

Interactive comment on “Simulating Secondary Inorganic Aerosols using the chemistry transport model MOCAGE version R2.15.0” by J. Guth et al.

Anonymous Referee #2

Received and published: 16 June 2015

This work described the treatment of Secondary Inorganic Aerosol (SIA) inside MOCAGE with a comparison of results from global and regional simulations to an extensive dataset of measurements. This paper will likely be a reference paper for the simulation of aerosols in MOCAGE. The comparison to the measurements is noticeable and provides a good insight on how well the model performs. However, some parts of the paper can be quite difficult to read because of numerous references to other studies (sometimes forcing the reader to look into these studies to have sufficient information about the model) and lack sometimes of precision. Moreover, the inconsistency of the emission dataset (data from 1990 to 2009 used to simulate the year 2005) and the distribution between bins algorithm could be problematic. These problems (detailed into more details below) must be addressed through revision before I can recommend publication in GMD.

C1143

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 2.5 μm . The fine mode cover particle with a diameter lower than 2.5 μm and is constituted by the accumulation mode and the nuclei mode (for particle lower than 0.1 μm). 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.

- 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

C1144

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.

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.

- 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. . .

- For deposition, 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. 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?

C1145

- 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.

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

Technical comments:

Page 3600: Isorropia has two configurations (metastable and deliquescent) 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.

C1146

C1147