"Predicting the mineral composition of dust aerosols. Part 2: Model evaluation and identification of key processes with observations" by Perlwitz et al.

Dear Yves,

As with the companion article, we are grateful for the thoughtful comments of the reviewers and we appreciate the extra time to make substantial revisions in response. Some of the comments have also been used in the companion article to expand the model description or clarify our discussion of the results.

Our response to each of the reviewer comments below contains a 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 also 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

Response to the Comments by Reviewer #1

(The pages and line numbers to which Reviewer #1 refers seem to correspond to the version submitted after the technical review (but before publication in *Atmos. Chem. Phys. Disc.*). We have listed in **bold** the page and line numbers that we believe correspond to the discussion version.)

General comments

This study addresses modeling and validation aspects of dust mineral composition. The subject of the research is crucially important for better understanding the multiple roles of dust in climate system. The authors conduct the analysis using global model with nudged wind fields and exploit the available observations and soil databases. They test two clear hypotheses about the mineral emission fractions. The main problem is that the observational base is relatively poor both for soil parameters database, which uses observations with insufficient spatial resolution, and for aerosol observation that are sporadic in time and space. However, we have to admit that this is the best available information at a time. It is important to start working in this direction. The global approach has advantage, as it allows incorporating available observations all over the world. However, it is low resolution and is very poorly supported by observations in the Southern Hemisphere. Similar regional studies have to be encouraged in future. The minor comments are given below.

Thanks for these supportive remarks. We agree that there are large uncertainties in the data base for the soil mineral fractions, which are used as input for our simulations, and that our evaluation would benefit from a far greater number of measurements. Like Claquin et al. (1999), we accepted these uncertainties because we believe that mineral composition is fundamental to how dust aerosols interact with climate. We hope that our work draws the attention of scientists who do measurements, who can offer improved contraints upon the models.

Minor comments

I would suggest the authors once more report their emission scheme. It would be more convenient for a reader to have this information just in the text but not in the references.'

As suggested by the reviewer, we have added a new section summarizing the dust aerosol module, including the calculation of emission (Sect. 2.2, starting on p. 8).

P 3, 25-30: It sounds like the most important assumptions are semi-hypothetical. Could you elaborate on this and add explanations. **(Page 3580, lines 1-6)**

We have tried to identify more precisely in Sect. 2 (with full details in the companion paper: Perlwitz et al., 2015) where our method depends upon hypotheses or empirical representations suggested by measurements. For example, we note that the allocation of silt emission into the size categories transported by the model is based upon measurements at a single location (as noted below) and we discuss the validity of this representation (p. 6, last paragraph).

P 4, 1-10: Dust particles will be processed in the atmosphere both microphysically and chemically. You have to clearly discuss this and mention what model actually accounts for. **(Page 3580, lines 7-16)**

We do not account for this process in the current version of the model. Following the referee's suggestion, we explicitly note this in Sect. 2.2 (p. 9, penultimate paragraph). We have also added this point to the companion article.

> "These modifications, which depend upon the mineral composition, alter the solubility and vulnerability of the dust particle to wet scavenging. We defer representation of this dependence to a future study and assume the solubility of each dust 25 particle to be constant (50%) and identical for each mineral (Koch et al.,

1999)."

P 4, 20: I do not think it is good idea to nudge winds at all levels assuming, e.g., that the surface flow is strongly controlled by the topography and it is different in the GISS ModelE and NCEP reanalysis. I would nudge in the boundary layer. Is nudging coefficient altitude dependent? Please elaborate on this issue. **(Page 3580, line 27)**

The reviewer raises an interesting point. However, the NCEP/NCAR reanalyses toward which we nudge have horizontal resolution of $2.5° \times 2.5°$ latitude by longitude, which is similar to the resolution of ModelE2. In practice, we don't expect that the difference between the topographies should be large. In response to the reviewer, we note in Sect. 2.3 (p. 10) that the nudging time scale is independent of height and equal to 100 s (and we also add this information to the companion article). We would be grateful to be pointed toward any study where nudging of winds only within the boundary layer provides better agreement with measurements.

> "The horizontal winds at each level of the model are relaxed every six hours toward the NCEP reanalyzed values (Kalnay et al., 1996). Relaxation occurs at all model levels (up to 10 hPa) with the globally uniform time scale of 100 s."

P 4, 25: Aerosol optical depth is the most important observed/retrieved characteristic. There should be some comparison included. **(Page 3581, line 6)**

We agree that this would be a useful exercise. However, in the present version of the model, we do not use the mineral fractions to calculate AOD because this task (along with model evaluation) requires substantial effort, and is deferred to a future study. We note this emission explicitly in the revised manuscript (Sect. 2.2, last paragraph on p. 9).

> "We also defer calculation of radiative forcing as a function of the aerosol mineral composition. As a result, radiative feedbacks between the mineral fractions and

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climate are disabled."

P 5, 3: Change "will be" to "are" **(Page 3581, line 10)**

Done. (Top of Sect. 2.1, p. 5)

"Two simulations are compared to our compilation of observations."

P 5, 5-10: MMT covers the entire world but how many observations they really had to build those mineralogical fields?' **(Page 3581, lines 12-18)**

The MMT is based on 239 "descriptions of soils", covering dry areas from the United States, Israel, Australia, North Africa, China, Iran, Iraq, and Northern India (Claquin et al., 1999). We are not sure whether "description" refers to soil samples or identifications of individual minerals (or something else), so we have not added this information to the article. Claquin et al. emphasize the uncertainty remaining in the MMT, which motivated subsequent refinements by Nickovic et al. (2012) and Journet et al. (2014).

P 5, 10-15: Is there any physical bases why we can assume that iron oxides are equally abandoned in clay and silt fractions? Could you please clarify this issue. **(Page 3581, lines 18-22)**

There are only limited size-resolved measurements of iron oxides and their fraction in the soil (e.g. Journet et al., 2014). Claquin et al. (1999) infers the soil fraction of iron oxides from color rather than measurements at specific particle sizes, and assumes that iron oxides are present only at silt sizes. However, subsequent aerosol measurements show that iron oxides are present at both clay and silt sizes (Lafon et al., 2006; Kandler et al., 2007; Engelbrecht et al., 2009; Jeong et al., 2014). Consistent with Nickovic et al. (2012), we assume that the iron oxide fraction at clay sizes is identical to MMT value at silt sizes

prescribed by Claquin et al. (1999). We are not aware of measurements that would allow a more precise distribution of iron oxides with respect to particle size. We discuss this in Sect. 2.1 at the bottom of p. 5.

> "According to the MMT, hematite is present in the soil only at silt sizes. Aerosol measurements show this mineral to be present at both clay and silt sizes (Lafon et al., 2006; Kandler et al., 2007; Engelbrecht et al., 2009; Jeong et al., 2014), so we extend the size range of emitted hematite to include clay sizes. Given the limited measurements of this mineral in soil samples, we follow Nickovic et al. (2012), and assume for simplicity that the hematite fraction at clay sizes in identical to the silt fraction provided by the MMT."

P 5, 20-25: Is it observations from one place used to verify the distribution of minerals over the size bins? Please comment on this and clarify in the text. **(Page 3581, line 28 to page 3582, line 5)**

Yes. For the AMF experiment, we use measurements at the single location of Tinfou, Morocco by Kandler et al. (2009) to derive a normalized distribution of mass for each mineral within the silt size category (with diameters between 2 and 50 μm). This distribution is used to apportion the emitted silt fraction into the size categories of ModelE2 at every source region. The distribution is normalized so that it is independent of the specific magnitude of emission at Tinfou. We assume that the (normalized) size distribution is invariant with respect to source region. There are few measurements that would allow us to evaluate this assumption, but we think that the emission increase with particle diameter is probably robust (Sect. 2.1, p. 6, last paragraph).

P 5, 25-26: Please clarify the sentence about "gravitational setting". **(Page 3582, line 6)**

This is a typo that was intended to say "gravitational settling", i.e., the falling out of dust particles due to gravity (p. 6, line 13).

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"Dust at Tinfou is measured after transport from the source, when the largest particles are removed preferentially by gravitational settling."

P 6 10-15: It is not empirically based, it is just an assumption. Could you explain. **(Page 3582, lines 19-24)**

We agree, and now describe our reconstruction as a "heuristic" representation, which we believe is more descriptive than "empirical". We are expressing the idea that fully dispersive measurements of the soil destroy aggregates that would have persisted during mobilization and brittle fragmentation. Thus, the emitted silt fraction contains silt particles present in the wet-sieved soil, but also aggregates comprised of wet-sieved clay particles. In Kok (2011), the number of aggregates of diameter D scales with the volume fraction of soil particles with sizes below or equal to this diameter. This expression predicts that the fractional contribution of clay soil particles to the emitted silt mass scales with the soil clay fraction. This is the behavior that we are representing heuristically by allowing wet-sieved clay particles to contribute to the emitted silt fraction in proportion to our coefficient γ (Sect 2.1, p. 7).

"In the AMF simulation, we reaggregate these fragments heuristically. For each mineral, the emitted silt fraction is comprised of silt particles in the wet-sieved soil augmented in pro- portion to the mineral's wet-sieved clay fraction . . . ."

P 6, 18-21: Please clarify both about the constrain and about transformation of mineral fractions. **(Page 3582, line 18 to page 3583, line 3)**

We deleted the word "transformation". What we are trying to express is the *contrast* between the SMF and AMF mineral fractions. This contrast results from accounting for aggregates that are potentially emitted from the original soil, but destroyed by wet sieving (Sect 2.1, p. 7).

"The emitted silt fraction consists not only of silt particles present in the wet-sieved soil, but also aggregates that were broken during wet sieving into clay-sized fragments. In the AMF simulation, we reaggregate these fragments heuristically . . . ."

P 6, 22-25: Please justify this assumption. **(Page 3583, lines 3-5)**

The extension of feldspar and gypsum to the clay-sized range, which are absent in the MMT in this size range, is justified by the fact that these minerals are present at this size in aerosol measurements (Leinen et al., 1994; Arnold et al., 1998; Kandler et al., 2007, 2009). We have added a justification and references for the extension in Sect. 2.1 (p. 7, bottom).

"Conversely, the MMT provides the fraction of feldspar and gypsum only at silt sizes, even though aerosol measurements show that these minerals are present at both clay and silt sizes (Leinen et al., 1994; Arnold et al., 1998; Kandler et al., 2007, 2009)."

P 7, 14-16: Does this emitted mass in your model produces reasonable dust optical depth? It would be useful to mention this in the text. **(Page 3583, lines 25-28)**

In the current version in the model, we haven't calculated the scattering and absorption of radiation by the mineral tracers. We agree that the aerosol optical depth should be evaluated. However, this is both beyond the capability of the current model and the scope of the current study. The focus of this study is the evaluation of the model mineral fractions, which is independent of the scaling of the absolute dust mass and the dust aerosol optical depth. Our model emission is within the range calculated by other models, which gives some confidence that the dust AOD in the new model won't be outside the current range of uncertainty. We intend to calculate the radiative effect and evaluate the model AOT in a future study. In

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Sect. 2.2 (p. 9, last paragraph), we note the omission of any radiative perturbation by the minerals.

> "We also defer calculation of radiative forcing as a function of the aerosol mineral composition. As a result, radiative feedbacks between the mineral fractions and climate are disabled."

P 9, 6-25: XRD and SEM have a disadvantage to be more sensitive to the particle surface layer that could be affected by coating. **(Page 3585, line 21 to page 3586, line 12)**

Thanks. We have added this caveat (p. 12, line 23).

> "Both XRD and SEM measurements are disproportionately sensitive to composition on the particle surface, which may include coatings resulting from chemical reactions with other species, compared to the particle interior."

P 14, 21-25: Isn't it directly follow from our assumptions for the SMF and AMF emissions? **(Page 3591, lines 15-19)**

The result discussed by the reviewer is the overestimate of the quartz fraction at silt sizes by the SMF method, with improved agreement within the AMF experiment (p. 18, around line 15 of the current draft). The reviewer seems to be asking whether this improvement is inevitable. We believe that this is not the case.

The smaller quartz fraction in the AMF experiment compared to the SMF simulation indeed follows directly from accounting for reaggregation in the former (that increases phyllosilicate emission at silt sizes at the expense of the quartz fraction). However, it is *not* inevitable that the AMF value is closer to the observed value. For example, the MMT could have indicated a quartz fraction at silt sizes in the SMF experiment that is consistent with the observed value. Then, reaggregation within the AMF simulation would still decrease the quartz fraction, but move it *away* from the measurements. The fact that the SMF value is an underestimate

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shows that accounting for phyllosilicate reaggregation is necessary. That the combination of the MMT and reaggregation of phyllosilicates (i.e. the AMF method) results in approximately the observed value suggests that the MMT quartz fraction is approximately correct.

P 15, 15-17: It would be useful to take more about how atmosphere could process dust particles. **(Page 3592, lines 9-12)**

We have expanded the discussion in Sect. 2.2 about physical and chemical transformation of dust particles during transport and our current neglect of these processes (p. 9, penultimate paragraph).

> "Measurements show that physical and chemical properties of aerosols evolve along their trajectory (cf. Baker et al., 2014). . . . These modifications, which depend upon the mineral composition, alter the solubility and vulnerability of the dust particle to wet scavenging. We defer representation of this dependence to a future study . . . ."

We have also added this discussion to the companion article. In the conclusions (p. 26, last paragraph), we briefly note our future plan to incorporate these processes.

P 18, 20-25: It would be useful to discuss what physical processes could affect this ration. E.g., it can not change within one size bin, I believe. **(Page 3595, line 20-25)**

We add a brief discussion to the second paragraph of Sect. 5.3 (p.21, line 4). For minerals other than pure crystalline iron oxides, the ratio with respect to quartz is relatively constant along a trajectory, as noted by the reviewer.

> "Like mineral fractions, mineral ratios will evolve downwind of the source region . . . ."

P 21, 13-20: What does control the emission of small particles? Is it availability of clay fraction in the soil layer or it is hydrodynamic entrainment that is less effective for small particles? Please explain. **(Page 3598, lines 14-22)**

Aerodynamic entrainment is inefficient for clay-sized particles because the cohesive force per unit area is large. These small particles mainly enter the atmosphere through bombardment by larger particles or else disintegration of larger particles. Kok (2011) gives a physical argument as to why the mass distribution of emitted particles is weighted toward larger sizes. The point that we wanted to reiterate in the conclusions is that the soil texture data are based on soils that are fully dispersed by wet sieving. This dispersion is much more destructive of soil aggregates than saltation and sandblasting which (according to measurements and brittle fragmentation theory) allows some of the smaller (silt-sized) aggregates to persist as aerosols. This is discussed in the opening paragraph of the conclusions (p. 23).

> "Minerals like phyllosilicates that are aggregates of smaller soil particles are almost exclusively observed at clay sizes after wet sieving, despite aerosol measurements showing greater phyllosilicate mass at silt diameters (eg. Kandler et al., 2009). This suggests that many of the aggregates that are destroyed during wet sieving would resist complete disintegration during wind erosion of the original, undispersed soil. "

There is also a fuller discussion in Sect. 2.1.2 of the companion article.

P 23, 13-17: Aging of dust is an important process especially for iron oxides. Do you account for it? **(Page 3600, lines 18-25)**

No; this is one of our next projects. We note our general neglect of particle transformations (but not for iron specifically) in Sect. 2.2 (p. 9, penultimate paragraph), the companion article, and briefly within the conclusions (p. 26, last paragraph).

Response to the Comments by Reviewer #2

This article provides an extensive evaluation of the model for size and mineralogy- resolved dust emission presented in part 1. The authors construct an impressive compilation of measurements, from about 60 studies, making for a detailed evaluation. I have only a few comments, and recommend that the article be published after the authors address them.
It would be helpful to include a comparison against the results of Scanza et al. (2015), who use some of the same measurements to evaluate their model for mineralogy-resolved dust emission. Can the authors discuss the effects of the purported improvements over this recent study? Do the additional processes they include actually im- prove the simulation?

We have done calculations to understand the difference between the two methods. However, we feel that this comparison is beyond the scope of the current article, so we are planning a future manuscript on this topic.

I found the discussion section quite tedious to read, in part due its length. I would suggest improving the writing in this section, for instance by adding more sub-headings.

We appreciate this suggestion. In response, we have separated the results presented in Section 5 into a number of subsections that are limited to the discussion of a single figure or topic. We hope this new organization makes it easier for the reader to recognize our main points.

Response to the Comments by Paola Formenti

Authors shoudl include the following paper for data comparison : Formenti, P., Caquineau, S., Desboeufs, K., Klaver, A., Chevaillier, S., Journet, E., and Rajot, J. L.: Mapping the physico-chemical properties of mineral dust in western Africa: mineralogical composition, Atmos. Chem. Phys., 14, 10663-10686, doi:10.5194/acp-14-10663-2014, 2014.'

Thanks for pointing us to this paper. We noticed that the surface concentration measurements at Banizoumbou, Niger, which are analyzed in that paper are the same data as in Formenti et al. (2008). Those measurements are included in our evaluation. We have cited the more recent study in the conclusions (p. 26, line 1):

> "These refinements can be complemented with studies that map the mineral composition of specific sources (Formenti et al., 2014b)."

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