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Interactive Comment

Interactive comment on "TEM analysis of the internal structures and mineralogy of Asian dust particles and the implications for optical modeling" *by* G. Y. Jeong and T. Nousiainen

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Reply to the comments by anonymous referee #2

We appreciate the referee's valuable comments.

Major comments

Comment 1: Throughout the manuscript, the descriptions of particle mineralogy are detailed but, I believe, most readers in the atmospheric field have little knowledge about mineralogy. The mineralogical descriptions should be written in general terms so that readers in atmospheric science can understand. For example, I suggest writing general chemical formula of each mineral so that readers can have idea about their chemistry.

Reply 1: In the final version after the end of discussion forum, we will add a short table of the general chemical formulas which are based on the literature and our EDXS analytical data. We will inspect the manuscript again and revise the mineralogical terms for readers in atmospheric sciences.

Comment 2: This manuscript shows examples of selected Asian dust particles, most of which are larger than 10 micrometer. The discussion in this manuscript mainly based on such large dust particles, although they are small number fraction within all dust particles (Jeong et al., 2014; ACP). (1) I question the significance of these large particles having relatively small number concentrations to the atmospheric aerosol optical properties. In general, atmospheric number concentrations are important as well as volume concentrations. Also, dust particles with several micrometer or smaller were more abundant in their samples (Jeong et al., 2014) and (2) may not neither have pore nor polycrystal structure that are proposed in the current paper. I believe further discussion regarding particle sizes including smaller but more abundant dust particles will be needed to clarify the implication of this study to ambient optical modeling.

Reply 2: (1) Although the diameter of the particle shown in Fig. 5a is ca. 5 μm, the internal structure shows quartz core coated with thin clay layers similar to the structures observed in larger particle (Fig. 6). We have prepared FIB thin slices from two dust samples collected in 2009 and 2012. The sizes of the dust particles collected in 2012 (Jeong et al., 2014) were larger those in 2009. More images were selected from the image data of 2012 dust particles, because they were up-to-date data prepared on the basis of further FIB experiences and improved skill of our FIB team. Another reason is that the larger particles are technically more suitable to FIB handling than small particles. The larger particles are rather stably attached to adhesive substrate, while smaller particles are often not stable because large portion of their rough bases with high curvature was often floated on substrate. Nevertheless, we have observed internal microstructures of some micron-size particles. Here we show two examples (Suppl.

Figs. 1 and 2). (2) We have to pay much more attention to the dust particles of representative sizes in future research by overcoming technical difficulty. However, we think that particles of several micrometers also have structural features similar to larger particles. In the surfaces of desert or arid soils, fine particles are formed by the repeated jump, impact, and fragmentation of particles during the wind erosion. As shown in Suppl. Figs. 3–5, coarser particles can be divided into finer particles of diverse internal structure types (Fig. 14–16 in text). We have not annotated scale bars on Figs. 14–16, because the models for internal structures could be generally applied to common dust particles. Of course, further attention should be given to the finer particles in next steps of research to refine the internal structure models. The models of internal structures can be certainly improved by the continued investigations for dusts from other sources and diverse size fractions in the next stage of study. We will revise the final version considering above discussion.

Comment 3: I am unsure if small pores (< 1 micrometer) within such large particles (> several tens micrometer) indeed have an effect to their optical properties. When dust particles are large enough (tens micrometer), the solar radiation may not be able to penetrate inside the particles. A simple core-shell calculation suggests that small core (pore) within large particles have negligible effects on the optical properties of the entire particles, depending on the choice of refractive index. Note that discrete dipole approximation (DDA) method (Page 6639 line 22) will not be available for large particles such as those used in this study (Draine and Flatau, 1994). As author said further investigations do need to confirm the effects, I would like to see more quantitative discussion.

Reply 3: We agree that one submicron pore inside a particle tens of microns across would most likely not have a substantial impact on the optical properties. However, some of the particle cross sections show tens of percent of the area are occupied by pores. This would certainly have a considerable impact. As to the penetration of the solar radiation inside the particle, the question is unfortunately complicated by the fact

ACPD
14, C1192–C1203, 2014

that dust particles tend not to be homogeneous but are mixtures of different minerals. Some species, such as calcite or quartz, have so low imaginary parts of the refractive index that even crystals tens of centimeters across are transparent. Whether radiation can penetrate a few tens of micrometers depends obviously on the composition, but most mineral species typically encountered in dust are relatively weakly absorbing. Mixed with them are some highly absorbing species, such as iron oxides, but they are seldom evenly distributed. If we, for example, take a quartz particle that has isolated grains of hematite with the total vol% of a few percent, the particle interior will definitely be exposed to radiation. If we take the same amount of quartz and hematite and mix them evenly, the amount of radiation penetrating the particle will be much less. The impact of pores on radiation will therefore depend not only on the size of the pore and the size of the particle, but also on the composition of the particle. The Referee is also correct that the DDA method will not be applicable to dust particles few tens of micrometers across at solar wavelengths. The practical upper limit for computing orientation-averaged optical properties will be somewhere in the size parameter range of 20–30, which corresponds to a particle diameter of 4-6 micrometers at 628 nm wavelength. For considerably larger size parameters, we cannot think of any method that would allow taking into account the irregular particle shape and the inhomogeneity. The coated sphere model can be applied, but mineral dust particles are not spheres, nor are the pores arranged in a concentric ways. Understanding obtained using coated-sphere model would therefore be quite limited. For particle with few micrometers across, the DDA could be used, and indeed we plan to carry out such investigations. A proper description of such investigations would however double the amount of pages of the manuscript, and will take months to carry out, and are thus left for the future. Needless to say, it would also be desirable to obtain data from micrometer-scale particles to guide the model particle generation.

Minor comments

Comment 1: P6620, L6-8: This sentence contradicts to that in P6622 (L1-6). For

ACPD
14, C1192–C1203, 2014

example, Jeong et al (2014) reported mineralogical composition of individual particles. In abstract, it says there have been many reports on the microphysical characterization, whereas in Introduction, it says the microphysical properties of individual particles have not been fully resolved.

Reply 1: In revised version, we will delete “or mineral composition” in Line 7.

Comment 2: P6620 L 25-28: “likely have a great impact”: This statement is qualitative, and no evidence is shown if they have a great impact. Please see Major comment 3.

Reply 2: I think the simplest way to address this is to change the sentence into something like “have to potential to greatly impact…”. The sensitivity studies in the literature show that, so this claim is not wholly qualitative. Something similar is discussed in response to Ref #1, too.

Comment 3: P6623 L18-19: I think there is at least one report by Jeong et al. (2014), who reports internal structures of dust particles.

Reply 3: Jeong et al. (2014) is one of a series of papers organized by the first author (GYJ). The internal structures provided in Jeong et al. (2014) are only small portion of large set of TEM image data which were added to help the readers to better understand particle mineralogy. This paper is in fact the first report dedicated to the systematic investigation of the internal structures of dust particles.

Comment 4: P6627 L25-26: Why they are unlikely to have formed?

Reply 4: Grain arrangement in Fig. 2c is random in overall, but subparallel locally. Long thin lenticular pores in Fig. 2c may have been formed by the dehydration and contraction of subparallel agglomerates. However, the circular pore (arrow in Fig. 2c) cannot be formed by this mechanism, but may have been formed by soil process, particularly repeated wetting-drying and freezing-sawing cycles in the dry and cool sources of Asian dust. We will add this explanation to the revised version.

Comment 5: P6632 L19-21: I guess Iberulite, which is formed from mineral aggregates within rain droplet in atmosphere, may be one of potential formation process of the clay-rich particles.

Reply 5: Cloud processing deserves to be considered as a possible formation mechanism of clay-rich particles. However, the internal structures of iberulite are quite different from dust particles considered in this study. The morphology and structures of iberulite formed from dust-bearing rain droplet are characterized by high sphericity, vortex depression, outward-fining grains sizes, and porous internal structures with some biological fragments (Suppl. Fig. 6). We did not find microscopic features typical of iberulite in the cross-sectional slices of Asian dust particles. However, since there are few available data and inevitably some uncertainty on this subject (cloud processing and any possibility of clay coating formation), we will delete ", and do not form in the atmosphere through, for example, cloud processing."

With the closing of discussion forum, final version will be prepared considering comment and reply above.

Sincerely

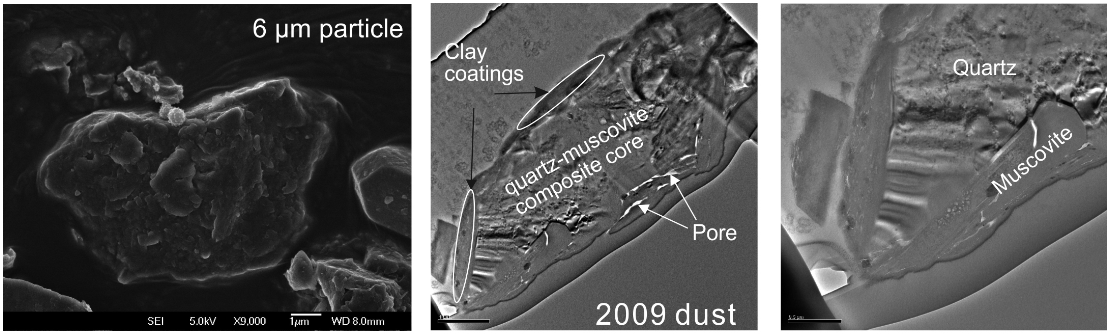
On behalf of co-authors

Gi Young Jeong Corresponding Author

Interactive comment on Atmos. Chem. Phys. Discuss., 14, 6619, 2014.

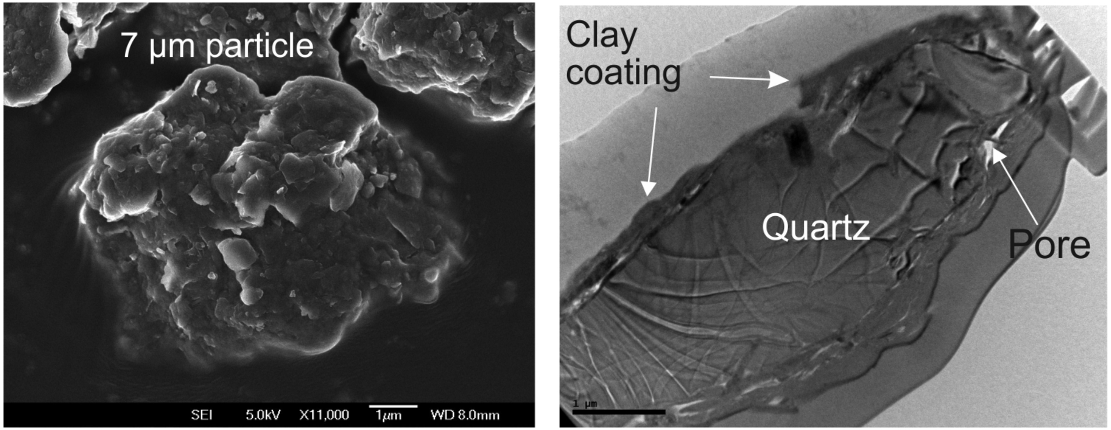
ACPD
14, C1192–C1203, 2014

Interactive Comment



Suppl. Fig. 1. Internal structures of ca. 6 µm dust particle showing quartz-muscovite polycrystal with clay coatings.

Fig. 1. Suppl. Fig. 1

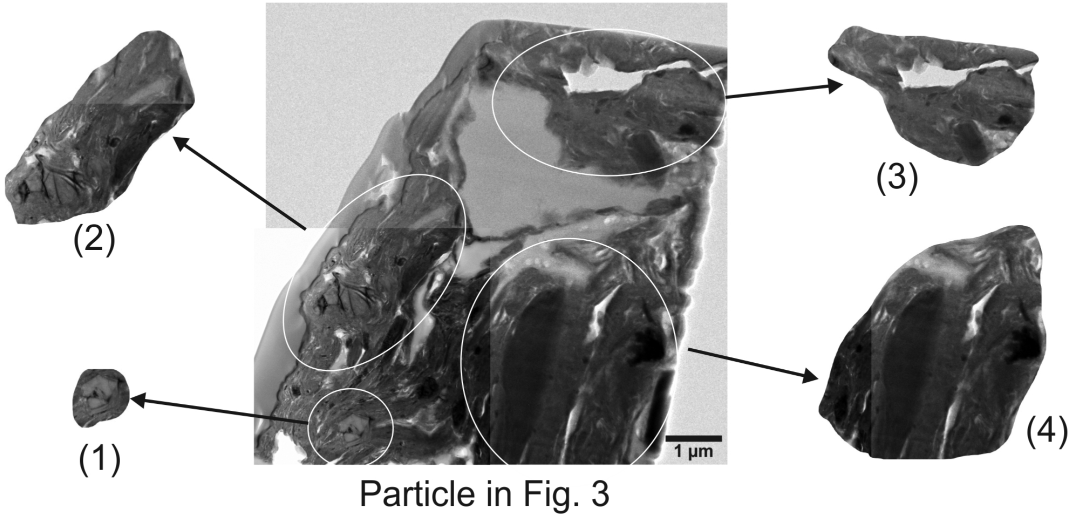


Suppl. Fig. 2. Internal structures of ca. 7 µm dust particle showing quartz core and clay coatings.

Fig. 2. Suppl. Fig. 2

ACPD
14, C1192–C1203, 2014

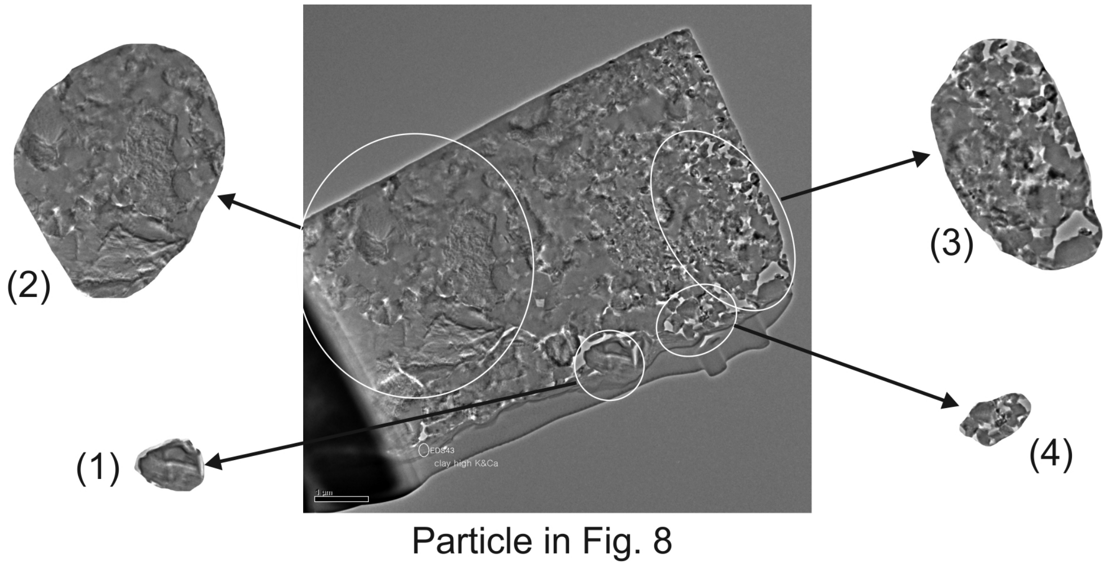
Interactive Comment



Suppl. Fig. 3. Fragmentation of large particle in Fig. 3 may produce small particles such as (1) quartz core coated with clays, (2) preferentially oriented clay agglomerate with quartz inclusion, (3) randomly-oriented porous clay agglomerate, and (4) preferentially oriented clay agglomerate with chlorite inclusion.

Fig. 3. Suppl. Fig. 3

ACPD
14, C1192–C1203, 2014



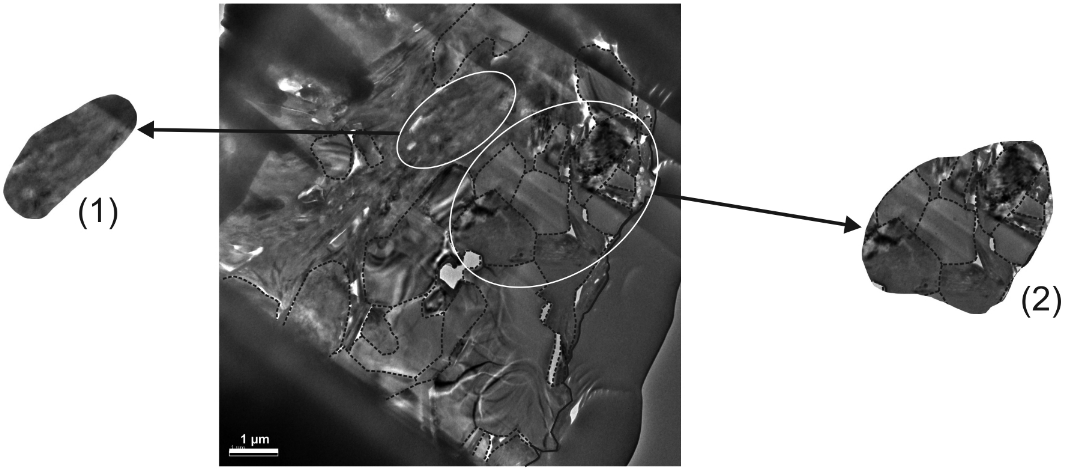
Suppl. Fig. 4. Fragmentation of large calcite-rich particle in Fig. 8 may produce small particles including (1) calcite single crystal, (2) compact calcite polycrystal, and (3, 4) porous calcite polycrystals.

Fig. 4. Suppl. Fig. 4

ACPD
14, C1192–C1203, 2014

ACPD
14, C1192–C1203, 2014

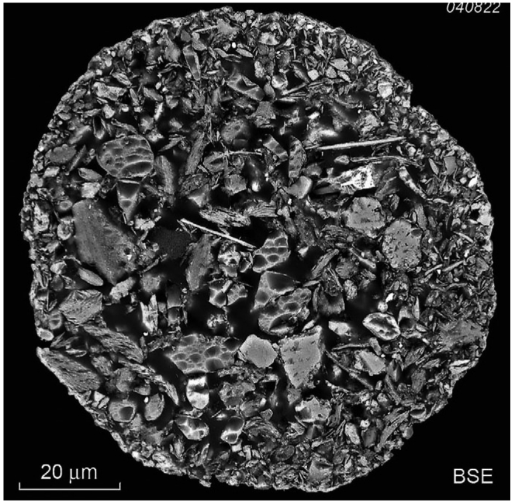
Interactive Comment

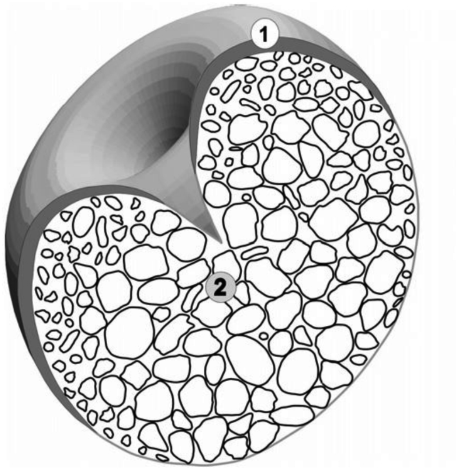


Particle in Fig. 12

Suppl. Fig. 5. Fragmentation of large particle in Fig. 12 may produce small particles including (1) biotite plate and (2) polymineral polycrystal with pores.

Fig. 5. Suppl. Fig. 5





Suppl. Fig. 6. Structures of iberulite from Díaz-Hernández and Párraga (2008, Geoochimica et Cosmochimica Acta ,72, 3883–3906).

Fig. 6. Suppl. Fig. 6