Responses to referee#2

We thank Referee #2 for his/her useful comments. Each response to the referee's comments is organized as follows: (1) comments from Referee in bold, (2) author’s response and author’s changes to the manuscript in normal font. The changes in the revised manuscript, except the small edit corrections, are highlighted in green color in the revised manuscript. Note that following referee#1's comment, the revised manuscript has been edited by an English speaker to improve its phrasing and grammar.

Major comments:

1) My main concerns are about the representation of distribution between bins of particulate concentrations (given by Isorropia) between bins and the absence of coagulation in the code.
- I was quite surprised that the first bin of aerosol ranged from 2 to 10 nm whereas nucleation is not represented and there is no mention of nucleation in the text. How can coagulation be neglected when there is a bin constituted by nanoparticles? Those particles are expected to coagulate very quickly with bigger particles.
- I think there is confusion between the fine mode and the accumulation mode. The two expressions are used in the text. According to Seinfeld and Pandis, the accumulation mode covers particles between 0.1 and 2.5µm. The fine mode cover particle with a diameter lower than 2.5µm and is constituted by the accumulation mode and the nuclei mode (for particle lower than 0.1µm). I guess that the author used fine mode instead of nuclei mode or the sentences like “we implicitly assumed that the coagulation has already been acting to transform fine model aerosols into accumulation mode aerosols” does not have any sense. But again, in that case, why are there bins between 2 and 100 nm?
- I find the fact to not represent coagulation (with almost no explanation) quite difficult to digest. As coagulation impact strongly the size of aerosol and therefore the deposition rate of aerosols, it is expected to impact strongly aerosol formation. More justification and references should at least be added in the text.

As a first approach for introducing SIA into MOCAGE, the choice made here was to assume that each type of aerosols is distributed into its bins all along its lifetime following the defined modes (and associated parameters) based on observations published in the literature. By doing this, we assume that the defined modes already include all the aerosol microphysical processes implicitly. This is why nucleation and coagulation are not explicitly in the model. This is a simple approach which has the advantage of requiring low computation time. The next steps will be to improve this formation part by working on the microphysical processes (nucleation, coagulation, etc) and will use the smallest bin. This is made clear in the revised version.
Concerning the specific remark on the confusion between the fine mode and the nuclei mode, the referee is right and we thank them for this remark. This is the nuclei mode that is not used in our study. The text has been corrected.

- When sea salts are taken into account in ISORROPIA, the model replaces all the Cl by NO3 (if concentrations of HNO3 are sufficient) because HNO3 is less volatile and a stronger acid than Hcl. However, Cl in sea salt is mainly present in coarse particle for which the assumption of equilibrium can be too strong because the kinetic of condensation/evaporation of HCl on coarse particles can be very slow and that it can be best to not take into account sea salt into an

inorganic equilibrium model. Have the authors look into that matter? It would have been best to add comparisons of modeled concentrations of Na and Cl to measurements to know if there is too much evaporation of HCl or not.

When using ISORROPIA, sea salts are taken into account for the calculation of the equilibrium. The nitric acid is allowed to be transferred in the aerosol phase but the chlorine in not transferred in the gas phase because the chlorine chemistry is not present yet in the model. Future work will include a better representation of the exchange between the aerosol and the gas phase, by using a method taking into account the kinetic of transfer between the two phases at least for big particles. Another planned work is to implement chlorine chemistry in order to use HCl evaporated from sea salt. This is now stated in the conclusion section as one of the next developments.

2) The manuscript lacks of description of the parameterization used in MOCAGE which could be critical for the understanding of the results of the model. This lacks of details make the paper difficult to read, especially if the reader doesn't know very well MOCAGE. Often, the manuscript cites other papers but does not describe the processes. It would be great to modify the manuscript so that the reader can have a good overview of all the processes impacting aerosol formation without having to look inside other papers as this manuscript could be one of the reference papers for the aerosols in MOCAGE. Maybe, the authors should present every of the main phenomena affecting aerosol formations with one part for each phenomena whether than presenting the old MOCAGE (and only referring to past studies) and separately the modifications.

Following the referee's suggestion, we have added description of the parameterizations used in MOCAGE and in particular for aerosol-related processes. Please note that the nucleation and coagulation are not described because they are not explicitly represented in the model.

- There is almost no mention in the paper of aqueous-phase chemistry except for some references of past studies which is one of the pathways of oxidation of SO2 into H2SO4. How aqueous-phase is taken into account? The authors mention (lines 3-10, page 3620) that pH and liquid water content can be sources of uncertainties. Therefore, the manuscript should be modified to clearly present aqueous-phase chemistry, how it is taken into account, how the cloud pH is computed, etc.

Note: Referee#3 asked a similar question. The liquid phase oxidation is taken into account following Pham et al. (1995) and Boucher et al. (2002). It takes into account the oxidation of SO2 by H2O2 and O3 after dissolving the necessary gases into the liquid water content. Liquid water content is a variable of the model extracted from analysed meteorological forcing fields. The pH is supposed to always be equal to 5 (Charlson and Rodhe, 1982). Details about aqueous phase chemistry and SO2 oxidation are given in Section 2.2.1 in the revised manuscript.

- For depositon, the authors cite, in section 2.3.1, a paper of Sic et al al. (2005) where they check the parameterization and propose improvement. But, the main conclusions are not cited.
The manuscript has been modified to include the main conclusions of Sic et al. (2015) as suggested by the referee.

Deposition is only described in section 2.4 without giving any formula to summarize how this process is represented. Deposition should be presented clearly inside one paragraph. What about deposition of gaseous precursors? How is deposition of SO2, NO2, NH3 and HNO3 taken into account? The authors mention sedimentation incorporating hygroscopicity. What does it stands for? Does the author compute a humid diameter (taking into account the amount of water inside the aerosol) that is used for sedimentation?

In order to be more explicit on the deposition processes affecting the aerosols, section 2.4 has been splitted into 2.4.1 concerning the advection scheme, 2.4.2 concerning the gaseous compounds and 2.4.3 concerning aerosols. Section 2.4.3 describes more precisely the processes affecting the aerosols. Concerning aerosols sedimentation, the referee is right. The hygroscopicity is taken into account by computing a humid diameter. Concerning dry deposition of SO2, NO2, NH3 and HNO3 are treated like the other gaseous compounds, following Wesely (1989).

- Heterogeneous reactions and gas-phase reactions: a reaction for the hydrolysis of N2O5 is mentioned. Are there any other heterogeneous reactions taken into account? What are the reaction of formation of H2SO4 and HNO3 and their reaction rate constant (models often have different values)? A table to summarize the formation of semi-volatile inorganic compounds could be added.

The other heterogeneous reactions taken into account are the reactions of aqueous phase oxidation of SO2 by O3 and H2O2. Concerning H2SO4, please refer to one of the preceding answer. Concerning HNO3, there are two ways for its formation. The first one, through gas phase reactions, corresponds to the RACM chemical scheme (Stockwell et al., 1997). The second way of forming HNO3, through heterogeneous hydrolysis of N2O5. As suggested by the referee, a Table summarizing the formation of semi-volatile inorganic compounds and their associated reaction rate constant has been added to the revised version of the manuscript.

- Nucleation: is it taken into account? There is no mention about it in the text.

This issue has already been discussed in a previous response about coagulation.

- On emissions (page 3604): the authors mix emissions for 2000 at the global scale with emissions for 2009 over Europe and biogenic emissions for 1990 and compare to measurements for 2005. Isn't there too much inconsistency in the years of used data? Due to reductions of emissions, the emissions are quite different between 2000, 2005 and 2009. It is important to be quite precise on emissions as the authors compare to a lot of measurements to evaluate the model but these results could be biased due to the inconsistency of emissions. Furthermore, biogenic emissions are very dependent on meteorology. Can biogenic emissions estimated for 1990 really be used for a complete different year?

Note: Referee#3 asked a similar question about biogenic emissions. Therefore, the part of the response dealing with biogenic emissions is the same.

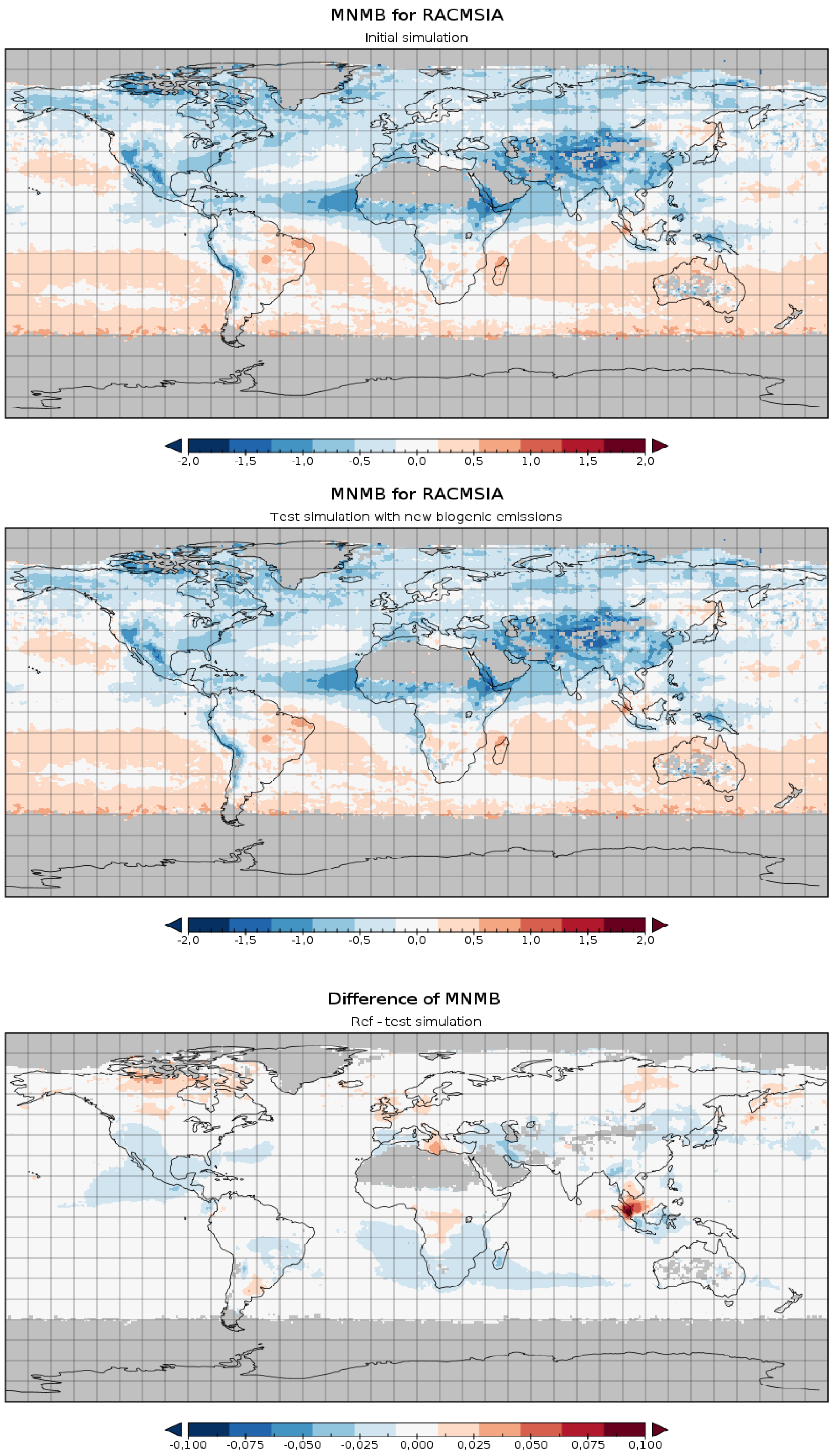

Illustration 1: Maps of Modified Normalised Mean Bias (MNMB) of aerosol optical depth against MODIS observations. The upper panel shows the RACMSIA experiment from the paper, the middle panel the test simulation realized with updated biogenic emissions and the lower panel shows the difference between the two first maps.

We agree that the emissions used here are not fully satisfying. This work started in 2013 with this old set of emissions, the new one not being available at this time. We realise now that we should have checked the availability of updated ones. Since it is not possible to entirely rerun all the experiments because of their computational cost and the time required, we have tried to address this issue in different ways. Firstly, we compared the MEGAN-MACC biogenic emissions for 2005 to those used in this study. This showed that differences are not very large. For example, when summing all emitted species it comes 972 Tg for MEGAN-MACC and 988 Tg for this study. The emissions for some species are close, like for isoprene for which the difference is about 5% (602 Tg for MEGAN-MACC and 566 Tg for this study). Other species shows more different emissions. Alkenes for example are emitted at 48,4Tg in MEGAN-MACC and 5,99Tg in this study.

Secondly we run a two months global simulation using MEGAN-MACC 2005 emissions for the biogenic part, starting on 1st of June 2005, from the simulation used in the paper. We looked at the month of July after a one month spin-up. Illustration 1 shows the MNMB of simulated AOD against MODIS observations of AOD. It shows small differences with a maximum local difference over Malaysia.

Table 1: Secondary inorganic aerosol compound statistics of RACMSIA simulation against HTAP observations. This table is the same as Table 5 in the paper but only on the period June/July.

	MNMB	FGE	Correlation
Sulfate total	-0,34	0,75	0,41
Sulfate corrected	-0,17	0,64	0,67
Nitrate	-0,31	1,13	0,32
Ammonium	0,28	0,77	0,62

Table 2: Secondary inorganic aerosol compound statistics of the test simulation using updated biogenic emissions against HTAP observations.

	MNMB	FGE	Correlation
Sulfate total	-0,39	0,78	0,4
Sulfate corrected	-0,18	0,7	0,66
Nitrate	-0,1	1,18	0,29
Ammonium	0,26	0,8	0,6

Tables 1 and 2 present the statistics of the initial RACMSIA simulation and the test simulation using updated biogenic emissions. These tables show the good coherence for sulfates and ammoniums between the two simulations using different biogenic emissions. For nitrates there is a larger difference between the two simulations which is still small. This indicates that the use of updated biogenic emissions does not change significantly the results presented in the paper. Moreover, a study on the Mediterranean basin based on the RACMSIA version of the model is ongoing and uses recent emission inventories. It is not published yet but it shows, on the Mediterranean region, similar statistical results as those discussed in this paper for Europe. This also indicates that the use of an old set of biogenic emissions does not affect the general conclusions of the paper.

Concerning 2009 emissions with 2005 measurements, the text was not clear enough. There are two separated sets of simulations. Each set of simulation is composed of one simulation noted RACM (without SIA) and one simulation noted RACMSIA (with formation of SIA). The first set of simulation is devoted to the global scale and corresponds to the simulation of the year 2005. For this set of simulation we use biogenic emissions representative of the year 1990 and anthropogenic representative of the year 2000. These simulations are compared to 2005 observations.
The second set of simulation focuses on the regional scale over Europe, while simulating the year 2010. We use for these simulations the same biogenic emissions but a regional emission inventory over Europe representative for the year 2009. These simulations are compared to 2010 observations. The paragraph concerning emissions in the text has been modified to make it clearer.

Technical comments:

Page 3600: Isorropia has two configurations (metastable and delisquescent) that can give different results (especially at low humidity). The configuration has to be added into the text. The authors compare modeled sulfate with the measurement of total sulfate and with corrected sulfate (without the contribution of sea salt) as the model simulates only the anthropogenic part of sulfate. Without changing the model, the authors could improve the comparison to measurements by assuming that there is a mass fraction of sulfate in the modeled concentrations of sea salt (7.68% according to Seinfeld and Pandis). If possible, it would be nice to compare the results of the models to other models by comparing to reported statistics in other studies.

ISORROPIA is used here within the deliquescent configuration. This information has been added in the text. We have performed the comparison asked by the referee using the sulfate fraction in the sea salts. Table 3 (below) presents the statistics obtained for Tables 5, 8, 9 and 10 in the original manuscript (Tables 6, 10, 11 and 12 in the new version). Taking into account the sulfate mass fraction from sea salt improves the bias (MNMB) against the total sulfate observations. The comparison to daily HTAP data (Table 6) shows nevertheless a decrease of the correlation coefficient and an increase of the error (FGE). The comparison with weekly HTAP data shows similar results (Table 10). Finally, when comparing the model results to daily EMEP data (Table 12), there is an improvement on both the bias and the error, the correlation being very similar. We thank the referee for this very constructive remark. We put these new results in the revised manuscript.

Table 3: Comparison of the statistics of sulfate total against MOCAGE simulation. "Initial" corresponds to the comparison with model nss and "including sss" corresponds to the use of sea salt to correct the sulfate concentrations in the model.

Table 5	MNMB	FGE	Corr
Initial	-0,32	0,79	0,52
Including sss	0,05	0,94	0,33

Table 8	MNMB	FGE	Corr
Initial	-0,1	0,68	0,66
Including sss	-0,05	0,67	0,64

Table 10	MNMB	FGE	Corr
Initial	-0,36	0,75	0,58
Including sss	-0,16	0,67	0,57