We would like to thank the Editor for his careful reading of this manuscript, and the associated remarks. The Editor's remarks are in bold and the answer are written after each point.

Answers to Referee 1:

9. The bias in AOD is much smaller compared to bias for the surface stations. Can the authors discuss the reason for this?

Please discuss this point in the paper, and try to give a more specific response than that given to referee one.

As already said, the AOD and the surface stations do not represent the same quantities. The AOD is a measure of the integrated column of aerosol quantities, while surface stations evaluate the aerosol concentrations at the surface only. Moreover, MODIS data cover a large area, while surface stations are limited to certain locations. When comparing to AERONET stations, the AOD is also representative of a limited amount of locations, but that are very different from the surface stations ones.

Moreover, there can be a good agreement with AODs while not with surface stations if the vertical distribution is not fully right. Also, the size distribution can affect the AOD computation, which is sensitive to the aerosol size, while the PM10 indicator is less sensitive to this aspect as long as the aerosols are smaller than 10 microns.

The manuscript has been updated in section 3.3 to add this aspect.

Answers to Referee 2: 1. How the results of the present study compare with these earlier studies?

P10,l32: Please name the differences in sulfate you make reference to.

The manuscript has been changed to:

Wet deposition is the main sink in Aan de Brugh et al. (2011) for sulphate, but sedimentation and dry deposition are the main sinks in this study. This can be explained by the difference of the simulated domains in both studies. The domain in our study is more southern and has high sulphate aerosol concentrations in the eastern part of the basin associated with less precipitation.

2. Also in all tables and throughout the manuscript where reference is made to budget, it has to be clarified they concerne PM10 aerosol (or is it bulk aerosol in the model?)

Could authors please add this information (on use of bulk aerosol as a target) also in Figure 7 and Tables 12 and 13.

The information has been added in figures 7 and 8 and tables 10 to 17.

I would have prepared having budget calculations for PM10, which is more easy to reference. Could authors please discuss qualitatively, by looking at size distributions for PM species, how results would be different for PM10.

Indeed, it would be interesting to have the same budget for PM10 if considering the air quality aspect. Nevertheless, the total mass aerosol budget is also interesting for domain such as visibility, or nutrient deposition. The simulation was set to compute the chemical production and destruction, sedimentation and dry deposition, wet deposition and burden of the total mass of aerosols. It would require to rerun the simulation by changing the outputs in order to get the budget over the PM10. Moreover, it would be very hazardous to extrapolate results to PM10.

4. Section 2.1 MOCAGE model Information is missing on how the aerosol size is parametrized in the model. Since later in the discussion the authors refer to the aerosol sizes are taken into account in the model and how they change during atmospheric aging.

Authors say, that there is no effect of ageing on aerosol size in the model. Certainly, there are many processes in the model which act on aerosol size over time, such as condensation or sedimentation. May be authors want to say that effects of these processes on size distribution are small, but even this would be surprising.

There has been a misunderstanding from our side. Indeed, by aging we were thinking of the processes that occur to a single aerosol particle, such as coagulation (this process is not explicitly represented), reaction with gas, etc. But, indeed deposition and chemical production (for secondary aerosols), and emission (for primary aerosols) processes affect the aerosol size distributions. The manuscript has been changed to be more precise on this point.

21. Page 9, line 15-18: Nitrate aerosol partitioning to the aerosol phase is very sensitive to the aerosol pH (Guo et al., 2016) and NH4NO3 is semi volatile. How well is this computed in your model? Do you consider dust aerosol alkaline components in ISORROPIA calculations?

Please discuss this point also in the revised manuscript. How much more nitrate would you expect, when taking into account alkaline compounds from dust ?

For Pacific Ocean, Fairlie et al. (2010) address this problem and give many references on other relevant studies.

Hodzic et al. (2006) showed that the inclusion of dust related alkaline components improves the behaviour of the model, over Europe, by reducing the PM10 bias of 6 to 10% when comparing results to selected EMEP stations. Moreover, Ansari and Pandis, (1999) showed the bias of the total nitrate aerosol is reduced by about 20% when including crustal species into the computation. Nevertheless, Fairlie et al., (2010) pointed out that the uptake coefficient usually used are too important. Hence the previous numbers might overestimated.

The manuscript has been modified to add this point. Moreover, the work to include the alkaline component in the model MOCAGE is ongoing.

Hodzic, A., Bessagnet, B., & Vautard, R. (2006). A model evaluation of coarse-mode nitrate heterogeneous formation on dust particles. *Atmospheric environment*, *40*(22), 4158-4171.

Asif S. Ansari & Spyros N. Pandis (1999). An Analysis of Four Models Predicting the Partitioning of Semivolatile Inorganic Aerosol Components, Aerosol Science and Technology, 31:2-3, 129-153, DOI: <u>10.1080/027868299304200</u>

I am quite convinced that ChArMEx provides data on alkaline compounds, or likewise data on coarse mode nitrate which are probably bound to such ions.

On the Charmex website, the few listed speciation data such as:

http://mistrals.sedoo.fr/?editDatsId=1324&datsId=1324&project_name=ChArMEx

or

http://mistrals.sedoo.fr/?editDatsId=2&datsId=2&project_name=ChArMEx

are either not available, or not during the Charmex period (2012-2014).

Answers to Referee 3:

1. The authors should make more clear the connection to the ChArMEx and the choice of the years, since they have not at all made any use of the campaign's data.

Even if 2012 and 2013 PM composition data from Cape Corsica are indeed not in the SEDOO

data base, I think they would be available from the PI (Jean Sciare), if needed. PM1 composition data from SOP2 (July, August 2013) are available and published, for example in Michoud et al., 2017. I recommend that authors use at least these SOP2 data for model evaluation.

I send at least 5 emails to Jean Sciare to get the data, but unfortunately I never got any answer.