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
March 20, 2020

1 Reviewer #2

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General comments

Numerical predictions of mineralogical effects on biogeochemistry and climate are highly uncertain. The authors implemented the mineralogical database to regional chemistry transport model. They confirm that this implementation does not substantially change the results of AOD, mass concentrations and deposition fluxes, following previous studies. I have some major comments to improve the paper.

Major comments

1. A fitting function to 4 data points, which were previously calculated by another fitting function, could introduce additional numerical errors. In fact, the fitting curve in Figure 3 is apparently different from that calculated by the original function. Although this would not substantially affect the results of AOD, mass concentrations and deposition fluxes, it would modulate the numerical predictions of mineralogical effects on biogeochemistry and climate. The original function should be used to avoid the error. At least, this caution should be noted in the manuscript.

   Answer:
   These remarks, and the associated doubt, about this function we proposed is a common point between the two reviewers. Our goal was primarily to use the Scanza et al. (2015) proposed function in our model. But results were not correct. We double checked the code (L. Menut and G. Siour) and found that we coded exactly what is in the paper. As, in addition, the formulation was very complicated in regard of the searched goal (i.e only provide a smooth transition from silt and clay, with values from 0 to 1), we prefered to implement a more simple and robust equation. We consider this is not adding potential errors as the reviewer write: the main idea of the authors in Scanza et al. (2015) is to propose a smooth transition but the absolute values they proposed are not exact and are proposed with constant values in large intervals.

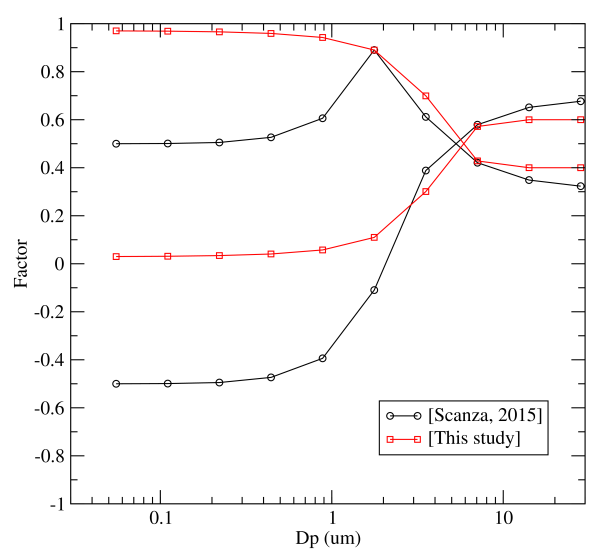


Figure 1. *Comparison between the function proposed by Scanza et al. (2015) and the one proposed in this study. The formulation by Scanza et al. (2015) is complex and appears to be numerically unstable with false values: negative values appear when we want only a smooth factor from 0 to 1. In addition, some unrealistic peaks are present, not conserving the total of 1 with the two parts of the function.*

   Figure 1 displays a comparison between the Scanza et al. (2015) equation and the one we proposed in this article. For mean mass median diamters below 1μm, it is clear that the Scanza et al. (2015) has unrealistic peak and negative values, when we expect to have a transition factor between 0 and 1. It is delicate to write in an article that the function we want to use is not correct and it is why we did not in the first version of the manuscript. But since the two reviewers have doubts about this, we modified this sentence in the manuscript:

> The relative part of clay and silt for each mineral depends on the mean mass median diameter of the emitted aerosol. We attempted to follow the formulation proposed in Scanza et al. (2015), with an equation and corresponding results in a Table. Unfortunately, the coding of the proposed formulation provides erroneous values, largely different from the results presented in their Table. Their formulation appears to be numerically not correct, in any case far from the simple goal which is to have a factor giving a smooth transition between 0 and 1. We thus define a new and simplified formulation as:...

2. Previous modeling studies have already implemented the mineralogical data to atmospheric chemistry transport models. The multi-model results and observational data are available over the model domain (Ito et al, 2019). Please discuss the results of the Fe solubility.

   Answer:
   Thanks for the reference of Ito et al. (2019) (note this paper was not published when our work was done and the draft written, then submitted). Then a discussion based on this multi-model work was added in the manuscript. About the Fe solubility, we are not in the same configuration than the models used in this study. For the moment, and in our model, there is no Fe in our anthropogenic emissions, thus it is not realistic to discuss the Fe solubility neither than compare the concentrations to measurements. But we added a sentence about this interesting point in the conclusion. This sentence was added at the end of the manuscript:

   > A step forward could be to add the Fe anthropogenic emissions in the model and then to have realistic Fe concentrations and deposited fluxes to make comparisons to measurements as done for example in Ito et al. (2019).

Specific comments

- p.4, l.104: Please describe the method to estimate the aerosols including nitrates, ammonium and sulphates. How do you consider the effect of mineralogical composition on these aerosol formations?

  Answer:
  In the first version of this manuscript, there was no description of active chemistry because the paper is dedicated to mineral dust. But CHIMERE is able to model inorganic species such as nitrates, ammonium and sulphates. It would be very long and not very useful to describe it in this 'dust' paper. Thus, we added references about the processes and chemical species. Note also that, for the moment, there is no link in the model between mineral dust and chemically active species. In this model version, mineral dust are considered as chemically inert. But these interactions could be the goal of future developments.
  The following sentence was added in the manuscript:

  > A complete chemistry is included in the model, a general description of gaseous and aerosol schemes is provided in Mailler et al. (2017) for this model version, including a detailed description of the aerosol scheme in Couvidat et al. (2018). The chemical evolution of gaseous species is calculated using the MELCHIOR2 scheme. The aerosol size distribution is represented using ten bins, from 40 nm to 40 μm, in mean mass median diameter as described in Menut et al. (2016) and updated in Mailler et al. (2017).

- Section 4: Please describe the method to estimate the mineral dust deposition flux and specify the effect of the aerosol density on the dry deposition.

Answer:
Yes, this request was also written by Reviewer #1. A subsection was added in the manuscript, describing the dry and wet deposition schemes used for aerosols. This new section is as follows:

Aerosols, including mineral dust, may be dry or wet deposited, depending on the meteorology and the surface characteristics. The dry deposition velocity is estimated following Zhang et al. (2001):

$$v_d = v_s + \frac{1}{r_a + r_s} \tag{1}$$

with v_s the settling velocity, r_a the aerodynamical resistance depending on the turbulence close to the surface and r_s the surface resistance, depending on the vegetation type. The aerodynamical resistance r_a depends on several turbulent parameters, such as the Monin-Obukhov length L, the friction velocity u_*, and the dynamical roughness length z_{0m}.
Depending on the atmospheric surface layer stability, r_a is estimated as:

$$\begin{cases} r_a &= \frac{1}{ku_*}\left[\ln\left(\frac{z}{z_{0m}}\right) + 4.7(\zeta_r - \zeta_0)\right] & (\mathrm{stable, L \geq 0}) \\ &= \frac{1}{ku_*}\left[\ln\left(\frac{z}{z_{0m}}\right) + \ln\left(\frac{(\eta_0^2+1)(\eta_0+1)^2}{(\eta_r^2+1)(\eta_r+1)^2}\right) + 2\left(\tan^{-1}\eta_r - \tan^{-1}\eta_0\right)\right] & (\mathrm{unstable, L < 0}) \end{cases} \tag{2}$$

where L the Monin-Obukhov length, z_r a reference height taken at the middle of the first vertical model layer, $\eta_0 = (1-15\zeta_0)^{1/4}$, $\eta_r = (1-15\zeta_r)^{1/4}$ and $\zeta_0 = z_0/L$, $\zeta_r = z_r/L$, k=0.41 the von Karman constant. z_0, the dynamical roughness length, depends on the fraction of land-use for each category and on the season (see Menut et al. (2013) for details and values).
The surface resistance r_s for aerosols follows the scheme of Zhang et al. (2001) and is calculated as:

$$r_s = \frac{1}{\epsilon_0 \times u_* \times R_1 \times (E_B + E_{IM} + E_{IN})} \tag{3}$$

with ϵ_0 is a constant set to ϵ_0=3, for all landuse categories, R_1 a correction factor describing the relative amount of aerosols sticking at the surface, E_B collection efficiency from Brownian diffusion, E_{IM} collection efficiency from impaction and E_{IN} collection efficiency from interception. The R_1 factor is estimated following Slinn (1982):

$$R_1 = \exp(-St^{1/2}), \tag{4}$$

where this factor is applied only for particles with $D_p > 5\mu m$. For Brownian diffusion, the resistance is estimated as:

$$E_B = Sc^{-\gamma} \tag{5}$$

where γ is a constant depending on the landuse type. In the model, this constant varies between 0.54 and 0.58. For the impaction, the resistance can have a lot of definitions, depending on the landuse. By default, the resistance value used is:

$$E_{IM} = \left(\frac{St}{\alpha + St}\right)^2 \tag{6}$$

The α values are landuse dependent and are tabulated following Zhang et al. (2001). In this model version, a distinction is made between northern and southern hemisphere, in order to use the correct

α for a specific modelled day. For specific vegetation types, the formulation is changed. For high vegetation (such as forests), the resistance proposed by Giorgi (1986) is used:

$$E_{IM} = \left(\frac{St}{0.6 + St}\right)^{3.2} \tag{7}$$

For grassland vegetation, a parameterization is proposed by Davidson et al. (1982):

$$E_{IM} = \frac{St^3}{St^3 + 0.753\,St^2 + 2.796\,St - 0.202} \tag{8}$$

Finally, the collection efficiency from interception is calculated as:

$$E_{IM} = \frac{1}{2}\left(\frac{D_p}{A}\right)^2 \tag{9}$$

with A a characteristic diameter given for landuse and seasonal categories. The settling velocity v_s represents the effect of gravity on particles and is calculated as:

$$v_s = \frac{1}{18}\frac{D_p^2\,\rho_p\,g\,C_c}{\mu} \tag{10}$$

with ρ_p, the particle density, D_p the mass median diameter of particles, C_c a slip correction factor accouting for the non-continuum effects when D_p becomes smaller and of the same order of magnitude as the mean free path of air, λ, Seinfeld and Pandis (1998). g is the gravitational acceleration with g=9.81 m s^{-2}, μ the dynamic viscosity (here the air dynamic viscosity is set to μ_{air}=1.8 $\times$ 10^{-5} kg m^{-1} s^{-1}). The slip correction factor C_c is estimated as:

$$C_c = 1 + \frac{2\lambda}{D_p}\left[1.257 + 0.4\,\exp\left(-\frac{1.1 D_p}{2\lambda}\right)\right] \tag{11}$$

with λ the mean free path of air, in meters, estimated as:

$$\lambda = \frac{2\mu_{\mathrm{air}}}{p\sqrt{\dfrac{8M_{\mathrm{air}}}{\pi R T}}} \tag{12}$$

where M_{air} is the molecular mass of dry air (here 28.8 g mol^{-1}), T the temperature (K), p the pressure (Pa), μ the air dynamic viscosity and R the universal gas constant.

Answer:
This also answers the question about the use of the density for the dry deposition. The wet deposition is calculated as follows:

The aerosols wet deposition calculation is separated between rain and snow. There is also a distinction between the wet deposition in-cloud and below cloud.
For below-cloud scavenging, aerosols are scavenged by raining drops. Following Willis and Tattelman (1989), a polydisperse distribution of raining drops is applied:

$$N(R) = 1.06 \times 10^{14} \times P^{-0.0295d0}(2R)^{2.16d0}\exp\left(-5679 P^{-0.153d0} 2R\right) \tag{13}$$

with P the precipitation rate in mm/h and R the radius of the droplet (in m).
The flux of deposition is calculated with:

$$F^i_{bc} = c^i \times \sum_R \pi R^2 u_g(R) E(R, r_i) N(R) \tag{14}$$

with i the aerosol species, r_l the radius of the particle (in m), u_g the terminal drop velocity (in m/s), $E(R, r_l)$ the collision efficiency of a particle with a raindrop, $N(R)$ (in m^{-4}) the raindrop size distribution.
For below-cloud scavenging of particles by snow, the particles are scavenged by appling the parameterization of Wang et al. (2014). A scavenging coefficient λ_{snow} is computed with:

$$log(\lambda_{snow}) = logA + B \tag{15}$$

with A and B fit function depending on the aerosol mean mass median diameter D_p. The flux of deposition is calculated with:

$$F^i_{in} = -\lambda_{snow} \times c^i \tag{16}$$

For in-cloud scavenging is here considered only when a precipitation occurs. The rate of deposition is computed by calculating the rate of impaction between hydrometeors and cloud droplets (assumed to have a diameter of 10 μm). The rate of scavenging is computed with equations 14 and 16 for D_p=10μm.

- p.5, l.122: To clarify a new implementation in this work, the first part should be moved before the several changes. Otherwise, please clarify the improvements from the Beegum et al. (2016), who implemented the MODIS erodibility to the CHIMERE model.

  Answer:
  Following this remark, the sections were redesigned. One part for the distributed CHIMERE version and another separated part for the new implementation related to dust mineralogy. Many details are already presented in the publication of Beegum et al. (2016) about erodibility and we added a few details.

- p.5, l.126: Please show the smooth function and the comparison with the measurements. How did you apply the function to different land surfaces? Please clarify the differences in the dust emissions with and without the function.

  Answer:
  The smooth function was already showed and explained in Mailler et al. (2017). As it is not authorized (and useful) to publish two times the same thing, we added a reference to the original publication. About the different landuses, there is no use of this information. This was added in the text.

- p.9, Table 3: How did you estimate the Fe solubility of 0.17% for illite? Please specify the reference, or correct the value. Please evaluate the Fe solubility with observational data over the ocean in this paper. Please clarify the differences from previous modeling studies in the estimate of the Fe solubility.

  Answer:
  The values are extracted from the Paris et al. (2011) work. Since we used exactly the values already discussed in this paper, we added the reference directly in the section 4.3 and Table 3. Our goal is not to evaluate the chosen constants in this paper since it was already done in other papers with exactly the same values, such as Balkanski et al. (2007). We remind that this study is dedicated to implement in the regional model CHIMERE exactly the same type of calculation than in the LMDz model: the goal is to have this functionality in a regional model in addition to the global one.

- p.10, l.177: How did you calculate the deposition fluxes for the mineral species?

Answer:
This question was already adressed and then already answered. Please see the answer to your first comment.

- p.10, l.193: Presumably, you used different size distribution of emitted aerosols. How did you calculate it? Please specify your calculation using their equation, which should provide the results presented in their Table 2a in your case.

  Answer:
  There is several questions in this comment. About the question on the calculation of the Scanza et al. (2015) formulation, at the beginning of this answer, we already presented the result of our calculation, checked many times and the Reviewer can see the problem. About the calculation of the aerosol size distribution used for the transport and then applied in equation (1), it is fully described in Mailler et al. (2017). A sentence was added about this point is the section about the model presentation.

  > The aerosol size distribution is represented using ten bins, from 40 nm to 40 μm, in mean mass median diameter as described in Menut et al. (2016) and updated in Mailler et al. (2017).

- p.16, l.284 and p.21, l.344: Please show the results of radiation effect, or rephrase the sentences.

  Answer:
  The term 'radiation' was used to say that a change in aerosol optical properties may induce a change in radiation and it is quantifiable via the AOD. The sentence was changed to eliminate 'radiation'. In the conclusion, it is correct since a change of the refractive index has an impact on the aerosol radiative effects.

- p.21, l.333 and Table 8: Please show the range of latitude and longitude for each region and compare the results over the same region.

  Answer:
  The range in latitude is expressed directly with the "Region" column. For the results of Lequy et al. (2013), there is no range since it corresponds to measurements at a specific location. The idea behind this table was to propose a discussion about the very large variability of this ratio and to show that very few studies were dedicated to its quantification. In our study, we propose a map and it is a novelty. Restricting the comparison by latitude bands is not at all the purpose of the table presented.

- p.21, l.349: Please clarify the strong dependency of settling velocity on the density in the method, or rephrase the sentence.

  Answer:
  This question was already adressed by this Reviewer and we answered with the complete description (as requested) of the dry and wet deposition calculation.

Technical comments

- Figures 1 and 2 as well as Tables 1 and 2 may be moved to supplementary materials to avoid the redundancy.

  Answer:
  These Tables are really useful since they concatenate all interesting informations about the choices made to represent the mineralogy in a model. We think they are their place in the article. The Figures are here to show the differences between the values and it is a complementary information that the quantified values in the Tables.

- p.1, l.4: Please correct in.

  Answer:
  Corrected.

- p.2, l.47: Please correct out.

  Answer:
  Corrected.

- p.3, l.49: Please correct et.

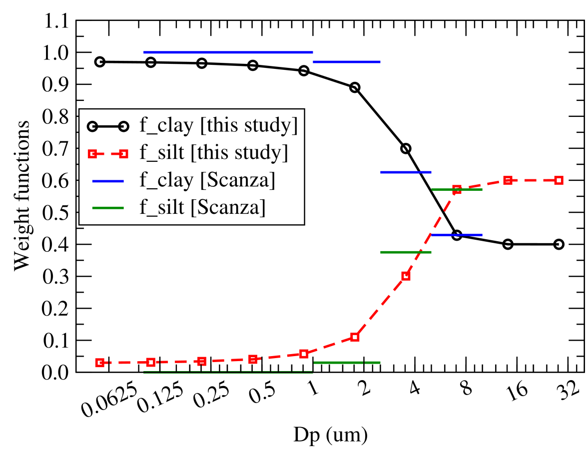
  Answer:
  We don't see the problem with line. But the text was corrected in a general manner.

- p.5, l.113: Please remove the.

  Answer:
  Corrected. The complete paragraph was rewritten.

- p.10, l.184: Please delete ,.
- p.11, Figure 3: Please correct f_clay [Scanza] and weight functions.

  Answer:
  Yes, right. The Figure was reprocessed to correct this error. The new Figure is:



- p.12, l.212: Please correct emission.

  Answer:
  Corrected.

- p.12, l.216: Please add the number to the equation.

  Answer:
  Corrected.

- P.17, Figure 7: Please correct the caption.

  Answer:
  Corrected. The caption is now:

  > 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).

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