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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Dust storms come to central and southwestern China, too: implications from a major dust event in Chongqing" *by* Q. Zhao et al.

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We sincerely thank the reviewer for the valuable comments and suggestions to improve the manuscript.

Comments: First, I think it is essential that the authors emphasize that dust is a minor component of thePM2.5 aerosol. Now, it isn't exactly clear, but the fraction of the PM2.5 attributable to dust during the "major ADE" may have been only 15-20%. Overall, the contribution of mineral dust to PM2.5 was 6-8%, with a major fraction of the PM2.5 undetermined (the percentage was not given, but looks like it could approach 50%). While beyond the scope of this paper, I'd think that it would be important to find out

what makes up that large undetermined component of the PM2.5 and that this issue should be included in statements regarding future work. Also, data for either PM10 or TSP also would almost certainly provide additional insights into ADEs.

Response: This comment contains two threads: that mineral dust didn't really contribute much to the PM2.5, and that much of the PM2.5 was unaccounted for by our measurements. We agree with the sense of the first thread, but with the major caveat that the fractional contribution of dust during a dust event will rise markedly during the event. We couldn't capture this strong increase because we were using weekly samples, and dust storms typically last only for hours. To supplement our measurements, we showed hourly PM10 at site JB in Figure 5. During the event, the PM10 rose by an order of magnitude, as it often does (Dillner et al., 2006; Han et al., 2007). Simultaneously, the pollution part of the aerosol usually remains stable or decreases, depending on the mechanism of transport of the dust (falling or blowing). Thus, the dust probably contributed much more than its weekly 15% to 20% during those few hours. As indicated in the reviewed manuscript (P27035 L10), our observation was not able to show anything about the mixing of mineral dust with pollution aerosol and the interaction with the gaseous precursors during transport, which are the subjects of future studies related to this work.

As for the second thread, it is not correct that up to 50% of the PM2.5 was undetermined. This can easily be seen from data in Table 1 and Figure 11a of the posted paper. The three sites at Chongqing had an average of $\sim$130 μg m-3 of PM2.5. Roughly 10 of that was mineral dust, while roughly 35-40 ($\sim$30%) and 45-50 ($\sim$40%) were secondary inorganics and organic matter (as stated in the text P27040 L2). Only $\sim$20% of the PM2.5 mass was attributed to unknown species probably dominated by water remaining in the weighing condition, due to the large amount of hygroscopic sulfates.

Dillner, A. M. et al, Size-resolved particulate matter composition in Beijing during pollution and dust events, Journal of Geophysical Research, 111, D05203, 2006.

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Comments: Along these lines, the paper's concluding statement is this: "Since dust episodes are natural events, their effects should be carefully considered when developing and promulgating China's National Ambient Air Quality Standard." While I agree with this statement in general, their study actually shows that dust is but a small fraction of PM2.5, and therefore its impact on air quality is arguably much smaller than those of other sources.

Response: (a) See above comment for the strong increase of dust during the hours of an event. (b) Note that the Chinese National Ambient Air Quality Standard is for PM10 rather than PM2.5, which will be more affected by the coarse dust than PM2.5 will be. It is known that coarse dust (PM2.5-10) is transported 1000–2000 km to Japan and Taiwan (Mori et al., 2003; Cheng, 2005) and still dominates the PM10 (e.g., 60%-80% in Taiwan, Cheng et al., 2005). So we can expect something similar for Chongqing. These statements have been added in the revised manuscript (Section 5.1) to make it clearer to the readers.

Mori, I. et al., Change in size distribution and chemical composition of Kosa (Asian dust) aerosol during long-range transport, Atmospheric Environment, 37, 4253–4263, 2003.

Comments: In their conclusions, the authors suggest, "that anthropogenic sources played a minor role during this dust event in Chongqing." This is based on their finding that some pollution-derived substances decreased during the ADE (see comment below), but they have not directly addressed the impacts of anthropogenic sources on dust loads in their study. Could this not be resuspended dust from construction activities or agriculture? They note that an assessment of sources is going to be presented elsewhere, but without seeing that information, the contributions from anthropogenic sources cannot be ascertained.

Response: Evidence against local resuspension of dust was twofold: (1) an abrupt decrease of wind speed just when the PM10 increased, and (2) the appearance of the

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dust peak four hours earlier than the normal midnight peak of pollutants. Both these points were shown in Figure 5 and noted in Section 4.1 (P20731 L25).

Evidence provided against industrial pollution (power plants, etc.) and for transported dust was chemical. (1) Ratios of pollution components (Pb, SO42- and OC) to Al decreased to a minimum during the event (showing the strong contribution of dust to the extra material). (2) The Ca/Al ratio also decreased to a minimum then (showing that the extra material was not influenced by construction in nearby urban areas). (3) The Si/Al ratio decreased to a minimum then (showing that the dust had been transported). These decreases were shown in figures 6 and 7, and discussed in sections 4.2.1 and 4.2.2. Supporting material is given for coal burning signatures in Okuda et al. (2008) and Guo et al. (2008), for biomass burning signatures in Duan et al.(2004), for the Ca/Al ratio in Zhang and Iwasaka (1999) and He et al. (2001), and for depleted Si after transport in Gatz and Prospero (1996).

Overall, the possibility of the major contributions from anthropogenic sources can be ruled out by the direct and indirect lines of evidence shown above. Most of them have been included in the reviewed manuscript, and others have been added in the revised version (see the first paragraph of Section 4.2.1 and 4.2.2).

Okuda, T et al., Trends in hazardous trace metal concentrations in aerosols collected in Beijing, China from 2001 to 2006, Chemosphere, 72, 917-924, 2008.

Comments: The method for estimating PM10 from the API needs to be described in some detail and more important, caveats included to make it clear to readers that the reconstructed PM10 not only includes dust but also other substances. The authors do acknowledge this albeit somewhat obliquely on page 27032 (l 32), but the limitations of the reconstructed PM10 should be made clear early on, preferably in the description of the method. The difference between PM10 and dust weakens the analysis of the chronology of the major ADE considerably, and furthermore, one would expect that much of the PM10 could be from local sources.

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Response: No PM10 was estimated from API here. The hourly PM10 concentration in Fig.5a was directly measured by TEOM (data obtained from local monitoring station of Chongqing EPB, as has been further clarified in the revised manuscript). From the points made in the previous response, we believe that local sources would also be of minor importance in the high dust loading for PM10.

Comments: If there are no useable data for crustal K, Fe, etc., it doesn't make sense to estimate them from Al unless there is a reason to discuss them independently, that is, no new information beyond what is contained in the Al concentrations is being added by this procedure (they are just weighting the Al data more than the other well determined elements). My suggestion would be to estimate the dust concentration based on the data for each of the crustal elements that is well determined and then take an average or weighted average of those estimates.

Response: We see little practical difference between the two approaches. Since crustal ratios to Al are well known and have been used for reconstructive purposes for many years (e.g., Malm et al., 1994; He et al., 2001), we consider our method to be acceptable for the purpose at hand.

Malm W.C. et al., Spatial and seasonal trends in particulate concentration and optical extinction in the United States, Journal of Geochemistry Research, 99: 1347-1370, 1994.

Comments: I also wondered why the authors relied on the Taylor and McLennan reference for crustal material. There must be good data for soils from the source regions available. This would likely not make a big difference, but would be a more valid approach.

Response: We agree that it is better in principle to estimate dust from soils rather than from crustal rock, there have always been practical difficulties associated with using soils. For example, we could not use the comprehensive data of spatial distribution of elements in Chinese soils from the China National Environmental Monitoring Center

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(1994) because they refer to bulk soil. Since the elemental composition of soils and their derived dusts varies strongly with particle size (Schutz and Rahn, 1982), bulk soil could bias the results considerably. So we decided to take the average crustal rock instead (Taylor and McLennan, 1995), which has been found close to Chinese soils (Yuan et al., 2008), as stated in the manuscript (P27028 L20). In the end, all available methods are compromises, and the evidence shows that our approach is as good as any other.

China National Environmental Monitoring Centre, The atlas of soil environmental background value in the People's Republic of China, China Environmental Science Press, Beijing, 1994.

Schutz L. and Rahn K.A., Trace element concentration in erodible soils, Atmos. Environ., 16, 171-176, 1982.

Comments: Further in terms of crustal signatures, I have two questions (1) how are the elemental ratio lines in Fig. 6 determined? And (2) in each of these plots, especially Mg/Al, there a numerous points below the crustal line, what do these points represent? One explanation is that they are the result of material containing Al from non-crustal sources, another is that the elemental ratios vary with dust load, which could be possible if there the mineralogy or particle sizes with dust load. Finally, this could be a consequence of analytical uncertainties, which brings me back to question (1): what are the uncertainties associated with the ratios determined in this way?

Response: (1) The lines in Figure 6 were determined from the 6 ADE samples at the three sites in Chongqing (as marked in black and indicated in P27033 L12), and represent our best estimates of the signatures for the transported dust. They had small scatter (Si/Al: 2.39$\pm$0.11; Ca/Al: 1.05$\pm$0.11; Mg/Al: 0.33$\pm$0.02), especially when compared to the whole sampling period (1.1–4.0, 0.3–2.4 and 0.2–0.6 for Si/Al, Ca/Al and Mg/Al, respectively). The scatter in the 6 ADE samples was at the level of typical analytical uncertainties (5% to 10%), although geographical variations over the source

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deserts may have also contributed. The standard deviations of the calculated ratios have been added to the revised manuscript.

(2) The points below the lines may represent normal variations of composition (pollution and local crustal) at the sites, analytical scatter, and special events such as fireworks during the Chinese Spring Festival in February of 2005 and 2006. The latter is supported by the finding that Al is one of the most enriched elements in fireworks (Wang et al., 2007). We consider such scatter to be completely normal for environmental samples. There is no reason in principle that these three ratios cannot occasionally be lower in polluted areas than in distant deserts.

Wang Y. et al., The air pollution caused by the burning of fireworks during the lantern festival in Beijing, Atmospheric Environment, 41, 417-431, 2007.

Comments: Throughout the paper the authors allude to differences or significant differences between concentrations (or that the concentrations for one site were higher than the other). There is no indication of statistical significance here or whether any tests were done to evaluate the significance of the differences; therefore, the validity of these statements and arguments is questionable, e.g., the difference between 130 and 118 on pages 27026 and 27. Further, there appears to be a mismatch in data between Chongqing and Beijing, and I did not see that the authors adjusted for this in their comparisons. The reason for the data gap needs to be explained and taken into account even if it is only a failure of the sampling gear.

Response: It is very difficult to satisfactorily address this question in the way that the reviewer has in mind. First off, the $\pm$ figures given in Table 1 do not represent true standard deviations, because atmospheric variables are distributed log-normally rather than normally. These sets of data are even more nonideal, because they contain systematic seasonal variations as well as pulsed increases from springtime dust storms. Thus, the samples were not chosen randomly from a well-defined distribution. Setting all that aside for the moment, however, we can get a rough idea of probabilities by

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comparing the two annual means and standard deviations of the mean (standard errors). For 50 samples at each site, the standard deviation of the mean will be 7 times smaller than the (single-sample) standard deviations reported in Table 1. This would give 130±6 μg m-3 for Chongqing and 118±6 μg m-3 for urban Beijing (TH). The two means are thus two standard deviations apart. This effective difference is confirmed by a standard probability test for the two means, which gives only a 14% chance that they are the same. Thus (a) we can't do anything better than taking the measured means at face value, but (b) if we try to check with unjustified statistics, we also get that they are not the same.

The data gaps are mainly due to missing samples. The samples covered 92%, 94%, 83%, 85% and 80% of the 52 weeks from March 2005 to February 2006 at JB, DDK, BB, TH and MY, respectively. Thus, their averages should reasonably represent the concentrations at each site, and so we didn't adjust them.

The plausibility of this approach can be shown by a worst-case example for MY, where 20% of the samples were missing. Even if the average of the 20% unsampled weeks was 50% higher than the sampled weeks (highly unlikely), it would only increase the average by 10%. A more-reasonable difference of 5% would change the average by only 1%. Mismatched datasets are common with long-term multisite regional observations (e.g., Zhang et al., 2003b; Querol et al., 2009). We have noted the possible bias caused by the data gaps (P27027 L14). The above discussion has been simplified and added to the revised manuscript.

Querol, X. et al., Variability in regional background aerosols within the Mediterranean, Atmos. Chem. Phys., 9, 4575-4591, 2009.

Comments: I am having some difficulty reconciling the argument (p27040), "This suggests the Asian desert dusts are influencing air quality over broad regions of China." with another argument (p27041): "Because meteorological stations are normally located at reachable sites such as urban areas or agricultural lands, there can be other

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days when ADEs are present in remote regions but go unreported." That is, the authors might want to modify their conclusions and argue either that the dust storms are of a broad regional scale or they are small enough to affect remote sites but not areas where observations are normally made. (I would argue the former is more often accurate).

Response: These two statements do not have to be "reconciled," because they refer to different topics. The first (from page 41 and 42, not 40) deals with the scale of dust storms, which our data indicate is broader than often understood. The second deals with reporting the eventsâĂŤwe feel that there may not be enough meteorological stations in remote areas to fully demonstrate the presence of dust storms there. In other words, the broad effects that we found might be recognized as being even broader and more frequent if more stations were available to report on dust.

Comments: Indeed, the x-y plot of the dust concentrations at Chongqing vs. Beijing shows that the correlations between the dust concentrations are driven by a small number of points. These presumably represent the large-scale events.

Response: Right. Thanks to the reviewer's earlier suggestion, we have been able to show the large scale covariation between Chongqing and Beijing more clearly with the x-y plot in Fig.3.

Minor points Comments: The African deserts are larger sources of dust than the Asian ones, so the first sentence of the Introduction is not accurate.

Response: The reviewer is correct. We have amended the first sentence to read: "Arid regions of southern Mongolia and northwestern China are among the main contributors to global dust emission . . . ".

Comments: P 27023 (henceforth just P23, L 19) Since alkaline dust helps offset acidic aerosols (awk) L 25 is restrained by climatic (word choice)

Response: We have changed "offset" to "neutralize" in the revised manuscript, but we

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see nothing wrong with “is restrained by . . .”.

Comments: P 24 L12 At the end is discussed the possibility (awk)

Response: We think this is also a proper construction in English. It shifts the longer part of the sentence to the end, where the reader expects to see the detail.

Comments: P 25 L3 The rural site was near the Jinyun L6 Great North China Plain, and <it> has

Response: We see nothing wrong with L3 and L6. In L6, the “, and has . . .” makes a parallel construction with “Beijing is . . .” (Beijing is . . ., and has . . .).

Comments: Sampling and analysis: technically, I think it is more accurate to state that concentrations were determined rather than measured.

Response: We disagree. True, mass rather than concentration was measured, but it is worse to use the general verb “determined”, which has many other meanings.

Comments: P27 Is it possible to compare the TEOM and gravimetric data directly?

Response: Yes. Studies that have compared TEOM data to gravimetric data found that TEOM data tends to be lower by up to ∼30% in Beijing and the United States, mainly due to the loss of water and semivolatile organics and inorganics (Chow J. et al., 2006; Wang et al., 2009). We needed to compare the two in order to show the relevance of this work to the National Ambient Air Quality Standard.

Chow, J. et al., PM2.5 and PM10 Mass Measurements in California’s San Joaquin Valley, Aerosol Sci. Technol, 40, 796-810, 2006.

Wang W. et al., Atmospheric Particulate Matter Pollution during the 2008 Beijing Olympics, Environ. Sci. Technol., 43, 5314-5320, 2009.

Comments: L24 “elsewhere” could mean in another section of this paper, but what is meant is in another paper.

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Response: Right. We have changed “elsewhere” to “in an accompanying paper.”

Comments: P29 L 28 I think of secondary aerosols as forming heterogeneously thus the dust would provide surfaces on which reactions would occur but not lead to secondary particle formation. In fact, reactions on the dust may well remove aerosol precursors from the atmosphere and reduce the number of secondary aerosols formed!

Response: In the first sentence above, the reviewer is thinking of numbers of particles, whereas we dealt with mass throughout our paper. We are both right. His second sentence is also true only for numbers of particles.

Indeed, aerosol precursors may be removed from the atmosphere through the reactions with alkaline mineral particles, but it does not necessarily lead to a net reduction of the secondary aerosols. As observed in many previous studies (Usher et al., 2003; Sullivan et al., 2007), coating of hygroscopic sulfate ((NH4)2SO4, NH4HSO4, or H2SO4) would also be favored during dust event when the surface area of dust can increase by up to an order of magnitude (Dentener et al., 1996). Besides, secondary aerosols are also supposed to be including the sulfate and nitrate in form of CaSO4, Ca(NO3)2, etc., which were found abundantly coating on submicron Asian dust (Li et al., 2009). To clarify this issue, these statements have been added in the revised manuscript after simplification.

Dentener et al., Role of mineral aerosol as a reactive surface in the global troposphere, Journal of Geophysical Research, 101, D17, 22869-22890, 1996.

Li, W. J. and Shao, L. Y., Observation of nitrate coatings on atmospheric mineral dust particles, Atmos. Chem. Phys., 9, 1863-1871, 2009.

Sullivan, R. C. et al., Direct observations of the atmospheric processing of Asian mineral dust, Atmos. Chem. Phys., 7, 1213-1236, 2007.

Comments: P35 L 17 what’s the significance of 3 km? (with reference to above or below it?)

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Response: The transport distance of Asian dust depends on its altitude it can reach (Sun et al., 2001; Tsai et al., 2008). Dust above 3 km tends to get caught up in the prevailing westerlies and travel longer distances, while that below 3 km stays nearer the surface, and is thus deposited nearer the source. We have modified the sentence to " (dust) can then be carried above or below 3 km to downwind regions of different distance".

Comments: L25 During this event: (which event?)

Response: We have modified the text to "the ADE in Chongqing" in the revised manuscript.

Comments: P36 L 5 "were kept" (word choice) L6 "took over" (word choice)

Response: We have changed the phrases to "stayed well below 2 km. . ." and "reached heights above 0.6 km . . . "

Comments: P37 L 2 throughout the event (which event?)

Response: We have modified the text to "the ADE in Chongqing" in the revised manuscript.

Comments: L5 low- and high- pressure systems began to accumulate over these regions simultaneously (not sure this is accurate)

Response: It is accurate. We checked.

Comments: L 19 observed (How?)

Response: By examining the hourly trajectories at various heights in Chongqing during 5-6 May 2005, as partly shown in Fig. 9. This has been added in the revised manuscript.

We found some incorrect figure names on P27037, and have corrected them as follows: L18: Fig.9b to Fig.9a; L13: Fig.9a to Fig.9c; L23: Fig.9c deleted.

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Comments: P38 APIs and transport pathways: even assuming the APIs were a measure of PM (the index also can be affected by trace gases), dust is a small fraction of PM2.5 and an unknown fraction of PM10 or TSP (but probably a minor fraction in most cases) and so the APIs are generally not directly related to dust concentrations.

Response: The APIs in Figure 10a (all representing PM10) were compared against each other only, and were accompanied by the statement that "APIs cannot directly represent the dust during this event."(27039 L1) Concerning the fraction of the dust in PM10, a recent review has found that dust generally accounted for ∼40% of the PM10 mass in central and western China (Fang et al., 2009), and a much more considerable amount of dust would be expected in PM10 during 5-7 May 2005 for the 6 western cities. Their consistency of PM10 evolution with the chemical signatures, meteorological evidences and modeling results does support the transport pathways of the desert dust from the northwest to southwest of China. Statements above have been added to the revised manuscript to clarify this issue.

Comments: L20 it activated a variety of deserts {caused the production of dust (or dust storms) in}

Response: We have changed the phrase to "produced dust or dust storms in . . ."

Comments: P42 L 9 The pollution-derived components, such as Pb, SO4, and OC, decreased significantly as Al concentration increased (In other studies, pollutants have increased during Asian dust storm, why should they decrease as dust loads increase?)

Response: A better statement might be "Ratios to Al of pollution-derived components, such as Pb, SO4, and OC, decreased significantly as Al concentration increased."

It is normal for pollutants to decrease during dust storms, because the dust comes from lightly polluted desert areas. Sometimes the increase of elements like S, which can have strong desert sources in addition to their usual pollution sources, is mistaken for an increase in pollution (e.g., Sun et al, 2005, as indicated by Rahn et al., 2005).

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Increases in Zn can also be misinterpreted in this way. Increases and decreases of pollution-derived components in previous studies have been summarized by Chan and Yao (2008), but must be examined with a critical eye. The changes in composition of aerosol during dust storms may depend on the type of mixing between the dust and the pollution, as proposed by Guo et al. (2004) and Zhang et al. (2005).

Guo et al., A mechanism for the increase of pollution elements in dust storms in Beijing, Atmospheric Environment 38, 855–862, 2004.

Rahn et al., Comment on "Chemical composition of dust storms in Beijing and implications for the mixing of mineral aerosol with pollution aerosol on the pathway," Journal of Geophysical Research, 110, D24209, 2005.

Sun et al., Chemical composition of dust storms in Beijing and implications for the mixing of mineral aerosol with pollution aerosol on the pathway," Journal of Geophysical Research, 110, D24209, 2005.

Zhang et al., Ground observation of a strong dust strom in Beijing in March 2002, Journal of Geophysical Research 110, D18S06, 2005.

Comments: L 15 dusts were (I'd change this to dust was)

Response: We have modified the text according to the reviewer's suggestion.

Comments: L 24 "normal pollution events (strange expression)

Response: We see nothing strange about "normal pollution events."

Tables & Figures Comments: Table 1. Give N, the numbers of samples. What do the +/- denote? There should be a footnote for the abbreviations. Too many significant figures, I'd think.

Response: $\pm$ donates the standard deviation. We have added the number of samples and a footnote for $\pm$ according to the reviewer's suggestion.

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Comments: Fig 1. Spell out names Fig 2. Are the enrichments the same or different among sites?

Response: Names have been spelled out in caption to Figure 1. Figure 2: We show the three Chongqing sites so that readers can see for themselves. Except for K, we regard the enrichments as functionally indistinguishable at the sites.

Comments: Fig 4. I would change the x-scale (maybe just the labels need to be changed) to be time and show the concentrations as horizontal lines, with samples connected by vertical lines.

Response: We are not sure what the reviewer means here. We regard this figure as the clearest way of showing the concentrations and their temporal changes, even though the labels are a bit small.

Comments: Fig. 6. Doesn't need to be in color.

Response: We disagree. The colors make it much easier to distinguish points from the three sites, especially when the dots of ADE samples are marked in black.

Interactive comment on Atmos. Chem. Phys. Discuss., 9, 27021, 2009.

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Interactive Comment