Dear Editor and reviewers,

Review of paper: *Analysis of the atmospheric composition during the summer 2013 over the Mediterranean area using the CHARMEX measurements and the CHIMERE model*, by L. Menut, S. Mailler, G. Siour, B. Bessagnet, S. Turquety, G. Rea, R. Briant, M. Mallet, J. Sciare, and P. Formenti

Our answers are in blue in the text and after each reviewers remark. To summarize our changes in the manuscript, we found the reviewers mainly focused on these specific questions:

- "Add evaluation of the modelled meteorology (e.g. 2m temperature)"
A new section was added, dedicated to the surface temperature. This is now in a section more clear also for the meteorological model set-up (subsections were added).
In this new section 4 "Analysis of meteorology":
- maps of 2m temperature and precipitation amount for E-OBS data and WRF results: new Figure 6
- Time series of daily 2m temperature and precipitation and for some sites studies in the paper: new Figure 7
- Time series of 2m temperature and 10m wind speed for the ADRIMED site of Cape Corsica: Figure 8
- Scores between E-OBS and WRF for 2m temperature (correlations, RMSE and bias) and precipitation (hit rate): Tables 3 and 4.

- **"Add more aerosols optical properties evaluations."**
A section 7.3 called "Aerosols size distribution" was added in the 'aerosols analysis' part, including new comparisons between measured and modelled volume size distributions retrieved from the inversion algorithm of AERONET.

- "Add more description of the airborne measurements"
A new paragraph was added in the section 2.4 "The specific ADRIMED measurements"

- "Improve the analysis of the atmospheric composition"
The addition of the meteorological comparisons and the size distribution comparisons enables to have a more accurate analysis of the atmospheric composition evolution during this summer 2013.
The analysis of ozone concentrations was improved with a new Figure 9. Figure 11 was reprocessed to have the same colorscale.

- **"Need for more adaptated references and better figure colorscales:"**
- The proposed references were added in the text.
- Some color figures were reprocessed with a more readable colorscale.
- A new map was added to see the AERONET stations locations
- New informations were added in Table (as altitude of stations in Table 1, biases in scores tables).

In addition, we also updated the scores by homogeneizing the daily mean, daily max and hourly calculations for the different studied parameters. The text was rewritten and is now more clear. The bias was added in all scores tables.
The abstract, introduction and conclusion were rewritten to be more "an analysis of the atmospheric composition" as requested by the two reviewers.

Reviewer 1 (15 october 2014):

This manuscript, submitted in the special issue "Special Issue: CHemistry and AeRosols Mediterranean EXperiments (ChArMEx)" studies the atmospheric composition in terms of ozone concentration, particulate matter and aerosol optical depth over Europe and the Mediterranean during summer 2013. The study is based on both field campaign measurements (ChArMEx/ADRIMED) and modeling using the chemistrytransport model CHIMERE. Results show the ability of this model to

reproduce the atmospheric composition over the Mediterranean in terms of ozone concentration, particulate matter and aerosol optical depth. It is also interesting to show the contribution of the different species to the pollution over this region. However, before publication in ACP, the authors should correct several issues presented below.

Major comments:

- there is an inconsistency between the title of the paper and the objectives announced at the end of the introduction. Reading the title, I expected to find in this study a detailed analysis of the atmospheric composition over the Mediterranean during the summer 2013, rather than a successive evaluation of different parameters. The analysis of the contribution of the different species presented in section 6.2 should come earlier in the paper. However, this evaluation is also interesting as CHIMERE shows its good performance with regards to the CHARMEX measurements. Therefore, I suggest either to change the title and explicitly mention the evaluation part of the paper, or reorganize the paper to give more importance to the analysis part.

The paper was re-organized to better highlight the analysis part of the study. Meteorological data are added and analyzed in term of the main meteorological situations observed during the period, discussions about the observed and modelled atmospheric composition is more analyzed and aerosols size distributions are also added for the particles behaviour analysis. Finally, the whole text was changed with numerous new sections to really propose an analysis of this period.

- The discussion regarding the performance of the model should be longer for some parameters, and the authors tend to be too affirmative while the figures and the table reveal some discrepancies between the model and the observations. For example, I would appreciate to have more explanations on the differences in AOD between MODIS and CHIMERE (notably in the Atlantic ocean, in the Middle-East, near the Caspian Sea), on the negative correlations in the daily mean PM10, and on the discrepancies in the measurements of Cape Corsica (Figure 14). These deficiencies of the model should also be mentioned more clearly in the abstract.

The discussion about MODIS and CHIMERE was extended. But this is not the main goal of this paper to have a detailed analysis of the MODIS data: these data were just used here to have an overall view of the main aerosols contents during the whole studied period.
For the PM10 model deficiencies, more details are given to explain why some sites have very high correlations and others have negative correlations. This is helped by the addition of the new section dedicated to the aerosols size distributions.
For the Cape Corsica site, the discussion was also changed to point out the fact that this site is difficult to compare to model results with a low horizontal resolution. A discussion about the model and observations representativities was added. But, please note that it was already mentioned in the original abstract: *"A comparison with sulphate concentrations at Cape Corsica exhibits some discrepancies related to the regridding of shipping emissions."*

- With regards to the aerosol part of the paper, other variables than AOD could also have been evaluated. I think that the ADRIMED field campaign has provided measurements of the vertical aerosol distribution, optical properties (single scattering albedo, asymmetry parameter), or size distribution, which could reinforce the quality of the evaluation presented in the paper.

The whole set of measurements performed during the ADRIMED campaign will be presented in a paper led by the project PI, Marc Mallet. Our goal here is not to evaluate the model against all measurements done, but to focus on the capability of our modelling system, WRF-CHIMERE, to catch the main gas and aerosols concentration behaviour during this period. The title of the paper was changed to be more on this specific topic.
About aerosols, we added a new section for comparison and discussion on the aerosol volume size

distribution with AERONET measurements.

Specific comments :

- Page 23078 L13-21: Other studies (Moulin et al., 1998, Middleton and Goudie, 2001; Israelevich et al., 2012) could be mentioned in this paragraph, to explain the transport of dust aerosols over the Mediterranean basin.

OK, these important references were added in the new version.

- In the introduction, the use of regional climate models such as COSMO (Vogel et al., 2009), RegCM (Santese et al., 2010), SKYRON (Spyrou et al., 2013) and ALADINClimate (Nabat et al., 2014) in order to quantify the aerosols over the Mediterranean and estimate their impact on regional climate should be mentioned. Such field campaigns like ADRIMED devoted in studying aerosol properties are important to improve the representation of aerosols in the climate models.

We agree and the introduction was also improved following this suggestion.

- Page 23081 L12-21: Is it possible to compare this data on vegetation fire events to a climatology ? Thus the authors could justify the last sentence of this paragraph (L20-21).

This kind of climatology was already presented in [Turquety et al., 2014]. This is why the article is cited but to present again the climatology is not necessary. This paper has no climatological aspect but is focused on hourly processes over a few weeks.

- Page 23081 L24-27 : It is true that meteorological variables are generally measured country by country, but there has been an effort in the ChArMEx and HyMEx programmes to gather datasets (http://mistrals.sedoo.fr). The authors could consider adding for example an evaluation of 2m temperature.

Yes, we agree with this suggestion and this was added in the paper in a new section, dedicated to comparison with meteorological measurements.

- Page 23082 L8-10: The resolution (50km x 50km) can indeed raise the question of the representativeness of the model compared to measurements, but I do not understand why ozone can be evaluated and not NO2 ?

Yes, the sentence was not very clear. In fact, the choice is due to the fact that we can not evaluate all existing pollutants. And regarding our model resolution, we consider as a priority to evaluate ozone and aerosols. NO2 measurements are more 'local' measurements compared to observations in stations which are more 'urban' or 'suburban'. And this 'small scale' representativity is not adapted to our model resolution. The main goal of this paper is to estimate the main pathways of pollutants over a large domain, including several different sources such as anthropogenic, biogenic, dust and fires. We thus are limited to comparisons with pollutants having a several days lifetime. The text was clarified and simplified accordingly.

- Page 23083 L2-6: The instrumentation regarding the ATR-42 and the Falcon-20 should be presented in section 2.3.

This very precise presentation will be also made in the ADRIMED overview paper of Marc Mallet. We added a short presentation of the instrumentation..

- Page 23083 L15-16: Why use only the MODIS instrument ? It could be interested to have also other

instruments such as MISR or SEVIRI. Notably the use of instruments onboard geostationary satellites enable to have a better spatial and temporal resolution.

In this study, MODIS was used only to estimate if the model is able to retrieve the observed aerosol optical depth over the whole domain. This is mainly a 'preliminary' presentation of our results but not the main goal of the paper. Our goal is more to select 'in situ' measurements rather than satellite measurements which deserve a complete independent study.

- Page 23084 L6-7: Could you give more details on the method to build an unified AOD product, including the two MODIS sensors, as well as the Deep Blue algorithm ?

In fact, we are directly downloading the deep-blue data on the NASA Giovanni web site. We added this reference in the text. All details about this product are in the cited reference: [Hsu et al., 2004]. To avoid confusion, we move the description of the MODIS data in the section where the results are presented. And since our goal is not to make an extensive comparison between model and satellite (this will be done in companion papers dedicated to this topic), the text was also simplified.

- Page 23085 L14: It is not clear if you use forecast or analyses for the global meteorological fields ? Could you also give more details on the WRF simulations ?

The WRF configuration and the simulations are described in pages 23084 and 23085. For the forecast or analysis used, this is here analysis.

- Page 23086 (Section 3.2) : Are there gaseous and aerosol species at the boundaries of the domain ? Further in the text (Page 23089, L19) you mention a global climate model for the boundary chemical fields. Could you clarify this point ?

Yes, this was not enough described. New sentences were added to clearly explained the boundary conditions we used. Gaseous and aerosols species are prescribed at the boundaries, using global model monthly climatologies. Note also that all these informations about the CHIMERE model version used are extensively described in [Menut et al., 2013, GMD].

- Table 3: The average bias should be added in the table.

Yes, this is correct and the bias values were added in all the tables scores.

- Table 4 / Page 23088 L4: Are the correlation and RMSE calculated from daily mean values as indicated in the text of from hourly values as indicated in the caption of Table 4 ?

OK this needs to be clarified. Some sentences were added in the text for that. Some explanations were already p.23088. The Table 3 is for daily maximum values and the table 4 for daily mean values. These two results are complementary.

- Page 23088 L17-18: Could you give examples of this contrast between Ajaccio and Bastia ? It does not seem obvious for me, notably between 6 and 11 July.

Yes, we added more explanations. This is right that for this time interval, the model underestimates the measured surface ozone concentrations for the two sites. The sentence of under- and over-estimation was general and about the mean value during the whole studied period.

- Page 23089 L12-13: Is this due to spectral nudging ?

Yes, certainly. This is more discussed in the new 'meteorological' section.

- Page 23089 L16: What are “trends” for you ? This should be clarified.

Yes, this is not 'trend' as in a climatological definition. This is more 'tendency' in this case and the sentence was corrected.

- Page 23089 section 4.2: The calculation of statistical scores could help the reader to judge the performance of the model.

OK this was added in a new table.

- Page 23090 L1-2: Please remove the “s” to “Aerosol Optical Depth”.

OK, done.

- Page 23090 L20: I think that the highest AOD values are rather recorded over the Sahara and the Middle-East.

This is right, the sentence was not correct: we want here to describe the most important differences and not the most important absolute values. This was corrected. And sentence about the absolute values was added.

- Page 23090 L23: There are however some discrepancies between MODIS and CHIMERE (see my remark in major comments).

Yes, some explanations were added.

- Page 23090 L25: What is ?

Lambda and phi are common notations for the longitude and latitude. The word 'latitude' was added in the sentence.

- Page 23091 L2-3: Could you give information on the location of the AERONET stations (with a map for instance) ?

Yes a second map was added with Figure 3, for AERONET stations. We are obliged to make two maps if we want that all sites are readable on the plot.

- Page 23091 L9-10: This affirmation should be moderated, as Figure 10 clearly shows differences between observations and CHIMERE.

Yes, we rewrite the sentences (as well as the conclusion) to be more realistic.

- Page 23093 L8-9: Could you justify the overestimation of the vertical diffusion in the models ?

Yes, this is a numerical problem due to the vertical transport schemes used. An explanation and references were added to explain this point.

- Page 23093 L14-15: Why not present in another figure the distribution between aerosol species only for PM10 ? It would enable to justify more precisely the differences between the model and the observations.

New figures, scores and analysis was added specifically for PM10, including size distribution.

- Figure 4: Could you indicate the flight numbers ?

We tried and the figure was not easily readable. This is why we add the period on the horizontal flights and the correspondence between period and flight number in Table 2.

- Figure 8: The caption should be detailed. Altitude is notably missing.

Yes, the caption was rewritten.

- Figure 9: The color scale is not really adapted over Europe and the Mediterranean. I think that the yellow colors ranging from 0.1 to 0.4 could mask local maxima. Could you modify the color scale ?

Yes, the figure was reprocessed to have another colorscale.

References:

Israelevich, P., Ganor, E., Alpert, P., Kishcha, P. and Stupp, A. (2012). Predominant transport paths of Saharan dust over the Mediterranean sea to Europe. J. Geophys. Res., 117 : D02205, 2012, doi : 10.1029/2011JD016482.

Middleton, N.J. and Goudie, A.S. (2001) : Saharan dust : sources and trajectories. Transactions of the Institute of British Geographers, 26: 165–181, doi: 10.1111/1475- 5661.00013.

Moulin, C., Lambert, C. E., Dayan, U., Masson, V., Ramonet, M., Bousquet, P., Legrand, M., Balkanski, Y. J., Guelle, W., Marticorena, B., Bergametti, G., and Dulac, F. (1998): Satellite climatology of African dust transport in the Mediterranean atmosphere, J. Geophys. Res., 103, 13137–13144, doi:10.1029/98JD00171.

Nabat, P., Somot, S., Mallet, M., Sevault, F., Chiacchio, M.Â˘aand Wild, M.Â˘a(2014), Direct and semi direct aerosol radiative effect on the Mediterranean climate variability using a coupled Regional Climate System Model Climate dynamics, in press, DOI:10.1007/s00382-014-2205-6

Santese M, Perrone MR, Zakey AS, Tomasi FD, Giorgi F (2010), Modeling of saharan dust outbreaks over the Mediterranean by RegCM3: Case studies. Atmos Chem Phys, 10:133–156.

Spyrou C, Kallos G, Mitsakou C, Athanasiadis P, Kalogeri C, Iacono M (2013) Modeling the radiative effects of desert dust on weather and regional climate. Atmos Chem Phys, 13:5489–5504. doi:10.5194/acp-13-5489-2013.

Vogel B, Vogel H, Baumer D, Bangert M, Lundgren K, Rinke R, Stanelle T (2009) The comprehensive model system COSMO-ART radiative impact of aerosol on the state of the atmosphere on the regional scale. Atmos Chem Phys, 9:8661–8680. doi:10.5194/acp-9-8661-2009.

Reviewer 2 (3 october 2014):

This paper analyzes the aerosols and ozone variability over the Mediterranean area during the CHARMEX measurements period. The methodology is based on the comparison between CHIMERE simulations and different sets of observations. The topic is very interesting. Unfortunately, the model-to-data comparison is too briefly discussed.

Many text was added to improve the model to data comparisons and the analysis of the whole summer pollution.

Also, the good CHIMERE performance is overstated throughout the paper while taking a look at tables and figures gives evidence of clear model deficiencies. I encourage the authors to rephrase the discussion in a more moderate way and to include a more indepth analysis of model uncertainties.

We agree and the discussion is now more moderated.

For example, a comparison of simulations with aerosol size distribution and single scattering albedo from AERONET measurements should be added.

The aerosol size distribution is a sensitive point in this type of modeling. We added comparisons to data with AERONET and a discussion about the results and the strength and weakness of the model about this point.

As CHIMERE biases could also come from uncertainties in the WRF simulations, the evaluation of simulated meteorological parameters should be maybe completed by using a large set of observations from the European Climate Gridded database that the authors used in a previous paper (Menut et al. 2013).

Yes, this is right that the meteorology is also a large part of the uncertainty in this modeling system. We added comparisons between meteorological measurements and model.

Please find below my **specific comments:**

- Page 23079 L 4-5: Please justify
The introduction was completely rewritten to be more clear.

- Page 23079 L 18-20: Give some examples of results
The introduction was completely rewritten to be more clear.

- Page 23080 L 4: Which developments ?
This is fully explained a few lines later in the subsection dedicated to the CHIMERE model description.

- Page 23080 L 15: What were the performances of WRF-CHIMERE compared to the other models ?
At this time, there is no evaluation of the simulations made during the field campaign. But this may be a future interesting study in term of previsibility.

- Page 23082 L9: Why the comparison is relevant for ozone but not for NO2 ?
Please see answer to reviewer#1.

- Page 23084 L 8: The study of Levy et al. 2010 is only for data over land.
Yes, we agree. This is what we wrote at line 3 of the same paragraph. To avoid confusion, we rewrite this part of the text.

- Page 23086 L 25-27: Do aerosols have an influence on the modelled photolysis rates?
Yes, by attenuating the direct radiative flux and thus limiting the photolysis of ozone. This is specifically studied in a companion paper by [Mailler et al.] for the same Charmex special section in ACP.

- Page 23089 L 17-21: Biases are clearly seen also near the surface.
Yes, we just want to say "more underestimated in altitude". But, this is right that that the over-estimation is also visible near the surface. The sentence was rewritten accordingly.

- Page 23090 L 10: The sensitivity depends also on the wavelength
This was added in the text.

- Page 23090 section 5.1: a statistical comparison between MODIS and CHIMERE should be more precise
This is difficult to add a statistical comparison because we use time averaged maps. But following this remark, and the one of the reviewer#1, we added more text to discuss the differences between the model and the data. But, in this paper, our goal was not to analyze averaged satellite data but more surface and airborne hourly measurements.

- Page 23091 L 10: For most of the time, the modelled AOD is not very close to the measurements.
This is right and the discussion is now more moderated.

- Page 23091 L 23-24: authors mention a possible bias in the modelled aerosol size distribution. A comparison with AERONET aerosol size distribution over available site should be added.
We agree and a new section was added in the manuscript.

- Page 23091 L 26-27: Uncertainties in measured AOD are low (0.01 see Holben et al. 2001) compared to model biases.
This was corrected.

- Page 23092 L24: "[...] the bias remains low [...]": I disagree. A bias up to 90 % (Chitignano) is not low.
That's correct and it was corrected.

Technical corrections:

- Please explicit accronyms when first use
Many acronyms explanations were added in the text.

- Page 23077 L 12: add "of" before "the atmospheric"
It was done but the abstract was also completely rewritten.

- explicit BL and FT on Figure 1
BL and FT were explicited in the Figure. But we also added their definitions in the caption.

Bibliography:

- Menut L., O.P.Tripathi, A.Colette, R.Vautard, E.Flaounas, B.Bessagnet, 2013, Evaluation of regional climate simulations for air quality modelling purposes, Climate Dynamics, doi:10.1007/s00382-012-1345-9, volume 40, issue 9, pages 2515-2533

- B.N. Holben, D. Tanré, A. Smirnov, T.K. Eck, I. Slutsker, N. Abuhassan, W.W. Newcomb, J.S. Schafer, B. Chatenet, F. Lavenu, Y.J. Kaufman, J.V. Castle, A. Setzer, B. Markham, D. Clarck, R. Frouin, R. Halthore, A. Kameli, N.T. O'Neil, C. Pietras, R.T. Pinker, K. Vass, G. Zibordi: An emerging ground-based aerosol climatology: Aerosol optical depth from AERONET, Journal of Geophysical Research, 106 (2001), pp. 12067–12097