(1) 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 particles-type relative abundances to supporting particles 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.

In this work, we analyzed 834 individual aerosol particles collected in Shanghai in four different days, with different weather and air quality conditions. On the basis of the morphology and chemical composition gained by TEM-EDX, carbonaceous aerosols were categorized into POC, soot, tar ball, biogenic particle and internally mixed particles. Since carbonaceous aerosols exert great influence on climate and public health, we expected to gain insight into their formation and sources on local, regional, and long−range levels, and to possibly make a contribution to the determination of the impact of these aerosols on regional air pollution in east China. To the best of our knowledge, single carbonaceous aerosols emitted from Shanghai are still not well known. However, just as the anonymous referee said, the manuscript need be further improved, especially about the TEM-EDX analysis.

(2) 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 cm^3 is a reasonable estimate. Using the data in Figure 3, the average number concentration is roughly 8000 per cm^3, so the TEM grid was exposed to approximately 8 million particles per sample. From Figure 3, approximately 20% of these, ~1.6 million particles, less than 300 were analyzed, a rate of ~0.02%!

Indeed, the manuscript in its current form has some flaws, but it supplied some useful information to understand carbonaceous aerosols in the Shanghai atmosphere. For example, carbonaceous aerosols were categorized into five types, including POC, soot, tar ball, char and biogenic particle. Most of particles were coated with secondary organic aerosols. The internally particles of sulphates, organics and soot were encountered frequently in the clean day. Aged particles were associated with days with low wind velocities, showed complex structures, and were bigger in size. Based on the TEM-EDX single particle analysis, coupled with air mass back-trajectories, meteorological conditions and air quality parameters, we discussed and interpreted the sources of the carbonaceous aerosols in Shanghai, and suggested that marine-originated air mass had significant influences on the Shanghai atmosphere.

On the basis of the reviewer's estimation, only a small fraction of particles were collected. We could not analyze all the particles collected on the grid using TEM-EDX due to huge amount of work. The main advantage of using microscope methods is that they can provide simultaneous information on single particle properties. Furthermore, TEM-EDX has the advantages of high spatial specificity for shape, composition, and structure, and can see particle interiors. However, it suffers from manually operated and labor-intensive, resulting in poor statistics. Beyond, it has a limitation concerning to the analysis of particles less than 100 nm in size and semivolatile particle material. In spite of that, the analytical TEM technique is extremely useful for characterizing aerosols, especially for internally mixed particles, which is a unique source of information on particle coatings, agglomeration, and possible atmospheric reactions. By using TEM techniques, Buseck' group supplied visual evidence that the radiative property of soot was determined, to a large extent, by the mixing state of soot within individual particles, suggesting that the external mixing assumption is unrealistic for many atmospheric situations. Li and Shao' work also deepened the understanding on the haze in east China using TEM-EDX methods.

(3) To build a useful general description of the aerosol population when only sampling 0.02% of the particles larger than 100 nm 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 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, 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.

Based on this reasonable suggestion, we reexamined this manuscript. Some important information was lack in the experimental section, which makes readers confused. In the revised version, we almost reorganized the experimental section, and supply the detailed information about individual particles' selection for analysis and how that selection process impacts the results.

The distribution of aerosol particles on the TEM grids was not uniform. Coarser particles were deposited near the center of the grids and finer particles on the periphery. Outward from the grid centers, the particles are more sparsely distributed. Because of the manual, labor-intensive operation of the TEM, only limited numbers of particles could be analyzed. To ensure that the analyzed particles were representative of the entire size range, three to four areas were chosen from the center and periphery of the sampling spot on each grid. Size of each particle was calculated using the best fitting ellipse to a particle outline, and the diameter of each particle was calculated as the arithmetic mean of the short and long axes of the ellipse. **It should be noted that** the upper limits of the resulting size distributions may be affected by the inhomogeneous distribution of particles on the grids, because some large particles pile up in the center, where particle overlap makes size measurements impossible. On the other hand, the lower ends of the size distributions may have been affected by poor collection efficiency of < 100 nm particles on the TEM grids.

Particles examined by TEM were dry at the time of observation, although some may have been droplets in the atmosphere because of the high relative humidity in the environment. Ammonium sulphate particles probably crystallized on the TEM grids. Therefore, the morphologies of some aerosol particles, as observed in the TEM, may differ from their shapes prior to collection. Despite possible changes in morphology, and except for volatile species, the compositions of the particles measured herein should reflect their original compositions before collection.

Decomposition of the particles in the vacuum or under the electron beam further complicates EDX analyses. Changes in the TEM are visible, and a low-intensity electron beam was used to avoid decomposition during spectrum acquisition. Tar balls, soot particles, and biogenic particle did not show any changes in the beam, whereas the organic coatings and the internally mixed particles containing sulphates invariably changed under the electron beam; an analysis of such particles indicates the composition of the residue after the majority of the particle evaporated. In general, even where the EDX results may not accurately reflect original particle compositions, the analyses show characteristic changed between groups of particle types and from specimen to specimen, and thus useful for distinguishing certain groups of particles and for studying the trends of changes.

(4) 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.

Indeed, the TEM-EDX analysis is a powerful tool to analyze the optical properties of soot particles and to study hydration/dehydration cycles of aerosols. By using TEM techniques, Buseck's group supplied visual evidence that the radiative property of soot was determined, to a large extent, by the mixing state of soot within individual particles, suggesting that the external mixing assumption is unrealistic for many atmospheric situations. Therefore, the core-shell model does not properly represent the shapes of ambient soot particles.

(5) 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 Nov 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.

Only one manuscript is in the works.

(6) 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.

We described the TEM analysis in detail in the revision version. In my opinion, this manuscript supplies the information about morphology, composition and mixing state of individual carbonaceous aerosol in urban Shanghai. On the basis of air mass back trajectory analysis and the temporal variability of particles-type relative abundances; we also explained the sources of these particles.

On the basis of the reviewer's suggestion, we improved greatly the quality of MS by major revisions. The major changes in the revised version are as follows:

i) TEM analysis was described in detail. We explained how individual particles were chosen for analysis and how that selection process impacts the results. The experimental section was almost reorganized;

ii) In the POCs section, Si in OC is very common but it is not quartz. Although the ratio of Si/O is nearly 1:2, we only can identify it as "Si-rich particles" (Fig. 5a);

iii) Only two particles in our samples were identified as "Char". In TEM study, the occasion is possibly. Therefore, they have not assigned as one kind of particles. We deleted the paragraph about "Char";

iv) We were lack of the proof that the particles shown in Fig 8d and e are biogenic aerosols. In the revised version, the figures were deleted, and relative abundances on the sample of November 13 were counted again. The corresponding discussions were also deleted or revised;

v) In the revised version, we have corrected a large number of English errors of MS, including incomplete and grammatically incorrect sentences and misspelling.

We acknowledge the comments of the reviewer.