General comments:

This paper presents information about the composition and morphology of carbonaceous particles in Shanghai, China, based on TEM-EDX single particle analysis. Just as the anonymous referee #1, I am not qualified to evaluate the TEM-EDX technique itself. However, I feel comfortable to evaluate the significance of the results to the aerosol scientific community. The radiative properties of aerosols depend on their size, composition, morphology, and mixing state. To my knowledge, there are not online techniques that could provide information about aerosol morphology and mixing state. Individual particle analysis by TEM-EDX provide this kind of information, but it has a limitation concerning to the analysis of particles less than 100 nm in size, as well as to the statistical significance of the analyzed particles representing the entire aerosol population. I think that one should take these limitations into account when interpreting the results from single particle analysis. The authors of this paper should emphasize this point in the discussion.

Even though, information about aerosol morphology and mixing state is critical for modeling the radiative impacts of aerosols. Also, since the aerosol morphology affects its optical properties, this kind of information is important when interpreting data from online aerosol instrumentation like optical size spectrometers and monitors of aerosol scattering and absorption coefficients. In my point of view, the authors discussed and interpreted the data appropriately, relating TEM-EDX single particle analysis with air mass back-trajectories, meteorological conditions and air quality parameters. I have seen recent papers using a similar approach (e.g., Geng et al., 2010 and Niemie et al., 2006). I recommend publication in ACP, after revision of the points I mention below. I strongly suggest an English revision, because the manuscript shows writing quality issues.

Just as the reviewer said, TEM-EDX analysis has a couple of limitation when it is used to study aerosols. In this work, 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.

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.

We have revised the manuscript. In the revised version, we emphasize these limitations in the experimental section.

Specific comments:

(1) Page 20947: a sentence should be added to the abstract, stating that particles were sampled in four different days, with different weather and air quality conditions, and under two particular conditions: one day impacted by the transport of dust aerosols from the Mongolian and Gobi deserts (Nov 12) and three days impacted by air masses coming from the Yellow Sea.

The sentence of "*Particles were sampled under different weather and air quality conditions.*" was added in line 2, page 20974.

(2) Page 20974: Also, it would be interesting to add to the abstract the aged particles were associated with days with low wind velocities, showed complex structures, and were bigger in size.

The sentence of "*Aged particles were associated with days with low wind velocities, showed complex structures, and were bigger in size.*" was added in line 11, page 20974.

(3) Page 20975, Line 6-7: that is not correct to say that light absorption is considered to occur only in soot. Dust and biogenic particles, for example, also absorb light. Please refer to Andreae and Gelencser, 2006 for further information about the current nomenclature of light absorbing aerosols. Also, the reference Adachi and Buseck, 2010 does not seem to fit here.

I have read seriously this paper, and considered this suggestion was correct. The sentence of "which is considered only to occur in soot". The reference of "Adachi and Buseck, 2010" was replaced by "*Andreae and Gelencsér, 2006*" in line 6, 20975 page.

(4) Page 20975, Lines 11-12: a more recent reference concerning to the effect of soot particles on climate would be Ramanathan et al., 2008.

The suggestion is reasonable. "*Ramanathan et al., 2008*" is added in line 12, page 20975.

(5) Page 20978, Line 17: "Experiments and Methodologies" is not a good name, since in my view there was only one experiment.

"Experiments and Methodologies" was replaced by "*Experimental*" in line 17, page 20978.

(6) Page 20979, line 21: How was the NO_x analyzer modified? How does it impact the observations you report? If this is not relevant in this work, just remove the word "modified".

"NO_x was detected with a modified commercial MoO/chemiluminescence analyzer (Thermo Fisher Scientific, Co., Ltd, Model 42i" was changed to "*NO_x was measured by a molybdenum NO_2-to-NO converter using the chemiluminescent reactions.*" in line 21, page 20979.

(7) Page 20982, Line 1 and 4: you say that air masses at 500 m and 1000 m do not affect ground-level air quality. That might not be true, since the referred air masses might be inside the diurnal mixing layer. Please comment on that.

The suggestion is reasonable. The sentence of "500 m and 1000 m do not affect ground-level air quality" was deleted, in line 26, page 20981 and line 4, page 20982.

(8) Page 20982, Line 7: actually, the mass increase is in the 1-10 micrometer size range.

"10 nm-10 μm" was changed to "*1-10 μm*" in line 7, page 20982.

(9) Page 20982, Line 12: fine mode particles were not cleared out. The average total number concentration did not change much from Nov 11 to Nov 12 (Figure 3b). What happened was that the coarse mode increased greatly, and therefore the total particle mass were dominated by the coarse mode.

The suggestion is reasonable. "…indicating that anthropogenic fine particles emitted from local sources were cleared out when the dust storm approached the sampling site and that the dust particles were mostly dominant during the storm period" was changed to "*…indicating that the coarse mode increased greatly when the dust storm approached the sampling site and that the total particle mass was dominated by the coarse mode during the storm period.*", in line 12-14, page 20982.

(10) Page 20983, Line 3: Add a reference to the statement that carbonaceous particles are the major fraction of aerosols by number in urban atmosphere.

In line 3, page 20983, "*Zhou et al., 2009; Ramana et al., 2010*" was added.

(11) Page 20989, Line 11: "as well as through homogeneous nucleation". If you are talking about organic coatings, what matters is the condensation of SOA onto preexisting particles. Homogeneous organic nucleation is controversial.

The suggestion is reasonable. "…, and primary organic aerosol as well as through homogeneous nucleation." was deleted, in line 11, page 20989.

(12) An important result that was omitted from the conclusion section is that aged particles were associated with days with low wind velocities, showed complex structures, and were bigger in size.

The sentence of "*Aged particles were associated with days with low wind velocities, showed complex structures, and were bigger in size.*" was added in the conclusion section, in line 20, 20994.

(13) Figure 3: what particle density was assumed to calculate the mass size distributions? This information should be somewhere in the text.

The sentence of "*Mass size distributions in this work were calculated by assuming an apparent particle density to be 1 g cm^{-3} and particles to be spherical shaped.*" was added in line 29, page 20979.

(14) Figure 6: the information about heterogeneous particle sizes is unclear. Please rephrase that. Also, there is a typo: the correct word is "graphene", and not "grapheme".

In the caption of Figure 6, "heterogeneous" has deleted, and "grapheme" was replaced by "*graphene*"

(15) Figure 7: include letters g and h in the corresponding figures.

In Figure 7, "*g*" and "*h*" have added in the suitable places.

Technical comments: (There are certainly more language errors, please provide a careful revision)

(1) Page 20974, Lines 16-18: this sentence makes no sense, please rephrase it: "With an exception of the sample collected during a dust storm on 12 Novemberember, soil-derived particles (68%) were relatively more frequently observed." I think you are using the word "exception" in a wrong way.

On the basis of this suggestion, we have rephrased this sentence, in lines 16-18, page 20974.

(2) Page 20975, Line 7: did you mean "oxalic acids"?

"oxalic" was changed to "*oxalic acids*" in line7, page 20975.

(3) Page 20975, Line 19-20: rephrase: "Jacobson (2001) SUGGESTS that internally mixed soot particles, COMMONLY PRESENT IN THE ATMOSPHERE, could be the second most significant component of global warming."

We have rephrased this sentence, in lines 19-20, page 20975.

(4) Page 20977, Line 25: remove the adjective "outstanding"

"outstanding" was deleted, in line 25, page 20977.

(5) Page 20978, Lines 6-7: "These factors make Shanghai air pollution DIFFICULT TO CONTROL…"

We have rephrased this sentence, in lines 6-7, page 20978.

(6) Page 20978, Line 11: Define HAADF-STEM.

"HADDF-STEM" was replaced by "*High-angle annular dark-field scanning transmission electron microscopy (HAADF–STEM)*", line 11, page 20978.

(7) Page 20978, Line 19: Typos. Sounds better: "…were collected BETWEEN October and November OF 2010…"

On the basis of this suggestion, we have rephrased this sentence, in line 19, page 20978.

(8) Page 20979, Lines 13-15: You only have 4 samples; therefore it is odd to say that sampling times were mostly between 60-90 s. Suggestion: "Sampling periods ranged between 30-180 s, depending on particle loading".

On the basis of this suggestion, we have rephrased this sentence, in lines 13-15, page 20979.

(9) Page 20979, Line 23: Define API.

"API" was replaced by "*air pollution index (API)*", in line 23, page 20979.

(10) Page 20981, Line 8: "…with CORREPONDING API DAILY AVERAGES OF 71, 298,…"

We have rephrased this sentence on the basis of the reviewers' suggestion in line 8, page 20981.

(11) Page 20981, Line 18-26: to make the text clearer, please include information about what figure you are referring to in each sentence. Ex: Figure 2b, Figure 2c, etc.

The revised version has included this information, in line 18-26, page 20981.

(12) Page 20981, Line 24: typo. "shanghai" (missed capital letter).

"shanghai" was replaced by "*Shanghai*" in line 24, page 20981.

(13) Page 20981, Line 24: For those who are not familiar with China geography, are Jiang-shu and Anhui names of Chinese cities, regions, counties?

"Jiangshu and Anhui" was replaced by "*Jiangshu and Anhui (names of Chinese provinces)*" in line 24, page 20981.

(14) Page 20982, Lines 14-15: rephrase: "AFTER 10:00 LT on 13 November, the influence of the dust storm became less IMPORTANT and the COARSE mode of the mass concentration distribution DIMINISHED.

We have rephrased this sentence in lines 14-15, page 20982.

(15) Page 20982, Line 18: You are actually referring to Figure 4.

We have corrected this error in line 18, page 20982.

(16) Page 20982, Line 19 and 26: Typo:SO_4^{2-} is an anion, not a cation.

We have corrected this error in line 19 and 26, page 20982.

(17) Page 20983, Lines 15-16: "Such particles ranged in diameter from 200 nm to 1.5 μm."

"Such particles ranged in diameter from 200 nm to 1.5μm, mostly from 200 nm to 1 μm" was changed to "*Such particles ranged in diameter from 200 nm to 1.5μm*" in line 15-16, page 20983.

(18) Page 20983, Line 28: you are actually referring to Figure 5d.

We have corrected this error in line 28, page 20983.

(19) Page 20984, Line 14: you are actually referring to Figures 5d and 5e.

We have corrected this error in line 14, page 20984.

(20) Page 20984, Line 18: you are actually referring to Figures 5c and 5d.

We have corrected this error in line 18, page 20984.

(21) Page 20984, Line 20: you are actually referring to Figure 5f.

We have corrected this error in line 20, page 20984.

(22) Page 20985, Lines 1-2: rephrase: "STEM-EDX mapping shows THAT the soot aggregate MOSTLY contained C and, to a lesser extent, O (Fig. 6b and 6c)."

We have rephrased this sentence on the basis of the reviewer's suggestion, in lines 1-2, page 20985.

(23) Page 20987, Line 16: remove quotes from tar ball.

The quotes have removed in the revised version, line 16, page 20987.

(24) Page 20989, Lines 16 and 18: you are actually referring to Figure S3.

We have corrected this error.

(25) Page 20989, Lines 6-8: rephrase "Organic coatings were lost under strong beam bombardment although they were less sensitive to the electron beam than sulphates and nitrates, which were different from tar ball and POCs.

This sentence will make readers confused. In the revised version, we changed it to "*Organic coatings were lost under strong beam bombardment*.", in lines 6-8, page 20989.

(26) Page 20989, Lines 13-14: "…suggested that the formation of SOA may be a significant pathway…". If SOA is formed, it is already in the particle phase.

In the revised version, "in the vapour phase" was deleted in this sentence, in lines 13-14, page 20989.

(27) Page 20989, Line 23: "…(see the section 3.4)."

"the section of relative abundances" was changed to "*the section 3.4*" in line 23 page 20989.

(28) Page 20989, Lines 24-25: "and exhibits a smooth water uptake, increasing THE PARTICLE SIZES".

We have rephrased this sentence, in line 24-25, page 20989.

(29) Page 20990, Lines 6-7, Rephrase: "Their size distributions were bimodal, with most particle diameters <500 nm or from 800 nm to 1.5 μm."

The sentence of "Their size distributions were bimodal, with most particle diameters <500 nm or from 800 nm to 1.5 μm" was changed to "*The most particle diameters were < 500 nm or from 800 nm to 1.5 μm.*"

(30) Page 20991, Line 5: "…and eventually make them efficient SCATTERS OF SOLAR RADIATION AND CCN".

We have rephrased this sentence, in line 5, page 20991.

(31) Page 20991, Lines 15-16: "The attached sulphates could AFFECT THE HYGROSCOPICITY OF THE PARTICLES, FAVORING THE ACTIVATION TO CCN."

We have rephrased this sentence, in line 15-16, page 20991.

(32) Page 20992, Lines 2-3: "… which is Characteristic of H_2SO_4 particles…".

We have rephrased this sentence, in line 2-3, page 20992.

(33) Page 20992, Line 9: "… AMMONIUM sulphate species…"

We have corrected this error.

(34) Page 20992, Line 24: typo: "…(R_2 of 0.74),…"

We have corrected this error.

(35) Page 20992, Line 26: use southeast instead of east south.

In the revised version, "east south" was replaced by '*southeast*" in line 26, page 20992.

(36) Page 20993, Lines 8-9: "…regarding the INCREASED CONCENTRATION of sea salt particles…".

We have rephrased this sentence in lines 8-9, page 20993.

(37) Page 20993, Lines 19-21: You are misusing the word "exception". Sounds better: "Specifically for the sample collected on Nov 12, soil derived particles were more abundant (68%) as compared to sulphates (15%) and soot (7%). That does not hold for the other samples, in which sulphate were predominant."

We have rephrased this sentence on the basis of the reviewer's suggestion in lines 19-21, page 20993.

Preferences:

(1) Andreae, M. O. and Gelencsér, A.: Black carbon or brown carbon? The nature of light-absorbing carbonaceous aerosol, Atmos. Chem. Phys., 6, 3131-3148, 2006.
(2) Geng, H. et al., Characterization of individual submicrometer aerosol particles collected in Incheon, Korea, by quantitative transmission electron microscopy energy-dispersive X-ray spectrometry, J. Geophys. Res, 115, D15306, 2010.
(3) Niemi, J.V. et al., Changes in background aerosol composition in Finland during polluted and clean periods studied by TEM/EDX individual particle analysis, Atmos. Chem. Phys., 6, 5049-5066, 2006.
(4) Ramanathan, V. and Carmicheal, G.: Global and regional climate changes due to black carbon, Nature Geosci., 1, 221-227, 2008.

In the revised version, we have cited these literatures.

We acknowledge the comments of the reviewer. In the revised version, we have corrected a large number of English errors of MS, including incomplete and grammatically incorrect sentences and misspelling.