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Interactive comment on “A comprehensive characterisation of Asian dust storm particles: chemical composition, reactivity to SO₂, and hygroscopic property” by Q. Ma et al.

Anonymous Referee #1

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This paper describes the multi-analysis of ambient particles collected during a dust storm in Beijing. Asian dust storms are widely known to have large effects on the region's atmospheric chemistry and radiation budget, with resulting climate impacts. This topic has been extensively reported on and is of interest to the Atmospheric Chemistry & Physics readership. As such a vast number of papers characterizing Asian dust storm particles have been published over the last three decades, I feel strongly that successive papers on this topic must present significant new findings and understanding to warrant publication in leading journals such as ACP. Unfortunately this manuscript is severely hampered by inappropriate sample collection and handling methods. This creates great difficulties in accurately and reliably interpreting the re-

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sults from the subsequent particle analysis. The discussion and interpretation of the analysis is incomplete. Furthermore no new significant reliable findings are reported here. Therefore I cannot recommend this manuscript for publication in ACP.

A major concern results from the sample collection methods. Particles were collected “in a clean jar on the roof of a building”. This is not an appropriate way to collect aerosol samples for analysis. Even for TSP measurements particles should be collected on a filter or other substrate. The jar impedes any sort of particle size cut and likely favors the collection of larger particles. There will also be substantial mixing of the particles together, and continued chemical processing as the particles remain exposed to the atmosphere. Furthermore, no details as to how long the sample was collected, the meteorological conditions that prevailed, or the extent of the dust storm (e.g. particle mass loadings) were provided.

As there was no size or other particle segregation during collection it cannot be assumed that only dust particles were collected and analyzed here. Even though collection occurred during a dust storm mixing with other particles and gases from the regions high urban pollution levels will have occurred. Therefore the observations of various components mixed with the dust particles can't simply be assumed to have occurred through reactive uptake or condensation on the particles, as the authors appear to assume here.

There is no description of how the samples were stored or handled before analysis. Serious sampling and storage artifacts can occur if precautions are not taken to prevent the uptake of gaseous components, or evaporation from the sample, for example.

The particles were then diluted in water with ultrasonic dispersion before SEM/EDX analysis. This will alter the chemistry of the particles, removing and redistributing any material that dissolves, and potentially altering properties such as hygroscopicity (Herich et al., 2009; Koehler et al., 2009; Sullivan et al., 2009). Grinding the particles before DRIFTS analysis will also distort the original particle physicochemical proper-

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ties. Alcohol was also used to disperse particles for the Knudsen cell experiments; this can also change particle composition and related properties.

The SEM, FT-IR, and other analyses were all performed on bulk particle samples. Much more valuable information can be derived from individual particle analysis (Falkovich et al., 2001; Jeong, 2008; Laskin et al., 2006; Ro et al., 2005; Russell et al., 2002; Shi et al., 2008; Sullivan et al., 2007a). Why was this not performed?

Since bulk samples with no size segregation were analyzed, these will be strongly biased by the larger dust particles present due to their much larger mass. However large dust particles experience different and often relatively less atmospheric processing than the more abundant (by number) smaller dust particles (Ro et al., 2005; Song and Carmichael, 1999; Sullivan et al., 2007a). This is an important factor to consider but is never mentioned here. I note the large 10 micron scale of the congested SEM image in Fig. 1.

How were the particles treated, if at all, before the TPD and FTIR experiments? Can you be certain that the sample did not absorb water and other gases after collection but before analysis?

Similarly, how were the particles treated before water adsorption isotherms were determined? Often samples are heated to remove pre-existing water to compare with unheated samples.

P. 8906, line 22: Why do you assume that carbonaceous components only coated the particles? If the particles are being heated until they char, these compounds could also be released from the particle core.

P. 8907, line 2: No evidence is presented to establish the process by which nitrate became mixed with the particles. Condensation of HNO_3 and reaction with alkaline components are just two possibilities. Coagulation with nitrate-containing particles and condensation of ammonium nitrate are others. Individual particle size and composi-

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tional analysis could help resolve this.

Little significance can be taken away from the small uptake value for SO₂ on the ambient aerosol sample as this was conducted dry and without oxidant. As the authors point out, adsorbed water and an oxidant are required to convert S(IV) to S(VI) via heterogeneous processes. The ambient particles may have contained secondary material deposited through atmospheric processing which could have impeded the uptake of SO₂ in the Knudsen cell or DRIFTS experiments, unfortunately this was not addressed here. The dry requirement in Knudsen cell experiments represents a serious limitation and is an important reason why this technique has seen decreasing application in recent years to study atmospheric heterogeneous reactions.

It is most surprising that sulfate was not detected. As SO₂ emissions are high in China, and several other papers have detected sulfur compounds in Asian dust storm particles (Ro et al., 2005; Shi et al., 2008; Song and Carmichael, 1999; Sullivan et al., 2007a; Zhang and Iwasaka, 1999), this deserves discussion and explanation.

Similarly, since sulfate was not detected in the particles, why did you choose to expose them to only SO₂(g). Why not also expose them to NO_x/NO_y(g), since nitrate was observed in the particles.

You detect chlorine in the dust particles, but this is not discussed. Dust particles can react with HCl(g) to accumulate chloride (Sullivan et al., 2007b; Zhang and Iwasaka, 2001).

P. 8905, 1st Para: The possible uptake of organic vapors by dust (or other) particles seems to be ignored here (Falkovich et al., 2004; Prince et al., 2008; Russell et al., 2002; Sullivan and Prather, 2007). I do not know what to conclude about the origin of various components detected by the SEM/EDX analysis.

The Reference list was notably lacking several important related papers. Some suggestions are listed below. The lack of papers from the comprehensive studies of Asian

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dust storms during the ACE-Asia, TRACE-P, and PACDEX studies was particularly surprising.

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