# Response to Reviewers

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Dear Yves,

Thank you for your comments on the initial draft that improved its clarity. We also greatly appreciate the extensive and thoughtful comments of the two reviewers that identified additional points of confusion. We have used the extra time you have given us to thoroughly revise the article. We have attempted to address the reviewers' comments while trying to simplify and improve the clarity of the remainder of the article. (As an example, we combined Figs. 9 and 10 to facilitate the comparison of the globally averaged mineral fractions of each experiment.)

One consequence of the extensive revisions is that a version showing the contrast of the original and revised articles is so marked up as to obscure the specific changes made in response to the reviewers. (Both latexdiff and Adobe Acrobat Pro gave marked-up changes that were similarly tangled and confusing.) As an alternative, our response to each of the reviewer comments below contains a precise reference to a page number or section of the revised article where a change was made. (Where the change is limited to a sentence or two, we simply quote the change in our response.) We think these references will make it clear how we have attempted to improve the article in response to the reviewers' comments.

Best wishes,

Ron Miller
Jan Perlwitz
Carlos Peréz García-Pando

**Reviewer 1:**

*This article provides a detailed treatment of the partitioning of minerals into dust aerosols at emission, which is critical for capturing the many mineralogy-dependent impacts of dust, for instance on radiation, biogeochemistry, and clouds. The methodology developed in this work is described in detail, is (mostly) well justified (where possible; sometimes experimental constraints are not available), and assumptions and weaknesses of the approach are discussed in detail.*

*The methodology draws partly from the recent previous study of Scanza et al. (2015). In essence, it partitions minerals in the soil into suspended dust aerosols using brittle fragmentation theory for D<20 um, extended to D=50 um by the detailed measurements of Kandler et al. (2009). This empirical extension is necessary because the soil mineralogy in Claquin et al. (1999) and Nickovic et al. (2012) is described in terms of clay (D<2 um) and silt (2<D<50 um). Combined with several other improvements, for instance in the treatments of iron oxide and quartz, the methodology in this study is advanced over that in Scanza et al. (2015). As such, this article makes an important step in progressing dust cycle models, and therefore makes a substantial contribution to the field. I anticipate that I can recommend the article for publication, after the following comments have been addressed:*

We appreciate the thoughtful comments, which have helped us to clarify and correct some important points.

*The authors description of the brittle fragmentation theory (Kok, 2011) is not quite correct. Contrary to what is stated on p.3503, this theory does not reconstruct the aggregated soil size distribution. Brittle fragmentation theory does not need (or assume) a particular undisturbed soil size distribution, and in fact hypothesizes that the emitted dust size distribution is independent of the aggregated soil size distribution. This confusion is repeated at several places in the article, and I think in the companion article as well, and it should be corrected throughout.*

Thanks for this correction. As the reviewer notes, the theory provides the size distribution of the emitted dust aggregates without reconstructing the distribution of the original undisturbed soil. The cumulative term accounts for the range of indivisible scales within the soil. For the materials used to test brittle fragmentation theory in a laboratory, this scale corresponds to a single value that is on the order of the crystal dimension. In contrast, dust particles are comprised of aggregates of different sized particles that are approximately irreducible during mobilization and fragmentation. The cumulative integral sums over the range of indivisible scales identified from the fully dispersed soil. Aggregates formed from smaller irreducible particles will obey the scale-invariant power law of brittle fragmentation across a larger range of emitted particle diameters. The cumulative basically acts to attenuate this power law at the smallest sizes that is beneath the indivisible scale of the the larger irreducible particles. Conversely, the emitted distribution exceeds this power law at larger silt sizes because aggregates are comprised of particles with nearly the entire range of indivisible scales. (The reviewer describes this equivalently, saying that aggregates at a certain diameter are comprised of all the particles with indivisible scales below that diameter: eq. 3, Kok PNAS 2011).

All this is now described in Sect. 2.1.2 (especially p. 12), and additional corrections are scattered throughout the revised article.

*If I understand the methodology correctly, the authors are assuming that, except for iron* *oxides, each aerosol is composed of an individual mineral, even when it is an aggregate.*

Yes. We are interpreting the mineral fractions within each size category as the mass available for combination with other minerals. The size category corresponds to the dimensions of the aggregate rather than the diameter of the individual minerals contributing to the aggregate. This interpretation is consistent with the size distribution of each mineral measured by Kandler et al. (2009), where particle size refers to the aggregate.

This interpretation allows for an economical representation of mineral aggregates. The economy results from the fact that we don't explicitly track individual aggregates. Instead, we represent the minerals as an external mixture, assuming that they all have identical (or approximately identical) removal rates. The mineral fractions are interpreted as an average over all particles within the size category. Only for iron oxides do we explicitly represent mineral combinations. This is now discussed explicitly and in detail on p. 9 (last paragraph) through the end of the subsection.

*That is, they scale the production of aggregated silt-sized aerosols of a given mineral by the prevalence of clay-sized particles of that same mineral. I would think that, in reality, the probability that a clay-sized particle ends up in a silt-sized aggregate depends on (among many other factors) on the fraction of other small (clay- sized?) particles in the soil, not just of the same mineralogy. Why did the authors make this assumption, and how do they expect it to affect their results? Some justification needs to be provided. Does this simplification explain some of the disagreements with measurements?*

According to eq. 3 of Kok (PNAS 2011), the number of aggregates of diameter $D$ scales with the volume fraction of indivisible particles with sizes below or equal to size $D$. The physical interpretation of this equation does not depend upon the identity of the minerals forming the aggregates, so we apply it to each mineral separately. Our application to individual minerals is consistent with the method used by Scanza et al. (2015). This is described in p. 18 ("We also assume that the emitted mass fraction of each mineral $n$ at silt sizes consists of two contributions from the wet-sieved soil ...")

*I also have a few minor comments:*

*Line 24-25 on p. 3500: The cohesive forces actually increase with particle size (e.g., Shao and Lu, 2000). What the authors probably mean to say here is that the cohesive forces per unit weight (or per unit surface area, which is more relevant for particle lifting) decreases with particle size.*

We have revised our discussion to incorporate this qualification along with a citation to: Shao, Y., and H. Lu (2000), A simple expression for wind erosion threshold friction velocity, J. Geophys. Res., 105(D17), 2243722443, doi:10.1029/2000JD900304.

In Section 2.1.1 (p. 8): "Larger soil grains or aggregates are more easily lifted because this cohesion can be overcome by the wind stress acting over a larger area (Iversen and White 1982; Shao and Lu 2000."

*Line 17 on p. 3503. Sow et al. (2009) actually argue that the size distribution depends significantly on wind speed. In addition to Gillette et al. (1974), better references here would be Shao et al. (2011) and Kok (ACP, 2011). Please correct that here and in the companion article (if appropriate).*

We have incorporated this clarification in Sect. 2.1.2 (p. 11): "However, measurements suggest that for the smallest particles that are transported globally, the emitted size distribution is approximately independent of wind speed and soil properties (Gillette et al., 1972, 1974;

Gillette, 1974; Kok, 2011a, Shao et al., 2011)."

*Line 25 on p. 3503. Reconstructing the aggregated soil size distribution from the fully dispersed one is very difficult and Im not aware of any theoretical models for this. I looked at the cited Shao (2001), but could find no reference to such a model. If this reference does present the (otherwise reasonable) assumption that the amount of aggregation scales with the clay fraction, can you provide a page number?*

Our citation of Shao was based upon a misinterpretation of the cumulative integral of the fully dispersed size distribution, as described above. The lesson we are now deriving from Shao's Figure 4a is that clay size particles in the fully dispersed soil were originally present as aggregates at much larger diameters (between roughly 20 and 300 um). These aggregates are large enough to be entrained directly into the saltating layer by the drag of the wind. According to brittle fragmentation theory, the emitted size distribution includes aggregates that would be destroyed during measurements of the fully dispersed soil.

The assumption that the amount of emitted aggregates scales with the fully dispersed clay fraction derives from eq. 3 of Kok (PNAS 2011). It reflects the effect of the cumulative integral when considering different soil textures (and therefore different distributions of indivisible scales).

We have corrected the discussion in Sect. 2.1.2 (especially p. 12) to reflect these points.

*I think d is a confusing variable name for the mass fraction, as many readers will inherently associate this with particle diameter. I suggest using a different variable name.*

This is a good suggestion. We have replaced "d" (intended to indicated dust) with "a" for aerosol (in contrast to "s" for soil). This necessitated finding another variable name for soil type. We now refer to soil type and texture, respectively, using "$\alpha$" and "$\beta$". (c.f. Table 5)

*Since there are no strong constraints on the global dust emission rate, I think it makes little sense to force the total emission rate equal to some somewhat arbitrary figure (2224 Tg/year in this case). For future studies, I would suggest setting the dust emission proportionality constant either by forcing the global dust AOD to some constant value (since we do have constraints on that), or by calibrating to some observational data set such as AERONET.*

Our original submission unintentionally gave the impression that we had some reason for choosing 2224 Tg of global annual emission. In fact, this value was a historical accident. Our comparison to observations is based upon mineral *fractions* that are independent of the global annual emission, and thus, we made no effort to adjust the global value. In the revised version (Sect. 2.2.3), we are explicit that the value of 2224 Tg is irrelevant to our comparison, which does not depend upon a particular value (p. 27, top).

Nonetheless, the reviewer raises a more general question about how to compare two simulations in a meaningful way. Despite identical global emission, the SMF and AMF experiments have emitted size distributions that are markedly different. Thus, the experiments have different particle lifetimes and thus different aerosol loads, resulting in different degrees of fidelity to observations. For this comparison, we agree with the reviewer that we should separately calibrate the global emission of each experiment using observations. Unfortunately, it is beyond the scope of this study to simulate the radiative effects of the different minerals (including their individual contributions to the total dust AOD). We have kept this caveat in mind in Section 4, where we compare the AMF and SMF size distributions and lifetimes.

*Im a bit confused by the term accreted. Is this the same as aggregated? Can you give exact definitions of both terms?*

We recognize the potential confusion identified by the reviewer. We were attempting to avoid confusing the aggregation of minerals with iron oxides with our frequent references to disaggregation of clays by wet sieving. In the revised version (top of Sect. 2.2.2, p. 22, top), we explicitly note that we are using two distinct terms to discourage this association.

*Why does this article include a comparison to data at Tinfou, Morocco when part 2 contains an extensive evaluations against measurements? I think this needs some justification at the beginning of section 4.5.*

We have two reasons for bundling the comparison of the size distribution at Tinfou to this article (that is otherwise a model description). First, the measurements at Tinfou are exceptionally detailed, allowing a more extensive model evaluation than is permitted by the other observations that are compared in the companion article. However, our main reason is that the comparison highlights some limitations of our method, and the need for future improvements. We felt that this discussion fit best in this article, whose focus is on the method. In the revised version (Sect. 4.5, p. 38 bottom), we try to motivate this organization. Nonetheless, we acknowledge that this is a somewhat arbitrary choice.

*Figure 15: keeping the color scheme consistent for panels e-h would make the figure more readable.*

We think this is generally good practice, which we have applied to the preceding figures (formerly Figs. 11-14, now Figs. 10-13). The problem for this particular figure is that quartz and iron oxides have a much smaller range of values compared to the ranges for feldspar and gypsum. We were not able to figure out how to choose a color scale that can incorporate both ranges while showing regional variations in detail for all minerals. In response to the reviewer request, we instead note explicitly in the figure caption for Fig. 14 (the former Fig. 15) that the color scale varies among the minerals.

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**Reviewer 2**

*The paper documents and preliminary evaluates the new methods of explicit calculation of dust aerosol mineral composition incorporated in the GISS ModelE. That was never done in full scale so far and is of great importance as dust mineralogy and chemical composition defines radiation effects of dust particles, their impact on atmospheric chemistry, and clouds. Currently models calculate dust size distribution but assume globally uniform mineral composition of dust particles. The authors show that the proposed approach gives results consistent with a few available observations of dust mineralogy. However, the simulations in this study do not account for radiative feedback of aerosols. It would be interesting to provide more detailed evaluation that would estimate the new approach from this point of view. The paper is well written and could be published after minor corrections.*

At present, we don't account for radiative effects, whose representation within the model would require additional development and assumptions (about mixing rules and particle morphology, for example). Nonetheless, we agree that this will be a useful direction for future work, especially given the abundant measurements and retrievals of aerosol radiative diagnostics like optical thickness. (We are explicit about our neglect of radiative effects at the end of Sect. 2.3 at the top of p. 28.)

*P. 3500, L 27-29: please clarify the sentence.*

We have tried to make clear that the smaller soil particles that become aerosols are bound tightly to the soil by cohesive forces, and are unlikely to be lifted directly by the force of the wind. Instead, these smaller particles enter the atmosphere either because of the impact of a larger particle (whose kinetic energy overcomes the cohesive force) or because a larger particle disagregrates into a collection of smaller particles.

On p. 8, we write "Most of the smaller particles that are transported globally are entrained into the atmosphere during the fragmentation of aggregates that are bombarded by larger particles, or else are large enough to be lifted directly by the wind and disintegrated through repeated collisions (Shao et al., 1993; Kok, 2011b; Marticorena, 2014)."

*P. 3504, L 5: You mean below 20 micron?*

We have rewritten this section (2.1.2) to provide a more precise description of brittle fragmentation theory. According to this theory (Kok PNAS 2011), soil aggregates are distintegrated by repeated collisions resulting in particles with diameters that are almost entirely below a scale $\lambda$. Kok estimates this scale to be around 12 um, while noting that is an approximate upper bound. In our model, we assume that brittle fragmentation is the process that controls emission for particle diameters up to around 20 um.

In the revised version, we have elaborated our discussion of brittle fragmentation theory to address these points. This discussion starts on p. 11. At the bottom of p. 13, we note our assumption that "The specific range of validity is below 20 um."

*P. 3505, L 1-2: Could you give a reference here?*

This model behavior described by this sentence is noted to be consistent with the size distributions of each mineral in Fig. 1, which we note throughout the article is derived from measurements by Kandler et al. (2009).

*P. 3506, L 10-18: Validity of the approach*

At the end of Sect. 2.1.2 (last paragraph on p. 14 to the end of the section), we discuss approximations involved in our use of an empirical distribution to prescribe the size distribution of emission within the silt size range. As noted in the text, we can only demonstrate the validity of this approach by using a more complicated model of emission that explicitly represents saltation and sandblasting. However, we note that the increase of emission with particle diameter is likely to be a robust consequence of the decreasing threshold for emission with diameter: "The increase of the emitted silt fraction with increasing particle size (Fig. 3, second panel from left) is probably a robust consequence of the wind speed threshold for emission that decreases with diameter within this size range (Iversen and White, 1982b)."

*P. 3506, L 21-25: What would be if we take an external mixture?*

We don't understand this comment. We are treating the different minerals as an external mixture. We have provided an expanded discussion of this point in Sect. 2.1.1 (the last paragraph on p. 9 through the end of the section).

*P. 3509, (8): No, if (8) satisfies, then (6) will satisfy. Not vice versa.*

We believe that any two of the three equations (6)–(8) are sufficient to establish the third.

*P. 3509, (9) and (10): It is the same assumptions as in GOCART scheme.*

On p. 13, we extend the comment of the reviewer to note that many current models of dust emission specify the emitted size distribution to be spatially uniform.

"The prescription of an emitted size distribution that is independent of location is shared by studies of the global dust cycle that do not resolve mineral variations (e.g. Miller et al., 2006; Albani et al., 2014). This approach has also been used by Scanza et al. (2015) to account for the effect of brittle fragmentation upon the aerosol mineral composition."

We reiterate this point on p. 18 in the context of the two equations cited by the reviewer. "As noted above, the assumption of an emitted size distribution that is spatially uniform is shared by many models, including those with uniform or else varying mineral content (e.g. Miller et al., 2006; Scanza et al., 2015)."

*P. 3509: (11) just defines the amount of clay particle aggregated to silt sizes. What was the reason to talk about corrected size distributions? Are they consistent?*

The reviewer is correct. We are 'correcting' the size distribution of the wet-sieved soil to create the aggregates that would be emitted after brittle fragmentation. This is consistent with the representation of Kok (PNAS 2011) that is described in Eqs. (1) and (2). However, instead of using a cumulative integral like Kok (Eq. 2), we restore the aggregates using a heuristic representation whereby the emitted silt includes a contribution from clay particles in the wet-sieved soil.

This is expressed on p.18 just above Eq. (13): "We also assume that the emitted mass fraction of each mineral n at silt sizes consists of two contributions from the wet-sieved soil. The emitted fraction combines soil mass at silt sizes along with clay particles whose aggregates were broken during wet sieving. This is expressed by Eq. (2), but we represent reaggregation more simply by augmenting emission at silt sizes in proportion to the fractional abundance of clay particles in the fully dispersed soil:"

*P. 3510, L 21: Change predominately to predominantly*

Done.

*P. 3520, L 10-11: It is difficult to believe that this distribution is invariant.*

We share this reservation for the larger size particles that are not subject to brittle fragmentation, and note this point explicitly. However, it is difficult to assess how much the distribution varies, given the paucity of size-resolved concentration measurements for individual minerals. As noted above, we discuss this approximation more fully in Sect. 2.1.2

(last paragraph on p. 14 to the end of the section).

*P. 3531, L 23-26: It is consistent with the larger radiation effect of coarse fraction in comparison with a fine dust fraction discussed in the literature.*

Our point here is that an increase at one size has to be compensated by a decrease at another, because the emitted size distribution of silt is based upon a normalized distribution derived from measurements. We have tried to express this more clearly in the final paragraph of Sect. 4.5 (p. 40).

*P. 3532, L 24: Change crytalline to crystalline*

Done. (Despite the kind efforts of the reviewers, we have probably introduced other typos during revision :)

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## Additional Comments by Zhang Xuelei, 1 March 2015

*Here, I have four viewpoints for the authors and the editor to improving this paper.*

*1. Mineral composition has questions. This manuscript adopted the relative volume abundance of minerals deduced from single particle analysis to calculate the mass distribution as a function of particle size for common airborne minerals in Figure 1. The original data is from Table 1 of Kandler et al. (2009) with identified specific mineral phases. In Kandler et al. (2009) used the method in Kandler et al. (2007) to classified the mineral phases, but this method only identified the whole silicate (which could not further classified as illite, kaolinite and chlorite) and Kandler (2009) also written as All silicates except quartz are sorted into the silicates class, as the different silicate minerals cannot be distinguished from each other reliably by the elemental composition only. in section 4.3.2.1 line 11-14. The content of Table 1 in Kan2dler (2009) is low quality and unusable. This question directly influence the second challenge mentioned in Page 3501.*

We agree that Kandler et al. assign identical size distributions to phyllosilicates and feldspars (as we noted in our original submission). We disagree that this makes Kandler et al.'s measurements 'low quality and unusable'. In general, the quality of measurements is meaningfully assessed only with respect to their specific application. Our application is to partition the silt fraction given by Claquin et al. (1999) into the size categories used by our transport model. We speculated in our original submission how the contrast between the size distribution of phyllosilicates and feldspars would alter our results. (In the revised version, we have expanded our discussion of the implications of any contrast. See the final paragraph of Sect. 4.5, p 40, along with the final paragraph on p. 42.). However, the commenter does not explain why this contrast would undermine any of our main conclusions.

*2. iron oxides: The reported hematite abundances in Table 1 of Kandler et al. (2009) were obtained from the measured content of elemental Fe by EDX, and then each value multiply 20 percent. Thus, the result is almost similar values (0.5 percent-0.8 percent) in different size bins. To my experimental experience, the size of hematite particle is presented as particles less than 10 um. But the iron-rich particles indentified by individual particle analysis actually are the assemble of hematite, goethite, wuitite and so on. These iron-rich particles is the so called pure crystalline form and as an external mixture with other minerals. For hematite with nanometer sizes which attached to phyllosilicates is hard to be identified by individual particle analysis, and this attachment is also the external mixing. Because the internal mixing is meaning that a small particle was coated or partly immersed into a larger particle. Once nano-sized hematite particles attached to phyllosilicates would not effectively change size of silicates. The volume contribution is mainly from the iron-rich particles with micrometer sizes. As mentioned above, the usage of volume fraction of hematite in Fig. 11 of Kandler et al. (2009) is more plausible for calculation in Fig 1 of this manuscript. 2. The usage of internal mixture and external mixture in this paper is easy to comprehend as terms for calculating the complex refractive indices for optical properties of mineral mixtures.*

High resolution images show that aerosols comprised of iron oxides are present as both pure crystals and as accretions on or within other minerals. In our model, we represent both forms because of the potential importance of this distinction to radiative forcing by iron oxides. However, we admit that our treatment is relatively simple in the absence of much guidance from observations. Our general strategy for representing iron oxides is discussed in

Sect. 2.1.1 starting on p. 10.
*3. settling speed: As mentioned in page 3519 line 1-3, Settling speeds are proportional to mineral density, and the minerals have nearly identical densities except for iron oxides. Quartz has similar density (2.67 g/cm3) with compare to phyllosilicates (such as Kaolinite with 2.67 g/cm3), this means that both quartz and kaolinite have almost the same settling speed. Phyllosilicates with flake shape is easy fly in air than quartz, and the difference of morphology will obviously affect the settling speed (Li and Osada, 2007). If possible, the effect of morphologies of different mineral particles on their settling speeds should be taken into account in future works.*
In general, dust aerosol models do not account for morphology in their representation of gravitational settling, and we have yet to add this effect. We appreciate the reference to Li and Osada (2007), who show that dust particles that most likely originated within Chinese deserts are largely spherical by the time they are observed in Japan. Li and Osada interpret this to mean that variations in particle morphology and their effect upon dry deposition are mainly important only near the source region. We have added this citation and a brief discussion to p. 10.
*4. The core of the paper is built on the brittle fragmentation ¿ theory of Kok et al. (2011).* **Once the core collapsed, and this manuscript will be collapsed.** *Many models simulate the dust size distribution as a sum of lognormal modes (e.g., Balkanski et al., 2007 and Zhao et al., 2010). This approach is computationally efficient for...*
Kandler et al. show that for clay minerals like illite, a large fraction of the aerosol mass is present at diameters larger than 2 $\mu$m, even though Claquin et al. (1999) find that within wet-sieved soil samples, clay minerals are present only at smaller sizes. (Kandler et al. use XRD to show that the total mass of clays and feldspars are comparable, so the presence of clay minerals at silt sizes cannot be explained by the misattribution of feldspars, even though the size distributions of the two minerals cannot be distinguished.) These measurements (and observations from other studies cited in the text) point to the need to consider silt-sized aggregates of clay minerals that are emitted as aerosols, but which are not present in measurements of the wet-sieved soil (a point also discussed by Claquin et al. 1999 and Shao 2001). We choose to calculate emitted aggregates using Kok's theory of brittle fragmentation. The commenter subsequently claims that other representations of the emitted size distribution provide a better fit to data, but assessing this claim is beside the point of our article. (In any case, brittle fragmentation theory has been shown to be consistent with a widening set of that have become available after Kok's initial study.) Our interest here is simply to relate the size distribution of the wet dispersed soil to the emitted size distribution. Again, aerosol measurements clearly show the presence of phyllosilicates at silt sizes, and our model needs to reproduce this behavior. We expect other representations of the emitted distribution proposed by the commenter would change our results only quantitatively.