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Geoscientific
Model Development
Discussions

Interactive comment on "Development of an inorganic and organic aerosol model (Chimere2017β v1.0): seasonal and spatial evaluation over Europe" *by* Florian Couvidat et al.

Anonymous Referee #1

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The paper describes a series of developments in the chemical transport model Chimere and the evaluation of the model against aerosol measurements in several stations in Europe during 2013. The changes include new biogenic VOC emissions, replacements of the inorganic aerosol model ISORROPIA with that of ISORROPIA-II, update of the secondary organic aerosol module and better description of wet deposition. The description of the changes and the evaluation of the model are satisfactory. The scientific content of the paper could be improved by better connection of the changes made with the predictions and also the performance of the model. Overall, the paper is suitable for publication in GMD after some improvements and corrections are made. These are described below.

General Comments

(1) One of the major weaknesses of the work is that there is very little about the effect of the various changes on the predictions of the CTM. I understand that repeating these simulations with the previous version of Chimere requires significant work and may not be worthwhile. However, some discussion of the results of previous evaluations of the model is necessary.

(2) Abstract. The use of the terms "good performance", "good seasonal evolution", "performs well", etc., should be avoided or at least the quantitative metrics should be presented instead.

(3) Abstract and rest of the paper. Most of the work focuses on PM10 with some analysis performed for PM2.5. However, it is not clear in most of the paper if the concentrations refer to the former or the latter. I think that the size-range should be mentioned everywhere in the paper to avoid confusion.

(4) Abstract. The conclusion that the model "strongly underestimates SOA concentrations" is not supported by the evaluation which is based on the comparison with total OC concentrations. The same applies to the explanation that "this underestimation could be due to a lack of anthropogenic SOA in the model".

Model description

(5) It is not clear if the composition of dust is taken into account in the calculation of cloud pH.

(6) The volatilities of the three POA components should be mentioned. Also despite the corresponding discussion it is not clear to me how their emissions are calculated from the OA emissions in the corresponding inventory for the various sources. A table with the corresponding volatility-resolved emission rates for the various sources would be helpful.

(7) The POA aging reactions (1)-(3) are a net sink of OH. In the rest of the SOA reactions it is assumed that the OH is recycled. Why are these treated differently?

(8) If my understanding is correct the authors assume that the primary OA emissions in the inventory that they are using represent 20 percent of the organic compounds emitted in a certain volatility range (less or equal than 100 μg m^{-3} as IVOCs are neglected). So the corresponding emissions are multiplied by a factor of 5. Given that a lot of these emissions have been measured at high OA levels this increase appears to be too high. It appears that the emissions used set the IVOC emissions to zero and at the same time add a lot of organic emissions to the more volatile SVOCs. This is an important issue for the model so some additional analysis of what exactly is done and why is necessary.

(9) The use of ISORROPIA-II is rather confusing. Its major advantage compared to ISORROPIA is its ability to simulate the thermodynamics of calcium, magnesium, etc., compounds. However, the authors state in Section 2.1.4 (page 5, lines 22-24) that the crustal elements are not taken into account for the partitioning. This counterintuitive choice requires some justification.

(10) Some discussion of the aerosol size resolution used in the model is needed.

(11) Section 2.1.8. The way that the mass transfer is between the gas and particulate phases is simulated is not entirely clear. If my understanding is correct, bulk equilibrium is assumed for both inorganics and organics for sizes below 1.25 micrometers. However, it is not clear what happens for coarse particles for compounds like ammonia, hydrochloric acid, organics, etc. Also is the formation of calcium nitrate the only way that nitrate can be transferred to the coarse particles? What about sodium nitrate?

(12) Tables 1-3. A list of the species names probably in the Supplementary Information is needed to understand the information in these tables.

(13) The Henry's law constants of a number of organic species are both in Tables 2

and 4 and in quite different units. These important parameters should be in one table with some explanation about their source. Also the fact that they appear to be known quite accurately (three significant digits in Table 2) is problematic.

Emissions

(14) The authors mention that they use the EMEP inventory published in 2003. Are they using any emissions updates for 2013 or the emissions used are that old? A table summarizing the anthropogenic emissions used for each major source category would be helpful together with additional information about the origins of these estimates. The biomass emission rates should be mentioned.

(15) How are the sea-salt emissions calculated? What is the assumed composition and size distribution? This scheme appears to perform quite well based on the Chimere evaluation presented here.

(16) Some additional discussion of the ammonia emissions used would be helpful. Is there a diurnal variation of the emissions? Is the monthly variation (shown in Fig. 10 for six countries) similar for all areas in the domain? What are the total emissions?

Observations

(17) A short description of the various stations used in the paper is needed. This could be part of the Supplementary Information. My understanding is that all of them are regional background stations. Is this correct? Are there any exceptions?

(18) There appear to be around 20 stations measuring sodium in Figure 2. However, 38 stations are mentioned in Table 5. What is causing this discrepancy? There are similar issues with other PM components.

(19) A number of urban background stations (e.g., in Paris) could have also been used in the evaluation. Is there a reason why they have been excluded? Also there should

be PM1 measurements from the ACTRIS network available for the same period.

(20) How well did the measured PM mass concentrations compare to the sum of the components? Could some of the measured PM mass be water as previous studies have indicated for Europe but also for the US?

(21) The number of measurements used in the evaluation should be mentioned in Table 5.

Boundary conditions

(22) A brief summary of the boundary condition values (or ranges) provided by Mozart for the various sides of the domain would be helpful. It appears that these boundary conditions may be partially responsible for some of the discrepancies between model predictions and observations in areas like Northern Europe.

Carbonaceous aerosol

(23) The authors discuss the potential errors in the comparison of model-predicted OM values with OC measurements. However, they never discuss the actual values of the OM/OC ratios that they use. I understand that these vary temporally, but the average values for each station for at least the summer and winter could be presented. Given that there are now a lot of measurements of this ratio by High-Resolution Aerosol Mass Spectrometers in locations around Europe a more informed comparison could be made.

(24) The study focuses on daily average measurements. However, some discussion of the diurnal variations of components like OM would have been welcome. There are measurements of these variations in stations like Melpitz that could be useful.

(25) The predicted high levels of OC in parts of central and eastern Europe during the winter appear to be too high. I am assuming that these are due to biomass burning.

Is this an indication of emissions that are too high? May be atmospheric mixing that is too weak? Some discussion is needed here because other authors have argued that the biomass burning emissions in the European inventory is too low. Could it also be a problem due to the high SVOC emissions used by the model that become particles under low temperatures?

(26) There is no discussion regarding the predictions of EC by Chimere.

Nitrates

(27) Some discussion of the evaluation of coarse nitrate predictions by Chimere are needed at least in Cyprus but also in other areas where PM2.5 and PM10 nitrate measurements may be available. A plot of this coarse nitrate in Figure 18 would be useful. Could some of the nitrate prediction problems be due to the challenges in predicting coarse nitrate?

(28) The model predicts high nitrate levels in the southeastern Mediterranean (Figure 6). This is rather unexpected due to the relatively low NOx levels and high sulfate concentrations. The values appear to be quite high compared to what has been observed (there are available measurements in Crete). Is this due to dust? How does the model produce so much nitric acid in that area?

Figures

(29) Figures 3, 5, 7, 8, and 9 are quite confusing because they contain too much information and do not have legends. One solution would be to show just the predicted and measured concentrations in these figures and then show the evaluation metrics in the supplementary information. It would be nice to also indicate the number of stations in each area in these figures.

(30) The axes of Figure 10 do not have titles.

(31) There are several cases with missing data in the time series of Figures 12, 13, etc. These are replaced by lines connecting the existing measurements. This is confusing and may be misleading. There should be gaps in the corresponding lines. Even better symbols could be used instead of lines for the measurements.

(32) An explanation of what is shown in the QQ scatter plot (Figure 16) is needed.

Other issues

(33) The use of Na instead of Na^+, SO_4 instead of SO_4^{2-}, etc., throughout the manuscript is problematic and should be avoided.

(34) There are a number of typos that should be corrected. Some of them:

Page 7, line 18. NH3

Page 7, line 26, Henr's

Page 11, lines 29-20, with strong observed of Na

Page 20, line 24, S. et al. (2016)

Page 26, lines 4-5. Missing author names.

Figure 3, 5, 7, 8 , 9 captions. monyhly.

Page 46, line 1. PM2.5

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