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## Reply on RC1

Akinori Ito et al.

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Author comment on "Less atmospheric radiative heating by dust due to the synergy of coarser size and aspherical shape" by Akinori Ito et al., Atmos. Chem. Phys. Discuss., <https://doi.org/10.5194/acp-2021-134-AC1>, 2021

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Our responses to the comments from Reviewer 1 are included in the followings.

Reviewer #1

### Comments:

The present work aims at investigating the direct radiative effect of mineral dust aerosols and in particular it focuses on the effect of dust asphericity. This analysis takes advantage of a strong background of work performed in past years by the authors of the manuscript (work on size distribution and inclusion of non-spherical spectral optical properties calculations, Kok et al. 2017 Nat geosci ; DustCOMM dataset creation, Adebisi et al., ACP, 2020 ; study of the asphericity of dust and inclusion of the effect on reducing gravitational settling, Huang et al., GRL, 2020) to realise new model simulations with the IMPACT model coupled with the RRTMG radiation code. The IMPACT model has been improved in this study compared to its default configuration by accounting mainly of : a better soil moisture dataset from satellite observations to improve dust emissions, to include the effect of asphericity on gravitational settling velocity, by integrating with the RRTMG radiative code, and by using the DUSTCOMM dataset to constrain simulations. The paper provides a series of sensitivity simulations varying mainly the size distribution and the spectral optical properties of mineral dust aerosols and assuming or not the effect of asphericity in the simulations. The results are compared to field observations of the dust DREE (direct radiative effect efficiency,  $Wm^{-2} AOD^{-1}$ ) to identify the simulation configuration that best reproduces field measured dust perturbations at the surface and at TOA. The main conclusion of the paper is that improving the simulation scheme (improved vs default simulations) does not significantly changes the TOA global annual net DRE, conversely both the surface cooling and the atmospheric heating are strongly reduced assuming coarse aspherical dusts.

The paper is potentially a nice contribution for the scientific community. It provides interesting insight on dust aerosol science and contributes to further advance in the modeling of the dust cycle and its direct effects. Despite, I find that the paper suffers from an unclear identification of the objectives and a poor contextualization compared to the recent literature. As well, the presentation of the modelling simulations and the description and discussion of the results should be improved. In the current form I find the paper a bit difficult to read and I suggest that some major revisions are applied for improving the organisation and the presentation/discussion of the results. I compiled

some comments below.

**Response:**

We would like to thank the reviewer for his or her constructive comments which helped us to improve the readability of our manuscript substantially. We thoroughly revised the manuscript following suggestions made by the reviewer.

**General comments**

**Comment 1:**

One of my problem, especially at the very first reading, was to understand the main objective of the paper. I had the feeling by reading the title that asphericity was the main topic, then in the introduction it is discussed that asphericity is not so important (see lines 65 to 74) and then the paper introduces many simulations testing the dust DRE sensitivity and many test studies. The overall introduction and description of the work should be more incisive and clear in the objectives and scientific questions to test.

**Response:**

Thank you for your suggestions. To elucidate the importance of asphericity is one of our main topics. In the revised paper, we emphasized the importance of asphericity for the calculation of dust aerosol spectral optical depth and radiative effect. To do so, we presented an additional simulation result (Experiment 5) to elucidate the asphericity effects. We revised the title, "Less atmospheric radiative heating by dust due to the synergy of coarser size and aspherical shape", and paragraphs in the introduction on p.3, l.68 and l.75:

"Second, previous studies have shown that the SW radiative effect of dust asphericity on climate simulations is minor on a global scale, partly because the larger DAOD is compensated for by the larger asymmetry parameter of aspherical dust, which reduces the amount of radiation scattered backward to space (Räisänen et al., 2013; Colarco et al., 2014)."

"However, the assumption of spherical shape in models leads to a substantial underestimation of the extinction efficiency and thus DAOD near the strong source regions, mainly because the assumption of sphericity causes an underestimation of the surface-to-volume ratio compared to aspherical dust (Kok et al., 2017, 2021; Hoshyaripour et al., 2019; Tuccella et al., 2020). Radiative effect efficiency is often used for the evaluation of the models and is defined as the gradient of a linear least squares fit applied to AOD and dust radiative effect at each two-dimensional (2-D) grid box ( $W \cdot m^{-2} AOD^{-1}$ ). Thus, the estimates of the dust radiative effect efficiency could be biased, in part, due to large uncertainties associated with the spherical assumption on AOD retrieval (Zhou et al., 2020)."

**Comment 2:**

Following the previous comment, I have also found a little bit tricky to follow/understand the scope of the many simulations performed despite the synthesis effort in Table 1 and 2. There are many things and concepts in these simulations and a non-expert reader could be lost. Probably be clearer in describing the strategy?

**Response:**

In revised paper, we changed the structure of subsection and added outline of the

methodology used to obtain the sensitivity simulations of dust radiative effects: (1) the size-resolved abundance, (2) particle shape, and (3) mineralogical variability on p.4, l.118:

“In section 2.3, we describe the DustCOMM data set used to adjust (1) size-resolved abundance of dust concentration. In section 2.4, we describe the adjustment factor of (2) particle shape for spectral optical properties. In section 2.5, we describe differences in spectral refractive indices due to (3) different mineralogical compositions for the radiative flux calculation.”

**Comment 3:**

The paper refers too much to the supporting information, in particular in the Results section. While on one side I appreciate the effort of synthesis, I feel that it is quite difficult to follow the reasoning when being obliged to go back and forth from the main paper to SI. I suggest the authors to consider revising their strategy of presentation of the results in order to help the reader following the reasoning.

**Response:**

In the revised paper, we showed the evaluation of the various model experiments against semi-observationally-based estimates in box plots and Taylor diagrams (Taylor, 2001) in the main paper to provide a concise statistical summary of the bias, correlation coefficient, root mean square errors, and the ratio of standard deviation on p.11, l.315. Accordingly, we deleted supplementary figures.

“We compared our model estimates of DAOD<sub>550</sub> against semi-observationally-based data in box plots and Taylor diagrams (Taylor, 2001) for the evaluation of the various model experiments against semi-observationally-based estimates (Ridley et al., 2016; Adebisi et al., 2020) to provide a concise statistical summary of the bias, correlation coefficient, root mean square errors, and the ratio of standard deviation (Fig. 2, Tables S1 and S2).”

**Comment 4:**

Referring to lines 51-53 « On the other hand, model errors due to the underestimated coarse dust load and corresponding warming might be compensated for in models by using a refractive index that is too absorbing (Di Biagio et al., 2019), and which depends on the mineral composition of the dust ». Isn't it the same case here based on your results? Here the size distribution is cut at 20µm despite field evidences that larger particles are efficiently retained during transport (see FENNEC or SALTRACE results) and the best agreement with observations is found then when a stronger absorption is assumed in particular in the LW range, where the contribution of the coarse dust component is more critical. Is this result just due to the missing coarse size in the model above 20 µm? I would expect such a discussion in the paper (I noticed a mention to this at the very end of the conclusions, but the issue is argued to be related only to « Godzilla » type events and not relevant elsewhere. Is this really true or this aspect deserve more discussion?)

**Response:**

We thank the reviewer for this insightful comment. Song et al. (2018) found that the combination of Fennec dust particle size distribution and OPAC-LW, which was originally taken from Volz (1983), yielded the best simulation of the dust LW radiative effect in comparison with the satellite flux observations (i.e., CERES OLR), compared to the Di Biagio et al. LW refractive index. Marine sediment traps suggest that giant particles are dominated by platy mica and rounded quartz particles (van der Does et al., 2016).

Indeed, the dust sample for V83 was collected from rainwater after strong wind by Volz (1983). Thus, mineral composition of the giant particles could be different from the aerosol samples generated from soils in the laboratory by Di Biagio et al. (2017), which may reflect less absorbing LW refractive index of DB17 than V83, as Di Biagio et al. (2017) found a linear relationship between the magnitude of the imaginary refractive index at 7.0, 9.2, and 11.4  $\mu\text{m}$  and the mass concentration of calcite and quartz absorbing at these wavelengths. In revised paper, we replaced Experiment 1 with the same LW refractive index of Volz (1983) as in Experiment 2, and presented additional simulation results (Experiments 8, 9, and 10) to elucidate the sensitivity of radiative effect to refractive index.

Di Biagio et al. (2020) mentioned, “the key role of particles larger than 20  $\mu\text{m}$ , however, does not only rely on their direct contribution to the DRE but mostly on the fact that their inclusion reduces the contribution by smaller (cooling) particles to the global dust cycle”. We revised the sentence on p.2, l.43:

“The model errors in dust size distribution and particle shape can lead to an overestimate of fine dust load after the dust emissions in the models are scaled to match observed dust aerosol optical depth at 550 nm ( $\text{DAOD}_{550}$ ). The corresponding overestimate of SW cooling might be compensated for in models by using a refractive index that is too absorbing (Di Biagio et al., 2019, 2020), which depends on the mineral composition of the dust.”

The coarse size in the model above 20  $\mu\text{m}$  deserves more discussion. The combination of less LW absorption and coarser size can be examined from revised Fig. 10b. The dust size beyond 20  $\mu\text{m}$  might be partly compensated for in our model by using a refractive index that is more LW absorbing. Thus, we added the comparison with other modeling studies which considered the dust size beyond 20  $\mu\text{m}$  (Di Biagio et al., 2020) to Table 5 and revised Fig. 9 on p.14, l.409:

“A relatively good agreement of net RE by dust at TOA with Di Biagio et al. (2020) ( $-0.06 \text{ W}\cdot\text{m}^{-2}$ ) could be obtained from both the IMPACT-Sphere-Mineral-V83 (E1) and DustCOMM-Asphere-DB19-V83 (E2) simulations (Fig. 9 and Table 5). On the other hand, our modeled dust net RE at the surface from DustCOMM-Asphere-DB19-V83 (E2) was much larger than Di Biagio et al. (2020) ( $-0.63 \text{ W}\cdot\text{m}^{-2}$ ) and IMPACT-Sphere-Mineral-V83 (E1). The synergy of coarser size and aspherical dust could contribute to the less surface warming of the DustCOMM-Asphere-DB19-V83 (E2). At the same time, the more absorptive LW dust refractive index (V83) than DB17 (red diamond in Fig. 10b) could also contribute to the less surface warming, which might be partially compensated for in our model by the omission of dust with diameters in excess of 20  $\mu\text{m}$ . Consequently, our estimate of atmospheric radiative heating by dust from DustCOMM-Asphere-DB19-V83 (E2) was lower than Di Biagio et al. (2020) ( $0.63 \text{ W}\cdot\text{m}^{-2}$ ) and IMPACT-Sphere-Mineral-V83 (E1).”

We added the following sentences to conclusions on p.15, l.456:

“Moreover, such large particles can be transported to higher altitudes and longer distances than the model prediction. The higher the dust layer resides, the larger the dust LW RE at TOA is estimated under the clear-sky conditions (Liao and Seinfeld, 1998). Marine sediment traps, which are located underneath the main Saharan dust plume in the Atlantic Ocean, suggest that giant particles are dominated by platy mica and rounded quartz particles (van der Does et al., 2016). Thus, mineral composition of the giant particles could be different from the aerosol samples generated from soils in the laboratory by Di Biagio et al. (2017), which may reflect less absorbing LW refractive index of DB17 than V83. Indeed, the dust sample was collected for V83 from rainwater after strong wind. On the other hand, the contribution of the LW scattering might be underestimated in the models, as Di Biagio et al. (2020) noted that the adjustment factor was estimated for dust of diameter less than 10  $\mu\text{m}$  and thus might be a lower approximation of the LW

scattering by coarse dust. Therefore, a better understanding of the effect of such large particles beyond 20 µm and mineralogical composition on radiation balance remains a topic of active research, given their potential to amplify the warming of the climate system.”

This was also reflected on introduction, p.3, l.91:

“However, the speciation of dust into its mineral components inherently comprises uncertainties on soil mineralogy, mineral content in size-segregated dust particles, and refractive index of mineral, partly due to the differences in prescribed parameters such as the particle size.”

### **Specific comments**

#### **Comment 1:**

Abstract : the concept of default and improved simulation si given, but I am not sure it is fully clear at this stage. I would remove this nomenclature from the abstract text.

#### **Response:**

The default and improved was rephrased by before and after the adjustment, as was suggested.

#### **Comment 2:**

Line 23 : please specify the temporal/spatial scale of these estimated effects (global annual I guess)

#### **Response:**

This was done, as was suggested.

#### **Comment 3:**

Line 27 : I would say « warming of the Earth-atmosphere system by trapping incident and outgoing radiation »

#### **Response:**

This was done, as was suggested.

#### **Comment 4:**

Line 27 : what do you mean with « climate feedback ». Is this the correct term here ?

#### **Response:**

We added following sentence before the climate feedback on p.1, l.34:

“Radiative effect by dust aerosols perturbs surface temperature, wind speed, rainfall, and vegetation cover, which may induce feedback on dust emissions (Perlwitz et al. 2001; Miller et al., 2004a; Colarco et al., 2014).”

#### **Comment 5:**

Lines 54-64 : probably a word here also on the minerals affecting LW absorption would be good

**Response:**

This was added, as was suggested on p.3, l.88:

"The dust complex refractive index in the LW also depends on the particle mineralogical composition (Sokolik et al., 1998). Di Biagio et al. (2017) found a linear relationship between the magnitude of the imaginary refractive index at 7.0, 9.2, and 11.4  $\mu\text{m}$  and the mass concentration of calcite and quartz absorbing at these wavelengths."

**Comment 6:**

Lines 65-74 : I have to admit that I am quite confused here. The scope of the paper is to include asphericity effects but here I understand that it is not a big issue for global modelling. Please be more clear and focused on the objective and contours of the work.

**Response:**

This was revised, as was suggested in general comment 1.

**Comment 7:**

Lines 80-84 : it is quite unusual to draw the conclusions or the results in the introduction of the paper

**Response:**

This was removed, as was suggested.

**Comment 8:**

Line 91 : what is the forward model referred here ?

**Response:**

This is the IMPACT model simulation before the adjustment. The "forward" was rephrased by "IMPACT" in revised paper.

**Comment 9:**

Lines 104-112 : could be possible to give a bit more information or reference to the model capacity in reproducing dust mineralogy reasonably ?

**Response:**

The references of recent review papers including multi-model evaluations were added, as was suggested. We added the sentence on p.5, l.148:

"In recent review papers, multi-model evaluations of aerosol iron concentrations and their solubilities have been comprehensively summarized on global and regional scales (Myriokefalitakis et al., 2018; Ito et al., 2021)."

**Comment 10:**

Lines 123-128 : even by accounting for an « optimized » asphericity factor for dust in the model the lifetime of the coarsest bin is too low compared to pas literature. What is the impact of this on the results and overall sensitivity ? I guess this is a crucial point.

**Response:**

This is the one of our main topics and the reason why we adjusted the size-resolved abundance of dust concentration. The impact of this on the results and overall sensitivity was evaluated by "Experiment 3 – Experiment 4", as was summarized in Table 2. To elucidate this, we added the following sentence on p.6, l.166:

"The impact of this underestimate of atmospheric lifetime is explored using the DustCOMM data set, as was summarized in Table 2 (E3 – E4)."

**Comment 11:**

Line 134 : given that the focus of the paper is asphericity effects I am quite surprised to see that Mie theory is used here. However is then in section 2.4 that it is explained what it is done. Not sure the two things should not be merged together.

**Response:**

We presented the additional simulation result (Experiment 5) to show the asphericity effects. To elucidate this, we added the following sentence on p.6, l.177:

"The impact of this spherical assumption is explored using aspherical factor, as was summarized in Table 2 (E5 – E4)."

**Comment 12:**

Section 2.3 : I have to say that it remains a bit unclear how the dustcomm database is used in practice until sect 2.4. This sect 2.3 is a bit confusing to me

**Response:**

This was revised, as was suggested in general comment 1 and specific comment 10.

**Comment 13:**

Section 3.3 : should be taken in mind and reminded to the reader that the comparison in the LW range is more tricky than in the SW since there is a stronger dependence of the DRE on the vertical profile of dust, temperature and water vapour profiles therefore affecting the measured and modelled comparison

**Response:**

This was added, as was suggested on p.12, l.360:

"The dust LW radiative effect efficiency depends strongly on the vertical profile of dust concentration, temperature, and water vapor, which would affect the comparison due to a high variability in these factors (section 2.6)."

**Comment 14:**

Section 4 and throughout the text : pay attention to refer to « spectral optical properties » when related to both the SW and LW spectra

## Response:

We referred to spectral optical properties when related to both the SW and LW spectra, as was suggested.

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