

Summary:

This article “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 Song et al. investigates the chemical composition of particles collected in springtime dust storms. The group has previously reported on an overview of samples collected from 2000-2006. One of these samples (from one day in Nov 2002) was unique in that calcium-containing particles were the most abundant particle type. This article investigates the single particle composition and mineralogy for 109 particles. Their combination of EPMA and ATR-FTIR spectroscopy was sufficient to extract mineralogy of the samples. In addition to their extraction of mineralogy information, the main results of this study are that (1) amorphous calcium carbonate was found in four particles and that (2) the paper states that this is the first field observation of CaCl_2 particles converted from CaCO_3 in a sample collected in the boundary layer.

I think this is a very interesting study, as single particle analyses are needed to understand more details of the aging process than can be obtained through bulk studies. I have a number of questions that are listed below, but I feel that with minor corrections, this article would be a good contribution to *Atmospheric Chemistry and Physics*.

Before my questions, however, I want to indicate a few rare, but very valuable pieces of information that the authors have included in their manuscript and supporting information. Specifically, the authors have included a table that includes all particles and their compositions as determined individually from the two techniques. This is a wonderful resource for laboratory researchers. Within the text itself, the figures of FTIR and EPMA data for “representative” particles are clear and well laid out. I want to commend the authors for including this level of detail in the figure set and supporting information.

Questions:

Much of the discussion of mineralogy depends on the observed FTIR transitions. The text would be much clearer if a table of wavenumber regions for different functional groups in FTIR was listed, perhaps by mineralogy. If hydrocarbons were the only type of compound discussed, I wouldn't find this necessary, but it would clarify the results for me for these mineral compounds.

I know that it is common to analyze particles collected on filters during field studies. From using impactors, however, I also know that collection efficiencies can be poor and can depend on the composition of the particles. I'd like to know how representative their distribution of particle compositions is compared to the particles that were in the dust storm. Also, with SEM, the authors are limited to investigating super-micron particles. Could they give a sense of whether the sub-micron particles have a similar distribution of compositions? Do the particles in the other 6 impactor stages have similar composition?

How do the authors know that no modification of the particle composition occurred between 2002 when the particles were collected and 2012?

The authors mention that amorphous CaCO_3 could originate from biogenic or soil origin, but the emphasis in the paper is on the fact that it could be biogenic. There are several sentences about biogenic amorphous calcium carbonate before the authors seem to backtrack and say that it is more likely that the amorphous compound came from soil. I think the authors should state in the

first sentences about amorphous CaCO_3 that it could have a biogenic or soil origin, but that it more likely comes from soil.

The paper states that this is the first field observation of CaCl_2 particles converted from CaCO_3 in a sample collected in the planetary boundary layer. I don't understand the claim because the Sullivan et al. 2007b measurements were ship-based and as a result, they were also collected in the lower troposphere.

It's unclear to me why the authors characterize COO^- as being indicative of humic-like organic compounds. On pg 27309, the authors make the qualification that these spectral signatures could also be from oxygenated carboxylic carbonyl compounds. In other places, the authors just refer to humic acid. Substances like oxalic acid (a carboxylic acid) have been found to be internally mixed with mineral dust aerosol (Sullivan and Prather, ES&T, 2007). Should all indications of humic be qualified as humic or oxidized organics? Also, in the table in the supporting information, sometimes the authors write "organic", "organic (CH, humic)", "organic (CH, COO^-)", "organic (humic)", etc. The differences between these should be explained.

In the silicate particles, I am surprised the authors don't see aluminosilicate clay minerals like illite and kaolinite, especially since illite is mentioned in the article as being found in Chinese loess. Are they able to differentiate, for example, between kaolinite and muscovite? If so, how?

Minor Corrections:

FT-IR is not a typical abbreviation of FTIR. The text should be changed to ATR-FTIR.

In the supporting information, could the major and minor FTIR peaks be indicated in the table of particle composition?

In the figures in the manuscript, I think that once the figures are reduced in size for the published manuscript, some of the fonts will be too small. The elemental composition, legends, and axes, for example, are likely to not be visible. The axes of the FTIR data should be labeled with words and units, not just units.

Pg 27303 line 8 "on the other hand" should be changed to "in addition".

Figure 1 needs a scale bar.

Did the authors study 178 particles (pg 27300) or 109 particles (everywhere else in the manuscript)?

Pg 27314 line 11 "without containing" should be changed to "that did not contain"

Pg 27316 line 24 I think the authors mean that the carbonate minerals are a minor mineral component, in which case "their minor mineral" should be changed to "a minor mineral"

In the table in the supporting information " MO_x " should be " MO_x " where M = metal (Al, Fe, etc.)