Responses to referee#1

We thank Referee #1 for his/her useful comments. Each response to the referee's comments is organized as follows: (1) comments from the Referee in bold, (2) author's response and author's changes in manuscript in normal font. Some responses are given to several comments at the same time when these comments are related to each other. The changes in the revised manuscript, except the small edit corrections, are highlighted in green color in the revised manuscript.

Following the suggestion of the 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 part proportion of sulfates in the sea salt aerosols.

General comments

1.) The introduction includes a general overview of secondary inorganic aerosol. However, it fails to put this work in the context of other efforts to include SIA formation in global models. To demonstrate the novelty of the work described here, the following questions should be addressed in the introduction: a) What treatments of SIA are currently included in other global models? b) How is the approach described here, or the nature of the MOCAGE model, expected to provide insight into global SIA that until now was not available?

The work presented here corresponds to a representation that is fairly similar to that used in other global or regional models. The special interest of this work is linked to the specificity of the model MOCAGE which is to be able to simulate several domains at different scales via grid-nesting. The aim is here to develop a representation able to simulate SIA well at different scales from global to regional. The introduction has been modified accordingly in the revised version.

2.) In general, the paper is well organized and clear. However, there are several 'uncommon' phrasings and grammatical errors in the text, and therefore the manuscript may benefit from general (non-technical) editing. At this point the quality of the writing of the paper makes it unacceptable for publication. Authors are strongly encouraged to work with an English speaker to edit the paper.

Following this comment, the revised version of the paper has been corrected by an English speaker.

Specific comments

P3596 L7-10. It is not clear how chlorine chemistry is related to this work. If it is just to provide an example of gas-aerosol interactions, could an example more directly related to the species involved in this work be found? Or, is this an example of multiphase dynamics/chemistry that has been added to MOCAGE? If so, it should be clearly stated, and then shown how this work relates to those efforts.

We agree that this sentence was misleading. This is just an example of gas-aerosol interactions

which is not included in the model MOCAGE. In the revised manuscript, the chlorine chemistry example has been removed and a short paragraph has been added on the hydrolysis of N2O5 into HNO3 since it is taken into account in the MOCAGE model. This is a reaction happening on the sulfate aerosol surface.

P3601 L8-11. Although the need for computational efficiency is understandable, Capaldo et al. also find that aerosol nitrate concentrations are poorly represented by the equilibrium method compared to the dynamic and hybrid methods in their box model simulations (results being off by as much as a factor of four for coarse-mode PM). Nitric acid concentrations in this work have a greater FGE and lower correlation compared to HTAP observations for RACMSIA than for RACM (Table 7). Could the assumption of equilibrium be a factor in this?

P3612 L13-20. See comment on the assumption of equilibrium (P3601 L8-11).

We agree with the referee on the statement that nitrates are poorly represented by the equilibrium method, especially for the coarse mode nitrate, according to Capaldo et al.,(2000). In the paper, the authors claim the nitrate underestimation is due, at least partially, to the lack of reaction with sodium chloride which here is taken into account. In the future, it would be necessary to work on this assumption and to have a more realistic treatment of the gas/aerosol equilibrium processes. This aspect has been added in the conclusion.
When looking at the behavior of the different stations measuring nitric acid between both simulations, we can not determine a specific pattern for every station. Some stations show a better agreement with the RACM experiment while others show a better agreement with the RACMSIA experiment. One could expect the best agreement being on stations far from coastlines because of the interactions with sea salt being far from the equilibrium hypothesis. It is not a systematic result. Moreover, nitric acid is a compound which is difficult to model because there are many reactions involving nitric acid. Nevertheless, we agree with the referee that the assumption of thermodynamic equilibrium can be an additional factor explaining the nitric acid performance. This is now stated in the revised version of the manuscript.

P3602 L13-17. This paragraph should be revised. There seems to be some confusion (possibly between (NH4)2SO4 and NH4NO3?) in the first and last sentences.

The referee is right, there is a mistake in this paragraph. (NH4)2SO4 is formed from NH3 and H2SO4 and not from HNO3 and H2SO4. This has been changed in the revised manuscript.

P3604 L 23-26. What effects will the assumptions made in section 2.3.2 regarding the size distribution of SIA have on modeled AOD? Will SIA in certain size bins have more impact on modeled AOD than those in other bins.

In order to answer to this question, we made some tests on modeled AOD. Firstly, we computed the modeled AOD for a gridbox containing one aerosol type (for the example we used sulfate aerosols). A sensitivity analysis was carried out whereby the same aerosol mass concentration was distributed into each size bin in turn to observe the effect of the different aerosol sizes on AOD. These calculations show stronger AODs for the size bins located between 0.1 and 2.5 microns. In order to have further confirmation, we also computed the resulting AOD for a constant mass concentration but using different distribution modes from the literature. To do this, we used the

measured modes of Zhuang et al., (1999), which is the one used in the SIA module. We also tested modes from Hering et al., (1982) and John et al., (1990). Again the computation was made for sulfate aerosols. The Table "Tableau 2" presents the results of this computation. It shows that despite the huge differences in AOD between the different size bin, when using different realistic distribution modes, the differences in AOD are not significant.

Tableau 1: Computed AOD folowing different distribution modes using the AOD computaion framework of MOCAGE. The results are presented for sulfate aerosols.

	AOD Computed	Diff with Zhuang In pct
Zhuang et al.,(1999)	0,312	0,00%
Hering et al., (1982)	0,313	0,13%
John et al., (1990)	0,310	-0,89%

P3605 L10-13. Do the same stations that measure sulfate also measure the other species? If not, are there significant differences in the spatial distribution of station for the other species? (It might be more useful to have color-coded species measurements and include all the measurement stations in Fig. 1, as opposed to including altitude and only the sulfate stations.)

P3605 L26 – P3606 L1. Same comment as for P3605 L10-13.

All the stations are measuring sulfates, but not necessarily nitrates or ammonium. Following the referee's suggestion, Fig. 1 and 2 have been changed in order to show the measured SIA composition parameters instead of the altitude of the measuring stations.

P3610 L2-4. The remaining negative bias in the model is attributed to secondary organic aerosol (SOA) not be included in the model. Can the spatial distribution of the remaining bias be used to suggest whether or not this is the primary contribution? (Aside from the specific case mentioned in L10-13.)

The referee is right to raise this question. When looking at global distribution of SOA from Tsigaridis and Kanakidou (2003) and Heald et al. (2008), it becomes clear that the main spots of SOA concentrations are located over Asia, western Europe, eastern US and central Africa. Our results show a significant negative bias in AOD over Asia, eastern US and central Africa that can be linked to missing SOA. Both RACM and RACMSIA simulations exhibit large negative biases on the western coast of South and North America. When comparing to AEROCOM results, these biases can also be linked to dust emissions missing in MOCAGE over these regions. The manuscript has been revised accordingly.

P3611 L17-19. Why was it important to choose a rural location for this comparison?

The gridbox size of the model is quite large, approximately 220 km × 220 km at the equator, and can only be compared to stations measuring background concentrations. EMEP stations are all supposed to be measuring background concentrations but depending on their location they can be sometimes affected by urban effects. The Irish station is rural but was chosen because it is not under any direct urban influence and samples the transport of chemical species from America leading to significant concentrations of SIA. The manuscript has been changed to make this clearer.

P3617 L16. How can the improvement in MNMB and FGE be interpreted in light of the decrease in correlation for PM2.5?

We thank the referee for this comment which allowed us to detect an error in the manuscript. Indeed the correlations are inverted in Table 12 (Table 14 in the revised version). The RACMSIA simulation has a correlation of 0.58 and the RACM experiment of 0.47. The numbers were right in the text ("The correlation also rises from 0.47 to 0.58"). Table 14 in the original manuscript) has been corrected in the revised manuscript.

Technical Corrections

P3596 L2-4. This statement could use a reference.

The reference Seinfeld and Pandis, 1998 has been added to the text.

P3596 L7-8. This statement could use a reference.

The reference corresponding to this statement is Saiz-Lopez and von Glasow, 2012, it has been moved to an earlier position in the text.

P3596 L21-22. The wording of this sentence makes it sound like the SIA module has been described previously.

We agree that the sentence was not very clear. It has been changed.

P3611 L22. Is there any reason to expect Table 6 and Fig. 7 to be inconsistent? Are they not based on the same simulation data.

Indeed, Table 6 (Table 7 in the revised version) and Fig. 7 represent the same simulation compared to the same observations. The sentence highlighted here might be confusing. We wanted to show through two different ways the good results of the model against these measurements. We have removed this sentence.