

Responses to referee#3

We thank Referee #3 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. Note that following referee #1's comment, the revised manuscript has been edited by an English speaker to improve its phrasing and grammar.

Following the suggestion of referee #2, the measurements of total sulfates are now compared to the sum of the sulfate field and 7,68% of the sea salt field of the model. This is based on the composition of sea water (Seinfeld and Pandis, 1998) in order to easily take into account the proportion of sulfates in the sea salt aerosols.

Specific comments

p.3597 l.19 give also resolution in km : have you tried the sensitivity to different resolutions? If not, what do you think the impact would be?

Approximate resolutions in km have been put in the text in addition to the latitude/longitude resolutions. We assume the question is about the sensitivity of the secondary inorganic aerosols formation versus the horizontal resolution. We have not tested the sensitivity to different resolutions at the global scale. In this paper two resolutions are used, $2^\circ \times 2^\circ$ and $0.5^\circ \times 0.5^\circ$, showing similar behavior. Moreover, this version of MOCAGE is currently used for operational forecasts with a regional domain at $0.1^\circ \times 0.1^\circ$ and an other study (not published yet) is in progress at a resolution of $0.2^\circ \times 0.2^\circ$. All these configurations show stable statistical performances against observations when using the secondary inorganic aerosols scheme.

p.3600 l.10-15 It's not clear to me how internal mixing is assumed when the species are treated separately, or are they all lumped into the different bins together? Please explain.

We agree that this part is not clear. When the partition between the gas phase and the aerosol phase is done, each aerosol type is treated using the six sectional size bins. To do this, for each different aerosol species given by ISORROPIA, we distribute the mass over these same size bins. Internal mixing is therefore assumed implicitly during this distribution. The manuscript has been modified to make this point clearer.

p.3602 l.19 what are the mass conservation properties of the scheme? Has this been looked into?

The advection scheme used in the model MOCAGE is a semi-lagrangian scheme. Semi-lagrangian advection schemes are known to not be conservative. Nevertheless, at the global scale a correction to ensure the mass conservation has been added. It can only be done at the global scale because of the lateral flux of the different regional domains. Moreover, at the scales considered here, the errors linked to the mass conservation properties of the semi-lagrangian scheme are of an order of

magnitude lower than the uncertainties of all the other processes.

p.3606 I.4 This biogenic emissions seem too old: how can it be assessed that they are still representative?

Note: Referee#2 asked a similar question, the part of the response dealing with biogenic emissions is the same.

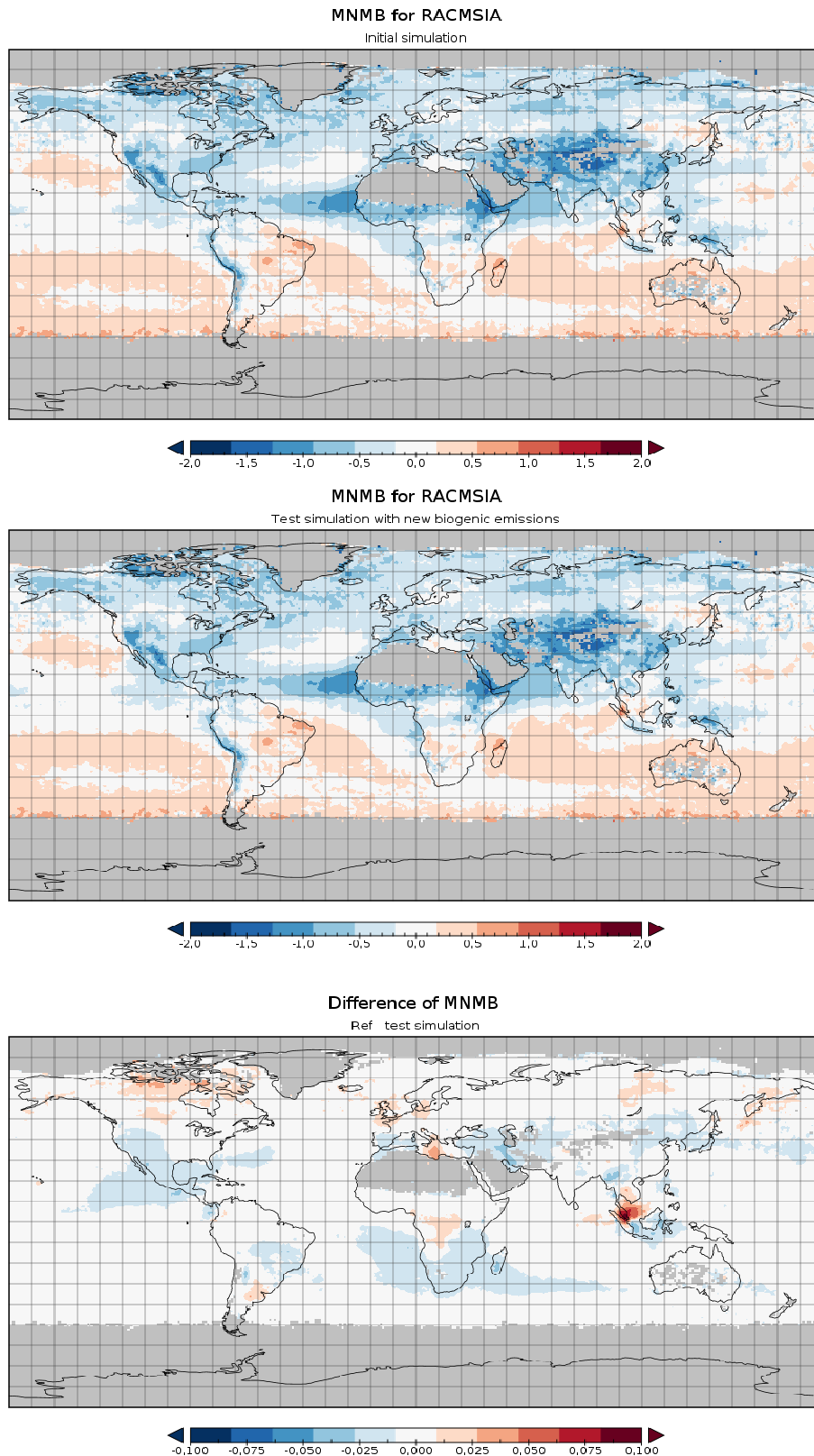


Illustration 1: Maps of Modified Normalised Mean Bias (MNMB) of aerosol optical depth against MODIS observations. The upper panel shows the RACMSIA experiment from the paper, the middle panel the test simulation realized with updated biogenic emissions and the lower panel shows the difference between 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 believe 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 show 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 month 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 long 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 ammonium 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 over the Mediterranean region it shows 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.

p.3607 I.3 In what sense misleading? Please elaborate.

The formulation is not clearly explained. Seigneur et al. (2000), explain the reasons for the use of MNMB and FGE. They state that past model performance evaluations have generally used observations to normalize the error and the bias. This approach can be misleading when the denominator is small compared to the numerator (Seigneur et al., 2000).

p.3609 I.6 The differences in figure 5 are really huge.

For ammonia, in the RACM experiment there is no chemistry. As explained in the original manuscript, ammonia is emitted, transported and deposited. In the RACMSIA experiment ammonia can be transferred in the aerosol phase leading to very large differences for NH₃ and consequently on HNO₃ between RACM and RACMSIA simulations. This point is more clearly stated in the revised manuscript.

p.3610 I.4 The bias in AOD over central Asia is also likely due to lack of adequate dust emissions as well as the lack of secondary organic aerosols. What is the positive bias over the ocean in Figure 6 due to, given the lack of DMS emissions (as I understood it)? Please elaborate.

We agree that the AOD bias over central Asia is likely not only due to the lack of SOA but also to an underestimation of the dust emissions in this region. MOCAGE includes desert dust emissions over Eastern Asia but the large uncertainties of the wind fields over this region due to complex orography produce large uncertainties on desert dust emissions.

Over ocean, there are no DMS emissions in the model indeed and therefore we expect AOD underestimation instead of overestimation. The positive bias over the ocean is likely due to the sea salt aerosols. The function used to calculate the sea salt emissions follows an exponential for big particles. These big particles remain in the atmosphere for a very short time and are very close to the surface but are often emitted in the model. The model overestimation can be linked to an uncertainty on the exponential emission function that possibly gives a too large number of big particles. Also the AOD measurements may not capture the transient presence of big particles in the very low levels. More explanations on the model AOD biases are given in the revised manuscript.

p.3611 I.7 what do you mean by "sulfate emissions combined with sea salt"?

Sea salt corresponds to a mixture of different ions, including sulfates. Referee #2 pointed out that a part of sea salt emissions is composed of sulfates (7,68% according to Seinfeld and Pandis). We agree the formulation is not very clear, it has been changed in the text.

p.3611 I.16 I was expecting to see also the RACM simulation for comparison in table 5, table 6 and figure 7. Can this be added? It would also be good to see how other stations behave (at least one more). Is the "Sulphate" in table 6 total or corrected?

The RACM simulation does not have any secondary inorganic aerosols i.e. sulfates, nitrates and ammoniums. It is then not possible to compare it the model SIA composition in RACM to the observations. An other station (CA0008R), located East of Lake Superior in Canada has been added to the text. Table 6 (Table 7 in the revised version) shows a comparison with corrected sulphate. This information has been added in the table caption and in the text.

p.3612 I.9 briefly explain how the SO₂ oxidation is included (i.e. parametrization, explicit chemistry, etc)

Note: Referee#2 asked a similar question. The SO₂ oxidation is included in the model by explicit chemistry. Gaseous oxidation by OH is a reaction from the RACM chemical scheme. The liquid phase oxidation is taken into account following Pham et al. (1995) and Boucher et al. (2002). It includes the oxidation of SO₂ by H₂O₂ and O₃ after dissolving the necessary gases into the liquid water content which is a variable of the model. A reference to the description of the SO₂ oxidation scheme (section 2.2.1) has been added in the text.

p.3613 I.4 I find it surprising that over the United States there are no daily data!

We checked again on the EBAS website (<http://ebas.nilu.no/>) and the data available on this platform for the United States are only weekly data and we have not found these data elsewhere.

p.3614 I.7 Compensation of what? Please explain.

Over Asia, the comparison to the aerosol composition shows an overestimation of secondary inorganic aerosols. The comparison of the AOD, in Fig. 6, where the stations in Japan and in the Islands south of China (see Fig. 1) are located, shows a good agreement. These good results for these AOD comparisons might therefore be due to an overestimation of secondary inorganic aerosols compensated by an underestimation linked to the lack of secondary organic aerosols. This part of text has been modified to make it clearer.

All other technical/small corrections have been taken into account in the revised manuscript.