

Review of manuscript MS n° acp-2012-732

Title: 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

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General comments:

This article is focused on the chemical modification of Asian dust particles which could occur during long range transport by using combined SEM-EDX and ATR-FTIR imaging.

Authors have previously collected and characterized particles during Asian dust storm events from 2000 to 2006; the results are reported in previous publications. One of the samples (from one day in 2002) was unique because it mainly contained calcium rich particles. This work focuses on the elemental and molecular characterisation of these calcium containing particles. The main results of this work concerns the detailed mineralogical characterization of 109 particles by complementary used of single particle analysis techniques. This original methodology was successfully applied by authors in previous papers published by the group. In addition to the detailed characterization of particles, authors highlighted in this manuscript, amorphous calcium carbonate and CaCl_2 particles in the unique sample. Undoubtedly, this work well demonstrates that using complementary single particle analyses are valuable to properly describe the chemical transformation of particles occurring in the atmosphere.

However, the main drawback of the paper concerns the number of particles which were analysed. In my opinion, results and discussion are based on a too restricted number of particles. For example the number of amorphous carbonate was found for 4 particles and CaCl_2 for 3 particles and described as “a significant portion of Ca-containing particles”.

The “chemical modification of Asian Dust Particles during long range transport” (as announced in the paper’s title) cannot be described with solely 109 particles collected during one day and for the only one size fraction of particles ($> 4 \mu\text{m}$). Authors should strongly demonstrate that such little number of particles is adequate to draw a general conclusion about chemical modification of Asian Dust particles. Finally, there are several over interpretations of the results (see specific comments) that should be revised.

The manuscript should be reconsidered before publication in ACP Journal.

Specific comments:

P 27299 line 21-22: some more recent references should be cited

P27300 line 16: 178 particles or 109 were analyzed?

P27302 lines 2-3: why only particles from the stage 3 were characterized?

P27302 line 10: High vacuum measurements should be specified.

P27304 lines 10-12: PCA analysis was performed on ATR-FTIR data. Is there any correlation between the different PC? That should be indicative for determining the mixing state within particles isn’t it?

P27304 line 15: by “equivalent diameter” Authors mean “geometric diameters”?

P27306 lines 27-28: cristobalite should be replaced by SiO_2 . Note that cristobalite is the high temperature crystallization form of silice. If cristobalite identification is confirmed (quartz is expected), what is the source?

P27309 line 2: I am not convinced that 4 particles can be considered as “significant fraction”

P27309 and 27310 lines 27-28 and 1-2: ACC can be collected within a water droplet without to be hygroscopic. In addition, the spectrum from the particle # 62 (Figure 3) does not clearly show an IR bands from H₂O. The broad band in this spectrum region can also be obtained from a false background correction.

P27310 lines 3-5: 3 particles of CaCl₂ is not significant and cannot be considered as “a significant portion”

P27310 line 4: CaCl₂-containing instead of “CaCl₂-conatining”.

P27310 lines 12-14: this is an over interpretation of the results. There is no proof for an atmospheric CaCO₃ to CaCl₂ conversion. Only 3 particles over 109 were found as CaCl₂.

P27311 lines 7-10. Results undoubtedly shown chemical modification but stating about an “... extensive heterogeneous reactions during long-range transport” is again an over interpretation. This should be true if the results were supported by related backward trajectories.

P27312 line 10: “...are real or due to...” should be replaced by “are of atmospheric origin or due to ...”

P27313 lines 11-12: “humic substances” should be replaced by “organic substances. FTIR spectrum does not provide any evidence of humic substances.

P27313 lines 17 and 24: There are some differences between elemental and molecular composition of particles. Sulphate and silicate (montmorillonite) were identified without detection of sulphur, Al and Si. Is there an explanation?

P27314 lines 8-9: Calcite is not a silicate.

P27315 line 1-2: The sentence “The non-swelling silicate particles also contain water as well as nitrate and organic” should be removed. This is probably a mixing of compounds, isn’t it?

P27315 line 27: (see also figure 5 – particle # 45) Peak attribution at 1623 cm⁻¹ is doubtful, I can only see a broad peak centred around 1635 cm⁻¹ on the figure.

P27316 line 2: “Of these, the major focus has been **on** the ability of...” (**on** is missing)

P27316 line 4: “..to enhance **their** reactivity”

P27318 lines 18-19: The sentence “In addition, a significant number of ACC particles were encountered in the samples” is an over statement. Only 4 ACC particles were encountered in the unique sample.

P27319 lines 1-4: This study provided detailed information on the physicochemical characteristics of 109 particles collected during a specific dust event containing a huge amount of Ca-rich particles. But there is no proof from the presented results of an extensive chemical modification of Asian dust particles during long range transport in general. The conclusion should be modified as regard to the few number of particles analysed. Finally, it should be valuable for the discussion to add backward trajectories in this paper.

A table containing FTIR bands with respective assignments should be added (in the manuscript or in SI) for a better reading of the paper and figures.