence. This word was therefore useless, and suppressed.

**P 7598, l 15 : anthropogenic**

Done

**p 7598, l 22: Saharan**

Done

**P 7599, l 11 : CHIMERE realistically reproduces**

Done

**P 7599, l 24 : Capo Verde islands**

Done

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

Done

**p 7601, l 19 : steadily decreases**

Done

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

We used LIDAR in all the revised version.

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

This sentence was cut into shorter sentences in order to clarify it.

**P 7603, l 18 : display a very similar structure**

The sentence has been rewritten as follows : « Modeled profiles display a structure that is very similar to the observed one »

**P 7604, l 18 : overestimation**

done

**P 7605, l 3 : boundary**

done

**p 7605, l 10 : total**

done

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

done

**P 7606, l 11 : most**

done

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

We actually meant « as a summary »:this is now corrected.

**p 7608, l 1 : remove “that”**

done

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

Only Fig. 10 needs to be referred to at that point : Fig. 11, which shows the daily cycle of the photolysis rates, is analyzed afterwards.

**P 7610, l 15 : different**

done

**p 7610, l 24 : we examine**

done

**p 7611, l 6 : at local noon**

done

**p 7614, l 1 : The ozone concentrations**

done

**p 7615, l 8 : a sensitivity simulation identical**

done

**p 7616, l 9 : to be biased**

done

**P 7617, l 29 : the REF simulation**

done

**p 7618, l 11 : with in situ measurements**

The sentence has been rephrased (and clarified) as follows :

Regarding  $J(O_1d)$ , the comparison of our model results with the results of Casasanta et al. (2011), obtained from {in situ} measurements, seems to indicate...

**Table 1 caption : Sectional bins**

done

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

These figures should be much easier in a full page, as it should be the case in the ACP format, shall the manuscript get published in ACP.

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

done

**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.**

The dateticks have been fixed and are the same (or very similar) in all the time series throughout the article.

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

This has been corrected

**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.**

As for Fig. 4, this should be fixed in the full A4 format of the ACP publications, should this manuscript get accepted in ACP. A plot for Nox emissions has been added, as suggested, as Fig. 2 of the revised manuscript. It could not be in a 2x2 matrix here as suggested because we feel that this map of Nox emissions needed to be described much earlier in the manuscript.

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

done