Modelling the mineralogical composition and solubility of mineral dust in the Mediterranean area with CHIMERE 2017r4

Menut, L., Siour, G., Bessagnet, B., Couvidat, F., Journet, E., Balkanski, Y., and Desboeufs, K.
https://www.geosci-model-dev-discuss.net/gmd-2019-337/

Dear Editor and reviewers,

We acknowledge the reviewers for the time spent to evaluate our work and for their minor revisions. We also acknowledge the Editor and we made all proposed changes in the revised manuscript. Please note that answers are in blue and after each reviewer's remark. When a large paragraph is added in the manscript, it is here described in a grey box.
All reviewers remarks were taken into account and are detailed in this letter.
Summarizing our answers:

1. Text, references and Figures (captions and labels) were checked and corrected as requested.

2. The two reviewers have questions about the function proposed to estimate the relative ratio of silt and clay as a function of the mean mass median diameter of the aerosol. We present here the problem we had: the goal of this function is to provide a simple and smooth transition between silt and clay fraction. The function proposed by Scanza et al. (2015) is very complex and when we computed it, we did not find the values presented in their article. Thus, we prefer to calculate this transition using another function, more simple and providing the same values.

3. The two reviewers ask for more details about the dry and wet deposition schemes used in the model. We add a section describing in detail these calculations.

Best regards,
Laurent Menut
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1 Reviewer #1

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The manuscript introduces a version of the CHIMERE regional chemistry-transport model, in which the mineralogical composition of dust aerosols is taken into consideration. It also presents results from an evaluation of the simulated dust mineral species and other aerosols versus observational data for the region of Europe, the Mediterranean, and North Africa, compared to a baseline simulation that takes into account only bulk dust without mineral speciation.

The explicit modeling of the mineralogical composition does not lead to an improvement of the simulated soil dust cycle or aerosol optical depth (AOD), according to the presented results. The model version with the minerals allows to explicitly simulate regional variations of $nssCa^{2+}$ and its ratio to total dust, though, which is important for biochemical cycles.

There have not been many models, regional or global ones, that consider the mineralogical composition of soil dust so far, and the presented one is a valuable addition to the few. I have a concern with respect to methodology and a few other points that should be addressed before publication, though.

1. Section 4.4.1: My main concern is with respect to the approach that is chosen to account for the wet-sieving bias in the database of the mineral fractions in soil by Journet et al. (2014), which is used as input. The authors present functions in Equation 1 to compensate for the wet-sieving bias, with the parameters chosen in a way that the values of the functions approximate the contributions of the soil clay and silt fractions to dust aerosols in Table 2a in Scanza et al. (2015). This may be a valid approach, principally. However, the reasoning by the authors for doing this is that they have not been able to reproduce the numbers in Table 2a by Scanza et al. (2015), using the equations in that paper. The authors assume anyhow that the numbers in the Table, which are approximated were correct. On what basis do they have confidence that their own formula adequately represents the contributions of the soil clay and silt fractions to the dust aerosol distribution then? That needs to be clarified.

   Answer:
   We understand this reviewer's remark. Probably, it was not well written in our manuscript. The problem was that we used the equation and were not able to calculate the same values as in table 2. The equation in Scanza et al. (2015) provides unrealistic values. This is why we preferred to write another equation able to provide values close to the ones in Table 2. This is illustrated in the answer to the Reviewer #2. About our confidence in the result of this simplified equation, we show in Figure 3 a comparison between the values of Scanza et al. (2015) and our equation: when Scanza et al. (2015) only provides mean values per size intervals, we propose a continuous fit, then not dependent on the model size distribution. And for all aerosol sizes, our fit gives the same values than the constant values of Scanza et al. (2015).

2. Page 4, Section 3.2, lines 107-108: The simulations with the GOCART model that are used for the boundary limits of the CHIMERE model domain only provide total dust fluxes. How are the mineral fractions of dust treated at the boundary limits?

   Answer:
   The reviewer is right, this problem was one of our major problem when developing the mineralogy in CHIMERE: how to redistribute the mineral dust at the boundaries without knowing their origin then their mineralogy? In fact, there is no rigourous answer and we then decided to create an 'other' category where we put initial and boundary concentrations. This is not very clean but it is without error. After tests, it was shown that this contribution is negligible compared to the emissions calculated in the modelled domain. This was added in the manuscript. The section 4.4 is in the manuscript about this point:

   > In addition, one has to note that the boundary conditions for mineral dust are entirely assigned to the "other" species, DuOT. Indeed, having no information from the global model used for the boundary conditions, it was not possible to assign these concentrations to specific minerals. To

> minimize the impact of this approximation on the boundary conditions, the simulations used in this study are done over a large domain.

Furthermore, for better reproducibility of the results from the current study, the information on what model simulations were used for the boundary limits of the model domain should be more specific than just naming on what papers they are based. Where can the simulations be accessed?

Answer:
The study was done with these GOCART boundary conditions and, apart the problem well pointed out by the reviewer, we are aware it is an old climatology, with a low resolution and too few informations. Our goal, for the next CHIMERE version, is too change these boundary conditions with a more recent dataset such as the CAMS global data. But, to be more accurate in this article, we added a new subsection called "Boundary conditions" containing:

> "Boundary conditions for mineral dust are calculated using a climatology calculated with the GOCART model, (Ginoux et al., 2001). This climatology was provided by Mian Chin and Paul Ginoux for the CHIMERE validation and distribution to users. The data are freely available on the CHIMERE download web site. The data represent a monthly global climatology simulation of mineral dust with an horizontal resolution of $2.5^o \times 2\ ^o$, Ginoux et al. (2001). The monthly mineral dust concentration fields are an averaged of years 1987, 1988, 1989, 1990 and 1997 and proposed in 7 size bins, later reprojected in the CHIMERE aerosol bins. The use of this climatology in the case of this study has a major weakness, knowing that the data are for the usual mean mineral dust species. The question was thus to choose how to redistribute this mean species into all mineralogical species. To avoid errors, it was decided to add this contribution into the 'other' species called DuOT. After some test cases, it was shown that the contribution of the boundary conditions was very low compared to the emissions calculated in the modelled domain. The impact of this hypothesis on the results was found to be negligible."

3. The authors should add a more comprehensive description of the wet deposition scheme that is used in CHIMERE to Section 4.4.2, since it seems to be a crucial part for understanding the results on wet deposition, which are presented in the manuscript.

   Answer:
   Additional informations about the dry and wet deposition fluxes calculation were added in the section 4 (see answer to Reviewer #2 where the whole text is extensively provided). This section was renamed because contains more informations than only about emissions. In place of "The mineral dust emissions", it is now "Model changes for mineral dust mineralogy". And it contains all changes made in the CHIMERE v2017r4 to include the mineralogy calculations. A new subsection was also added called "Boundary conditions" to better answer the previous remark of reviewer #1.

4. It will be informative, if results (figures and/or tables) for the resulting simulated volume/mass size distribution of the total dust concentration as well as the distribution of the mineral mass fractions over the dust size bins are also shown in the manuscript, even though measurements for evaluating them are not available. It still will be valuable for comparing with other models that simulate the mineralogical composition of dust aerosols. The lack of differentiation between different dust minerals with different particles densities by gravitational settling during transport may be due to the simulated size distribution.

   Answer:
   Yes, that's right. The goal of such study is to have more detail about the mineralogical and chemical composition of the mineral dust. We already provide this information with the Figure 4: monthly mean surface concentrations are presented for several sites, representative of different regions and with the mineralogical composition. We think it makes sense with this format. The conclusion of this figure

was that the variability remains very close to the emission and the emission of the different minerals was not so variable in space. Figure 4 is thus really representative of the result we could obtain and we concluded that to present the same kind of composition but with another choice of plot would be not very different.

5. Page 16, lines 285-286, and legend of Figure 7: The experiment for which the absolute AOD is shown in the top panel should be explicitly named there.

Answer:
Yes, of course, this information was missing. The absolute value of AOD is shown for the 'DUST" simulation. It was added in the text and in the Figure 7 caption. The new text is:

> The top panel displays the mean averaged value of AOD for April 2012 (when the largest surface concentrations were modelled) and for the simulation 'DUST'.

The corrected caption is:

> Monthly averaged Aerosol Optical Depth for April 2012 and over the whole modeled domain. (top) AOD absolute values are presented for the simulation DUST. (bottom) The map of difference represents the calculation of AOD(DUST)-AOD(MNRLO).

6. Page 17, line 292: The manuscript states, 'Statistical scores are calculated over 32 AERONET stations. Results are presented for selected sites in Table 6'. The phrasing is confusing. The statistical parameters are calculated and valid for the sample of all 32 stations, are they not? In what way are results shown for 'selected sites' in the table?

Answer:
The text was wrong and was corrected. The statistical scores are calculated over 32 AERONET stations and there is no 'selected sites' in Table 6.

7. Page 18, lines 312-313: A conclusion is stated there: 'Since the precipitations are well represented in the model, it indicates that the strength of the mineral dust plumes is overestimated in the simulation.'

This conclusion is not clear to me. The model simulated wet deposition flux of $nssCa^{2+}$ has a low bias, compared to measurements. Even if the error in the simulated precipitation is not large, the error in the deposition flux can have different causes. The entire simulated dust cycle, including the wet deposition flux may be too weak, or the simulated fraction of $nssCa^{2+}$ in dust may be too small, or the wet deposition scheme that is used to calculate the deposition fluxes may not be sufficiently efficient with respect to dust tracer removal.

Answer:
We are OK with this remark: our conclusion was too fast and too definitive. The reviewer is right: a lot of different processes may be at the origin of this underestimation. We change the sentence to open more to several possible causes. The sentence is then replaced by:

> The capability of a model to simulate the dust cycle contains many processes and, then, many possible errors. Here, the precipitation is correctly represented by the model. It means that the underestimation of modelled wet deposition fluxes compared to the measurements is probably due to other processes than a misrepresentation of the precipitation. This could be the altitude of the precipitating clouds, the trajectory of dust plumes (missing a station or not), the efficiency of the parameterized scavenging, possible errors on dust size distribution, a too small simulated fraction of $nssCa^{2+}$, among other possibilities.

8. Page 18, lines 317-320: I do not understand what this paragraph says. Please rephrase and explain this more clearly.

   Answer:
   Yes, it was an attempt to explain that the biases should be quite spatialized but are not spatialized at all in the end. Indeed, it is not clear. But as it is just a general idea, but nothing very accurate, we prefer remove this part from the manuscript. The paragraph is replaced by:

   > Since dust plumes are very spatially extensive, there is usually a bias between model and measurements for groups of stations located beneath these plumes. This is not the case here, since there are highly variable biases for nearby stations. The origin of the bias is therefore not due to a 'large scale' error: it is therefore probably not a transport problem. But it may be a precipitation problem, which is often a phenomenon of greater spatial variability on a small scale.

9. Units of shown variables should be added to the legend of those figures and tables where they are missing.

   Answer:
   That has been corrected for all Figures and Tables where it was the case.

Typos and language issues:

1. Page 3, line 71: Remove 'latter'.

2. Page 4, line 107: Replace 'where' with 'for which'.

3. Page 7, line 159: Fix typo in 'individual'.

4. Page 12, lines 217-218: Replace 'M' with 'i' in the description of the denotations of the equations, since 'i' stands for the individual mineral and 'M' for the total number of the minerals.

   Answer:
   The description of equation 5 was corrected and is now:

$$\left\{ \begin{array}{lcl} DF^b_{Nso} & = & \sum_{i=1}^{M} \left(DF^b_i \times \%_{Ni} \times \%_{solubNi} \right) \\ DF^b_{Nin} & = & \sum_{i=1}^{M} \left(DF^b_i \times \%_{Ni} \times (1. - \%_{solubNi}) \right) \end{array} \right. \quad (1)$$

   with DF^b_N the deposition flux of the chemical element N (i.e. Fe, Ca etc.) for its bin b. This flux is splitted between the soluble (so) and insoluble (in) parts. $\%_{Ni}$ is the percentage of chemical element N in each mineral M, $\%_{solubNi}$ the percentage of soluble fraction of chemical element N in each mineral M.

   Please note that the number was missing for this equation and was added for the revised version. Note also that we replaced nb by b for the bin number to avoid confusion with "number".

Answer:
That has been corrected for all remarks.

References

Ginoux, P., Chin, M., Tegen, I., Prospero, J. M., Holben, B., Dubovik, O., and Lin, S. J.: Sources and distributions of dust aerosols simulated with the GOCART model, Journal of Geophysical Research, 106, 20 255–20 273, 2001.

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Scanza, R. A., Mahowald, N., Ghan, S., Zender, C. S., Kok, J. F., Liu, X., Zhang, Y., and Albani, S.: Modeling dust as component minerals in the Community Atmosphere Model: development of framework and impact on radiative forcing, Atmospheric Chemistry and Physics, 15, 537–561, https://doi.org/10.5194/acp-15-537-2015, 2015.