

## ***Interactive comment on “Modeling the global emission, transport and deposition of trace elements associated with mineral dust” by Y. Zhang et al.***

**Y. Zhang et al.**

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Review comments are in black, while responses to the reviewer are in red. When text from the manuscript is quoted, new text is in bold face. The authors would like thank both reviewers for both their positive feedback, as well as their constructive criticism, which improved the manuscript. We revise the text as suggested by the reviewers.

Anonymous Referee #1:

1 General comments The modelling study by Zhang et al. that is presented here attempts to assess the effect of the spatial variability in the elemental composition of dust sources on the transport and deposition of trace elements by dust. Presently, the depo-  
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sition of trace elements is often calculated from bulk dust deposition, assuming a fixed elemental composition of dust; given that dust sources can differ quite dramatically in their elemental composition this is a significant progress and certainly justifies publication in Biogeosciences. The first step in the study by Zhang et al. is the compilation of a map of soil elemental composition using a high resolution soil data set from the FAO, then estimating the fractions of different minerals in the different soils, following Claquin et al. (1999) and Nickovic et al (2012), finally combining them with data sets on the elemental composition of different minerals from the literature. As the authors acknowledge, the assumptions on the mineral composition of soil types likely underestimates the variability present. The authors also note that impurities in gypsum, calcite and quartz can lead to variability in trace elements that is disregarded here. Very likely thus the spatial variability in dust source elemental composition is underestimated by the approach taken here; nevertheless, the first-order-trends are likely correct. For iron, similar attempts have been undertaken by Nickovic et al. (2013) (side-note: Only the precursor to that paper, Nickovic et al., 2012, is cited) and Journet et al. (2014), but the extension to more elements is a significant step that also allows a better validation. The second step is then to calculate the the emission, transport and deposition of this dust, using the Community Earth System Model that has already been widely used for dust transport modelling before. The novel aspect here is that the model now transports the different elements individually, so that at each point in space and time the elemental composition varies. Finally, in a third step the modeled elemental composition of dust and of dust deposition are verified by comparing them to a dataset of ground-based observation at a number of sites around the world. Although the results of the validation are somewhat mixed, the paper presents a significant step forward, and I think the paper should be published after suitable revision. However, before coming to my points of criticism, I'd like to mention that the whole paper is still written in an English that contains too many errors to list all of them at the end of this review, so I will limit myself to listing only a subset. In this form the paper cannot be published and I would urge the native English speaking coauthors to help the first author to rewrite it.

Response: We thank the reviewer for their comments, and have worked to ensure that the English is improved in the resubmitted version.

2 Main points of criticism For the review of the paper I have several main points that I would like the authors to answer:

Firstly, the transport of the different elements in the dust by the earth system model is applied to each element individually; in reality, the elements are bound together in different particles of variable composition. The assumption of individual elemental transport is likely to introduce some smoothing, i.e. an error. The situation is similar to that in marine ecosystem models with variable stoichiometry, where the variable stoichiometry of individual hytoplankton cells is mixed through by the ocean models advection and diffusion. For the latter case, Christian (2007) has examined the magnitude of the error introduced by that assumption (and generally found it to be handle-able); maybe the authors could have a look into that paper and come up with a similar argumentation? Response: In CESM, we treat eight elements as tracers in model, like dust. As the reviewer says, by splitting the dust into parts, we are introducing an error; usually most of this error comes from the advection algorithm itself. Of course, there is no advection algorithm that is mass conserving, monotonic, shape preserving and computationally efficient. There is quite a bit of literature on this issue, and we are using a state-of-the-art advection algorithm, which minimizes many of these issues. Because in our methodology we did not include every element that is in dust, we cannot explicitly examine the size of this error, but from other studies (including the one cited) we can assume it is not a large source of error for our calculation, but rather the errors associated with the source mineralogy is probably larger. We add in the following paragraph in the methods section: "By splitting the dust into its different mineral elements, we may add in additional numerical errors, because the elements are transported separately. There has been considerable work on improving advection algorithms in atmospheric models, and here we use the finite volume advection algorithm as part of the CAM [Lin and Rood, 1997]. While no advection scheme is perfectly mass conserving, mono-

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tonic, shape preserving and computational efficient, this scheme does a good job of balancing these multiple goals and maintaining strong gradients required in modeling atmospheric constituents (e.g. [Rasch et al., 2006]). Studies focused on elemental distributions in ocean models have suggested the relatively small uncertainties associated with these types of numerical errors (e.g. [Christian, 2007]), and compared with the errors in the source distribution of the minerals, errors from advection are likely to be small and are neglected here."

And we also add in a sentence in the conclusions highlighting that we think the soil map is the largest source of uncertainty in this study.

Secondly, the authors validate their model to a large extent with averaged elemental fractions in dust (line 21 to 23 on page 17505), but do not describe adequately how they calculated the average. Did they calculate the elemental fractions and then average those temporally for each location, or did they first average the amount of element and dust (or element and dust flux) at a location and then form the ratio of the two? And then, averaging the elemental fractions at the different validation sites, did they weigh the fractions by the amount of dust or dust flux? Depending on what you do the results may differ quite a bit. It would be interesting to see method of averaging affect e.g. the elemental fraction of iron (lines 8-10 in page 17506). Response: This is an important point, and we agree that we should discuss this more. We add in the following calculation into the section identified by the reviewer, and we add to each table and figure caption how the % is calculated to be clear. In this study we calculate the elemental fractions and average those temporally for each site. We have added the following paragraph into the paper to illustrate this point better: "For this comparison (above), we calculate the elemental fractions and average the fractions temporally for each site and compare to observations, but alternatively, we could average the elemental concentrations and divide by the elemental dust concentrations instead, and this will make a difference in our interpretations. For example, taking site 2-Tazhong, the averaged fraction is 3.5% when we calculate the fractions of iron firstly and average

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those temporally. However, when we calculate the averaged iron mass and dust mass separately, their ratio is 2.3%. For site3-Yulin, the ratio is 3.6% and 3.1% for the first method and second method, respectively. This difference maybe due to dust storm events. For this comparison, we use the first method, as we think it is more suitable for our goal of simulating the % of each element correctly. ”

Thirdly, the authors describe that the comparison between modeled and observed elemental fractions is not very good for two elements, namely magnesium and manganese (see e.g. the correlations in table 3). However, the authors do not discuss why that is. I suspect that it has to do with the uncertainty in the assumed average mineral composition (table 1); calcite e.g. often contains quite a bit of magnesium, but the assumed fraction in table 1 is zero. Maybe the authors could try to discuss the propagation of errors in table 1 onto their results a bit. Response: This is a very good question to answer. Originally we just discussed in paper separately with Table1 using “Underestimation of Mg and Mn could be due to a deficiency of minerals containing high concentrations of Mg and Mn in our model, as dolomite(MgCO<sub>3</sub>) or palygorskyte ((Mg,Al)<sub>2</sub>Si<sub>4</sub>O<sub>10</sub>(OH)·4(H<sub>2</sub>O)) are often identified in dust particles for Mg. Moreover, it is known that the chemical composition of minerals could be variable according to the regional origin of minerals and possible impurities. For example, the Mg content in calcite ranges from 0% to 2.7% in the natural environment.” Now we have added in P17507 with the sentence of “But in this study, the assumed fraction of Mg in Calcite is zero because we took Calcite as a pure mineral ( see Table 1). So the underestimation of Mg in dust could be a propagation of errors in previous compositions in minerals considered in this study”.

Fourthly, and most importantly, the authors use the ratio of the median elemental fraction in model and observations (documented in table 3) to 'tune' their results. This is a quite drastic step, and I wondered what the justification for that step is. I think the authors should give more reasons for this step than just the last line in table 3. Does it bring the models closer to the observations at all measurement sites uniformly? Does

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it reduce bias? What is the variance ratio between model and data? Response: For the tuning ratio, we aim to bring the models closer to the observations at all measurement globally, i.e. uniformly, so that downstream users (e.g. oceanographers), can use more accurate estimates. From observations, we have found a wide range in fractions of elements in individual site and all sites together, the ratio of the maximum and minimum in measured fractions could reach more than 700 for element K, and more than 200 for Ca and Mn. Using the ratio of averaged ones could introduce a bigger bias. So it is safer to choose a tuning ratio of medians from model and observation to adapt our model result. We add in a discussion of our motivation and the rationale better in the new text in Section 3.4. A better solution would be to solve the problem at the source, of course, which is why we highlight the problem (as discussed in the previous comment by the reviewers). We add text to better explain this point at beginning of Section 3.5.

And finally, at the present size many the figures resemble more a stamp collection and are almost completely useless to the reader. The authors should think about ways to present their results in a form that allows the reader to have a look without magnifying glasses.

We have revised the figures to make them more readable. Please note that an additional problem is that the discussion paper uses a square format, while the final paper will have the figures be rectangular, and thus will be larger using the format we use here.

3 An incomplete list of typographical, language and other errors The list of smaller language errors would quite long, and I have therefore not listed minor ones, such as omitted 'the' etc. Page 17497: many errors on this page; one example: line 27, 'calculating' should be 'calculation' Response: In updated manuscript the 'calculating' has been changed into 'calculation'.

Page 17498: What does the sentence 'Here the mineral dependent method is defined

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as M1' mean? I have no idea. Response: To compare the mineral method with Sillanpää method, we define the mineral method as Method 1. For the clarity, we have rewritten this sentence to "Here the mineral dependent method to calculate soluble element is defined as method 1 (Sol-1). To present uncertainties, the other approach (Method 2, defined as Sol-2) is introduced as reference. It is based on the extractable elemental fraction of in-situ 20 $\mu$ m sieved soil samples, reported by Sillanpää (1982) (Table S1) to combine with FAO soil dataset to get a global soluble elemental inventory independent of soil minerals".

Page 17502, lines 4-5: What does the sentence want to say? Response: We mean the global source areas are emitting dust with variable elemental fractions so differentiating in soil elements in source areas in model is necessary and meaningful. We have rewritten this sentence into "The simulated elemental fractions in dust suggest the differentiating in elements in soils between global source areas is necessary and meaningful" to make the clarity. Page 17502: Many small errors, like missing empty spaces between word, missing word like 'are found (in) dust' Page 17502, lines 23-25: what is a 'relative location'? I don't understand the sentence. The importance of something adds complexity in applying something else? Response: We have tried to fix the errors listed here and other similar errors in the updated manuscript. Page 17503, lines 15 ff: 'The monthly variability is calculated by': No, the variability is something that is already defined. I would write 'An index describing monthly variability.. Response: Yes, the sentence has been changed into "An index describing monthly variability is calculated by". Page 17504, lines 22-24: Where is the verb in that sentence? Page 17508, lines 16-18: Sentence unclear. Table 2, column 1: textbfAfrica -> Africa Response: Here "yielded" is a verb. The sentence of "Due to the high Ca/Al ratio (4.0-10.0) in a range of desert soils in some regions including South Africa, yielded Ca/Al ratios in dust emissions of 1.0, being much larger than those from North Africa." has been changed in "The high Ca/Al ratio (4.0-10.0) in a range of desert soils in some regions including South Africa, yields a Ca/Al ratio in dust emissions of 1.0, being much larger than those from North Africa".

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Page 17508, lines 16-18 has been changed into "The Greenland ice sheet accounted for the dominant part of receiving elements deposition to ice sheets regions, which is equal to the total amount of elements deposited in the whole of the South Atlantic Ocean."

Table 3: Capitalization of words in column 1 needs to be checked Table 4: caption: iffereent -> different footnote b: tunning -> tuning Response: We have tried our best to fix all the errors listed here and other errors in the updated manuscript.

Table 5: I don't understand the footnote! Also, the table is much too small to be read Figure 4: Why do the right and left panels have different sizes? Also the colourmap in d) is different from the others. Figure 5: All colorscales are identical! This is probably wrong. Figure 13: The text in the caption is almost un-understandable Response: For the footnote of Table 5, it means the modeled element deposition has been tuned to adapt the model results to the observed element. It is changed into "Here the soluble element deposition using Sol-1 has been tuned by timing tuned ratios (Table 3); Sol-1 refer to mineral method after tuning, Sol-2 refer to Sillanpää method described in the methods section (2)" to make it more clear. For Figure 5, the colorbar scale is identical due to the value means the ratio of the elemental fraction in atmospheric dust and dust deposition. It is the same order for all the elements. For Figure 13, the caption has been changed into "The percentages of elements in dust deposition (%) after tuning. It is tuned based on original percentages of elements in dust deposition in Fig. S1 by timing Obs./Mod. ratios listed in Table 3. Si did not change because there are not enough observational data available."

References not already present in the manuscript Christian, J.R. (2007). Advection in plankton models with variable elemental ratios. *Ocean Dynamics*, 57(1), 63-71. doi:10.1007/s10236-006-0097-7 Nickovic, S., Vukovic, A., and Vujadinovic, M. (2013). Atmospheric processing of iron carried by mineral dust. *Atmos. Chem. Phys.*, 13, 9169-9181, doi:10.5194/acp-13-9169-2013, 2013. Response: The references above have been cited in the updated manuscript.âĀĀ

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Anonymous Referee #2 The paper presents a method of using soil mineral maps to model the elemental content of atmospheric dust. The paper focuses on eight elements (Mg, P, Ca, Mn, Fe, K, Al, and Si), which are mostly of importance for ocean biogeochemistry. The technique represents an attempt to improve upon models that assume fixed fractions for these elements to simulate ocean deposition. This is a daunting task, since gridded soil maps can not capture all of the regional mineralogical variabilities, the range of elemental concentrations in soils and minerals is quite large, and the concentrations of minerals and elements in soils is different than the concentrations of minerals and elements in the atmosphere. Although the description of the model is quite brief in this paper and I am not a modeler, I suspect that many of the model parameterizations required to simulate elemental concentrations in the atmosphere are rudimentary at the present time. Nonetheless, this work is important for evaluating and improving key linkages between soil and atmospheric aerosol composition, and the effect dust deposition on ocean biogeochemistry. I have only minor comments that should be considered before publication. We thank the reviewer for the very helpful comments, and revise the text to clarify the points addressed by the reviewer. We also agree that this is a first step, and insert a sentence in the conclusions discussing that we think the largest source of uncertainty is in the soil map conversion to elements in the source regions.

There are a few spelling errors here and there. For instance, words like fractionsof, dustis, and observedin appear on line 19 of page 12. This may have occurred in the typesetting process, but a spell checker could easily weed out these problems. Thank you for identifying these typographic and English errors: we have tried to improve the English in the text and correct some errors in the updated manuscript.

There are some grammar issues in a few places: line 28 on page 15, line 28 on page 16. Page 7, line 5 and Table 1b: I find it a little odd that the authors are citing "personal communication" with one of the co-authors. Perhaps "unpublished data" would be more appropriate? Yes, it is already changed into "unpublished data".

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Pages 8 & 13: SD is never defined. I know that it means standard deviation, but it might be a small barrier for some readers. SD is defined in the updated manuscript.

Page 15, line 23: I don't know that I would say that the model and observations are generally consistent in Figure 10, but then again, I am having a really hard time analyzing such small figures. At first glance, I see a lot of red bars that are much higher than the blue bars. Perhaps a scatter plot with a 1:1 line would be more appropriate for such a comparison? You could use different shapes and colors of the points for the various sites. At any rate, figures are important for "hooking" your readers into reading more, and these small panels will hook few people. As you suggested, we have used the scatter plot to replace the bar figure in the updated manuscript. It is clear the values are close to 1:1 line (most in the range of 2:1 and 1:2 line) for most elements at most sites except for Mg, Mn and Si.

Figure 2: Way to many world maps for one figure – break it up! We have split Figure 2 to 2 pages to make the size bigger.

Figure 10: Figure panels are also way too small, and the resulting axes fonts are too small, too. Try to limit yourselves to four panels per figure. Figure 10 has been replotted. Also all the figures in our paper has been reorganized and are much more readable.

Please also note the supplement to this comment:

<http://www.biogeosciences-discuss.net/11/C9415/2015/bgd-11-C9415-2015-supplement.pdf>

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Interactive comment on Biogeosciences Discuss., 11, 17491, 2014.

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