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Interactive comment

# Interactive comment on "Development of an inorganic and organic aerosol model (Chimere2017 $\beta$ v1.0): seasonal and spatial evaluation over Europe" by Florian Couvidat et al.

### Anonymous Referee #3

Received and published: 22 August 2017

This manuscript presents an update of the CHIMERE's aerosol module. The chemical mechanism was modified for the formation of the secondary organic aerosol precursors. The equilibrium between the aerosol and the gas phase is then treated using the module SOAP. For the secondary inorganic aerosols, the thermodynamic equilibrium is computed using ISORROPIA, which has been updated to the version 2.1 here. Biogenic emissions have been updated and are now computed using MEGAN 2.1. Below-cloud scavenging has also been updated. After a presentation of the model CHIMERE and the aerosol module, the developments are validated over the year 2013 using surface measurements from the EBAS database by separating Europe into 5 coherent sub-regions based on country borders. The authors give some recommendations for

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future development. Finally the manuscript ends with a conclusion.

This manuscript is interesting for the aerosol community as it presents recent features in aerosol modelling. However, it presents some serious issues for a publication as it is.

The manuscript presents a new aerosol module (cf. abstract), but otherwise in the text it is treated as an update of the existing module. It is sometimes difficult to differentiate the parametrization and developments related to the 2013 version from the new 2017 version. The authors need to be clearer about this point that highlights the important development work done here. I would recommend to present the 2013 version of the parametrizations before introducing the new features. Following this remark, there is no comparison between the 2013 and 2017 versions over the year 2013. It would be very interesting to see the evolution in the performance of the model between the two versions.

The comparison to the observation set is very interesting as it uses a lot of collocated information on several measuring stations. It is then possible to evaluate the aerosol load, but also the aerosol composition and the seasonality. All these pieces of information could point out easily the strengths and the weaknesses of the model. However the authors only use ground based stations. It would have been interesting to compare the simulation to vertically integrated measurements such as aerosol optical depths, especially to evaluate the impact of the changes on wet deposition.

Also, almost all the mathematical formulations need to be reviewed. There are for example undefined variables used in equations or discrepancies between the name of a variable in the text and in an equation.

I would then recommend major revisions before publication.

#### General comments:

1. The use of paragraph breaks is sometimes puzzling, for example on page 13 line

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19. This sentence might make think that the next paragraph does not refer to the figures 7 and 8, which is not the case.

- 2. Page 2 line 2: I do not understand the presence of the "Vestreng, 2003" reference for the air quality model.
- 3. Section 2.1.1: Maybe a table that summarizes the reaction rate for the oxidation of SO<sub>2</sub> in clouds could be useful.
- 4. Section 2.1.3: I didn't understand how are the anthropogenic emissions managed for the POA and SVOC. You use POA emissions from an emission inventory. These POA are emitted into the species POAIP, POAmP and POAhP. Then you use the quantity of POA emitted to emit the SVOC by saying that SVOC = 5 \* POA and say that you don't take into account the IVOC. Is this right? How are then the SVOC emitted into the MELCHIOR species?
- 5. Page 6 line 4: I understand the dynamic method requires a lot of computation time explaining you choices. But did you run a test to know the impact it could have on a specific test-case for example?
- 6. Page 7, line 1: pleas also add "the mass fraction of respectively the solid phase, the aqueous phase and the organic phase **in the particle**" for a better understanding.
- 7. Section 2.1.6: The title is "Dry deposition of particles and semi-volatile organic species" but you talk about other gaseous species such as O<sub>3</sub>, SO<sub>2</sub>, etc. Please change the text to be consistent.
- 8. Section 2.2: When talking about the simulation set-up, you don't talk about the vertical resolution of the simulation made. Please add these informations.
- 9. Page 10, line 18: You wrote "Boundary Conditions were generated from [...]". Could you explain how they were generated?

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- 10. Section 2.3: This part is too small. Could you please add a table with the total number of stations and the number of stations in each of the regions you defined. According to Fig. 1 there are no stations for western Europe. Could you please explain why? Also, please add the link to the EBAS database in the text.
- 11. Page 11, line 19: You wrote "Only one station in Spain underestimates [...] concentrations for Na." while on Fig. 1 there are three blue triangles.
- 12. Page 11, line 29: You talk about the station ES0008R, but without showing any figures. Also this sentence is hard to understand.
- 13. Page 12 line 2: "The other stations [...] the opposite trend". I do not understand which other stations? All the stations or the other stations from southern Europe? Could you please make this part clearer?
- 14. Section 3.2: You are talking about sulfate observations. But are you using "total sulfate" or "corrected sulfate" (or non sea salt sulfate) measurements?
- 15. Page 14, line 23: I do not understand how an overestimation of NH3 emissions could induce an overestimation of TNO3. Could you please explain it with more details?
- 16. Section 3.3.1: How many measurements do you have for each stations in Table 6? it seems that some stations have very few measurements points, e.g. CH0002R looks like to have around 10 points. What is the confidence in the statistics you can have in this case?
- 17. Page 16, line 16: "especially in the Alps". How is the relief represented in the model in this area? Can it explain the overestimation of the PM by the model? Maybe you could try to interpolate the model at the altitude of the stations to improve the comparison made.



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- 18. Page, line 30: What is the effect of the strong overestimation over the Alps on the results of Fig. 15 and 17 for central Europe?
- 19. Section 3.3.3: You talk about Ca and NO<sub>3</sub> in  $PM_{10}$ , but in Fig. 18 there is "total Ca" and "total NO<sub>3</sub>". Are these two quantities supposed to be the same? Please change the text to be clearer.

## Figure related comments:

- 1. The manuscript preparation guidelines for authors claims that: The abbreviation "Fig." should be used when it appears in running text and should be followed by a number unless it comes at the beginning of a sentence, e.g.: "The results are depicted in Fig. 5. Figure 9 reveals that...". Please check that the right word is used in the text.
- 2. Also the authors should check the legend of their figures that are not always satisfactory. For example, the legend of the first figure does not mention what are the measuring station marked by dots.
- 3. Concerning the time series, it seems that there are missing values in the observations linked by a segment in the figures (e.g. On Fig. 12 between Feb and Mar 2013 for GR0002R). This can be misleading for the interpretation of the figure. Could you please change this? Maybe you could use symbol for the measurements and have continuous line for the simulation results.
- 4. Figure 2, 4, 6, 11 and 14: These figures does not seem to be complete on the right side for negative numbers. What represent the squares on these figures?
- 5. Figure 3, 5, 7, 8, 9, 15, 17: Is it possible to have slightly thicker lines? Is it possible to draw a line for 0 in order to read more easily some quantities such as the MFB.

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- 6. Figure 3: All your sub figures does not have the same size or position.
- 7. Figure 12 and 13: All your sub figures does not have the same size.
- 8. Figure 13: There seems to be a mistake. I think "CH0005R" should be "CY0002R".
- 9. Figure 16: The figure for  $PM_{2.5}$  is very small. Is it possible to enlarge it?

#### Table related comments:

- 1. Table 2, line "BiDer": hydrohilic -> hydro**p**hilic.
- 2. Table 2: for the type you mention type A, B and C, but in the table you only write hydrophilic or hydrophobic. Please explain this a little more.
- 3. Table 4: What are the Henry's law constant used for the other species such as HNO<sub>3</sub>?
- 4. Page 14, line 8: Please add a reference to Table 5 when talking about TNO3 and TNH4.
- 5. Table 5 presents results for  $O_3$  and  $NO_2$  but you never talk about them in the text.

Technical comments (when a letter or a word is missing, it is in **bold** in the comment):

- 1. Page 3, line 15: I guess the word "first" is missing in the sentence.
- 2. Page 5, line 19: "Thermodynamic of Secondary organic and [...]" -> "Thermodynamic of secondary organic and [...]"

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- 3. Section 2.1.5: the symbols w are different but refers to the same quantity. So are d and d. There is also a discrepancy between  $d_{sol}$  and  $d_{solid}$ .
- 4. Page 7, line 9-12:  $F_d$ , *i* is different from  $F_{dry,i}$ . Same for  $v_{d,i}$ ,  $v_d$  and  $v_d$ , *i*. What represents  $C_i$ ?
- 5. Page 7, line 25:  $\rho_{eau}$  and  $\rho$ . What is  $M_{eau}$ ? Does  $H_i$  stand for the Henry's law constant?
- 6. Page 7, line 26: Henry's law.
- 7. Page 9, line 12: there is no verb in the sentence.
- 8. Page 9, Eq. 18: What is *P*?
- 9. Page 9, Eq. 19:  $Delta \rightarrow \Delta$ ,  $k^{bin} \rightarrow k_i^{bin}$
- 10. page 9, Eq. 20: What is R and T?
- 11. Page 9, Eq. 21:  $Delta \rightarrow \Delta$
- 12. Page 10, line 2:  $J_{coaq,i}^{bin}$  and  $J_{coaq,i}^{b}$
- 13. page 10, Eq. 23:  $K_{j,k}$  instead of  $K_{j,l}$ . What is  $A_{p,i}$ ? The second sum symbol does not have attribute. Is the sum going from 1 to b over j?.
- 14. Page 10, line 13: Wind Speed -> wind speed (also line 15).
- 15. Page 10, line 13: Planetary Boundary Layer (PBL) -> planetary boundary layer (PBL).
- 16. Page 10, line 15: Temperature -> temperature.
- 17. Page 10, line 18: Mozart -> MOZART.

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- 18. Page 10, line 18: "Boundary Conditions" -> "boundary conditions"
- 19. Page 11, line 5: Figure 1 -> Fig. 1.
- 20. Page 12 line 17: Tsyro et al. (2011) and Neumann et al. (2016).
- 21. Page 15, line 11:"OM/OC simulated" -> "OM/OC ratio simulated".
- 22. Page 15, line 29: "summer concentrations are underestimated in summer". Summer is written twice.
- 23. Page 16, line 18: "PM2.5 and PM10, respectively" -> "PM2.5 and PM10 respectively", no comma.
- 24. Page 18, line 9: "Figure 12" -> "Fig. 13".
- 25. Page 19, line 9: "to take into IVOC emissions", it seems that a word is missing here.

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