Atmos. Chem. Phys. Discuss., 11, C7742–C7745, 2011
www.atmos-chem-phys-discuss.net/11/C7742/2011/

Atmospheric Chemistry and Physics Discussions

Interactive comment on "Morphology, composition and mixing state of individual carbonaceous aerosol in urban Shanghai" *by* H. Fu et al.

Anonymous Referee #1

Received and published: 15 August 2011

This manuscript presents the results of TEM-EDX analysis of several hundred particles collected in Shanghai during late Fall of 2010. Particles were broadly placed into once of several categories based on the observed morphology, composition, and mixing state. The authors also attempt to explain the sources of these particles by comparing the temporal variability of particle-type relative abundances to supporting particle data and air mass back trajectory analysis. This reviewer is not well qualified to critique the TEM-EDX analysis itself, and so the review will instead focus on the motivations for the study and how the results of the TEM-EDX analysis are interpreted with respect to the overall air quality situation in Shanghai.

The manuscript in its current form has significant flaws. The descriptions and images of the different types of particles found during the study are inherently interesting, but it is difficult to see how these results contribute in any real way to our understanding of the causes or nature of particulate air pollution in Shanghai. This is largely a critique of the technique itself. TEM-EDX analysis as described in the manuscript results in a qualitative description of a very small subset of atmospheric particles- a subset that seems to be heavily biased toward the particles most easily observable by TEM-EDX. To make the point, consider the numbers of particles analyzed in this study. The authors note that their samples were collected at 1 lpm with sampling durations of 30-90s. So a total sampling volume of 1000 cm3 is a reasonable estimate. Using the data in Figure 3, the average number concentration is roughly 8000 per cm3, so the TEM grid was exposed to approximately 8 million particles per sample. From Figure 3, approximately 20% of these, $\sim$1.6 million, would have been larger than 100 nm. Of these 1.6 million particles, less than 300 were analyzed, a rate of $\sim$0.02%!

To build a useful general description of the aerosol population when only sampling 0.02% of the particles larger than 100nm is challenging to say the least. To be successful at all, it seems critical that the sampling biases be eliminated wherever possible, and that they be characterized in detail when they cannot be eliminated. Unfortunately, the authors have not done either of these very well. The authors give a brief but adequate description of the hardware used for the TEM-EDX analysis, and allude to a few major sources of sampling biases (especially the loss of semivolatile particle material). However, there is no discussion at all regarding how individual particles were chosen for analysis and how that selection process impacts the results. Was every visible particle on the TEM grid analyzed? If not, were the selection criteria for analysis formalized, or random, or at the discretion of the user? Clearly, any biases in what particles were available for analysis, or in how specific particles were selected for analysis, could have enormous impacts on the statistical distribution of the particle types reported. Without resolving these questions it is impossible to use the results of the TEM-EDX analysis to quantitatively describe the aerosol population in a general way.

Again, the above critique is not meant to imply that the TEM-EDX analysis has no utility in particle analysis. When used to probe a narrow well-defined set of questions, the technique can be quite powerful. A good example of this is the work of the Buseck group, who use the technique to analyze the optical properties of soot particles and how soot agglomerates respond as they are aged in the atmosphere. It may be possible to use the data set here to address a narrow question in this manner, and the authors are encouraged to pursue this option.

There is also some value in a more explicitly qualitative analysis in support of a more general case study using a larger data set. Indeed, the authors do seem to have the beginnings of a solid analysis along this line. They mention the influence of dust event on November 12, and a period of stronger marine influence the next day. They also suggest periods of heavier pollution during atmospheric stagnation compared with other periods. They appear also to have quite a bit of additional data for describing these case studies. Perhaps another manuscript is in the works, but from what's available here, it seems that using the differences in particles found in the TEM-EDX analysis could be strong supporting evidence for some of these case studies.

Ultimately, despite the apparently high-quality TEM imagery included in this paper and the extensive work that went into it, it is not possible to accept this paper without major revisions. The qualitative images and accompanying descriptions are interesting but by themselves do not contribute significant new knowledge to the field. The authors seem to have been aware of this; they tried to convert the qualitative images to a statistical assessment, but the potential for sampling biases is too great for such an assessment to have validity. They also try to place their results in the context of the larger atmospheric environment at the time of sampling. This approach has merit, and potential, but in current form the arguments are too scattered and incomplete to form a complete paper. In addition to these critiques, it must be noted that while much of the paper reads well, the abstract and introduction have significant writing quality issues. Copy-editing for these sections at least is strongly recommended.

Interactive comment on Atmos. Chem. Phys. Discuss., 11, 20973, 2011.