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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Observation of chemical modification of Asian Dust particles during long-range transport by the combined use of quantitative ED-EPMA and ATR-FT-IR imaging" *by* Young-Chul Song et al.

Anonymous Referee #2

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Song et al. present the single-particle analysis of just 109 individual particles collected in Korea during one day of an Asian dust storm event. The particles were analyzed using two complimentary methods: quantitative energy-dispersive electron probe X-ray microanalysis, and attenuated total reflectance FT-IR. The combined analysis by these two complementary methods appears to be the only novel aspect of this work. The single-particle analysis is presented largely as a few case studies in with the chemical composition derived by the two techniques is described in great detail. Little effort is made to really summarize the measurements from the rather small number of particles

analyzed. The fact that so few particles were analyzed from a sample collected on just one day makes it difficult to draw conclusions about the typical composition and behavior of atmospheric dust particles. The analysis of this same one-day sample has in fact already been presented by these authors in three other publications. Overall, I am not sure what has been learned from this analysis. The combined methods certainly provide unique and valuable information, but I do not feel that any new significant findings have been reported here. The atmospheric aging of Asian dust particles has been extensively reported on in numerous reports. While these new single-particle methods could certainly provide valuable new information regarding the composition and aging mechanisms of mineral dust, no such new knowledge appears to be presented in the manuscript in its current form. Therefore, I must conclude that this paper does not satisfy the requirements for ACP, and recommend that it be rejected with the possibility for re-review after extensive revisions and expansion.

My main criticism is that it is not clear that questions or hypothesis are being probed by this study. The atmospheric aging of dust particles is too broad a topic to properly define these questions, and has already been explored by a very large number of publications – including many by these authors. What specific questions or processes are being explored here?

The unique chemical information the authors are able to derive using their combined methods can provide valuable new insights into the aging of dust particles, but I did not think that this was explored very deeply in this paper. Several findings that have been reported in other recent dust aging papers could be explored using these methods. Some questions to explore include:

Is sulfuric acid present with nitric or hydrochloric acids in the same dust particle, or is sulfuric acid not found mixed in high concentrations with other acids as has been reported by Sullivan et al. (2007a)? When is ammonium found in dust particles, if at all? What other secondary compounds are present with ammonium? What other compounds are found when organic compounds are found in dust? Is there any evi-

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dence for photochemical versus cloud processing as a formation pathway for carboxylic acids and other secondary organic compounds in dust? Refer to [Mochida et al., 2003; Sorooshian et al., 2007; Sullivan and Prather, 2007]. How does the dust mineralogy influence the aging of the dust particles? Some insight into this is briefly given for silicate-containing particles that "did not experience chemical modification". This is an important question and should be explored more deeply. An important issue is that the small number of particles examined here, all collected from the same sample, makes it difficult to reliably draw conclusions from this limited analysis to the real atmosphere. It can't be determined if the results obtained from this one sample represent typical atmospheric dust particles, or were unique.

No discussion of the results is presented, just the results, and the conclusions are rather uninspired.

The number of self-citations is excessive here. I counted 18 different papers by Ro, C.-U. cited in this not very long manuscript. Most of these papers appear to describe highly similar topics. The number of self-citations should be reduced to a more appropriate level, while the related work from other researchers needs to be more properly and thoroughly cited here.

[Jeong and Chun, 2006; Jeong, 2008] have presented some interesting single-particle analysis of Asian dust mineralogy that is closely related to this work and should be cited here. Do you find any evidence for or against his report of "microfibers" of calcite in Asian dust particles?

The presence of amorphous calcite was implied by [Sullivan et al., 2010] and the authors might find that report, and the references within, useful.

Analyzing different collection stage sizes would have produced more meaningful results. Apart from the particle size limitation of the ATR analysis, why was only one sample from one stage examined?

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The ability to determine the specific minerals present in dust particles is a big advantage and should be exploited more. For example, on page 27316 the authors state: "On the other hand, in this Asian Dust sample, the CaCO3 moiety originally present in silicate mineral particles appears to be more important for their aging than their silicate mineral type." This is the type of interesting finding that should be explored in much more detail.

The individual particle mixing state naming scheme used here is clear and efficient.

Tables are badly needed to properly summarize the findings. The single-particle analysis must be summarized and digested much more thoroughly.

There are other reports of HCl reacting with dust particles [Ooki and Uematsu, 2005].

Page 27310: "This is the first report of the field observation of CaCl2 particles converted from CaCO3 in a sample collected in the planetary boundary layer." I don't think this is accurate. See [Sullivan et al., 2007; Tobo et al., 2009] for example.

Should also reference the closely related work of [Shi et al., 2008].

Fig. 1C: Does the different color of the particles correspond to anything?

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