Atmos. Chem. Phys. Discuss., 11, C8250–C8256, 2011
www.atmos-chem-phys-discuss.net/11/C8250/2011/

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 #2

Received and published: 26 August 2011

General comments:

The paper presents information about the composition and morphology of carbonaceous particles in Shangai, 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 Niemi 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.

Specific comments:

1) Page 20974: 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.

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

3) Page 20975, Lines 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.

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Please refer to Andreae and Gelencsér, 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.

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.

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

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

7) Page 20982, Line1 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.

8) Page 20982, Line7: actually, the mass increase is in the 1-10 micrometer size range.

9) Page 20982, Line 12: fine mode particles were not cleared out. The average total number concentration did not change much from Nov11 to Nov12 (Figure 3b). What happened was that the coarse mode increased greatly, and therefore the total particle mass was dominated by the coarse mode.

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.

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 pre-existing particles. Homogeneous organic nucleation is controversial.

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,

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and were bigger in size.

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

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”.

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

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

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.

Page 20975, Line 7: did you mean “oxalic acids”?

Page 20975, Lines 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.”

Page 20977, Line 25: remove the adjective “outstanding”

Page 20978, Lines 6-7: “These factors make Shanghai air pollution DIFFICULT TO CONTROL. . ."

Page 20978, Line 11: Define HAADF-STEM.

Page 20978, Line 19: Typos. Sounds better: “. . .were collected BETWEEN October and November OF 2010. . .”

Page 20979, Lines 13-15: You only have 4 samples, therefore it is odd to say that

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sampling times were mostly between 60-90 s. Suggestion: “Sampling periods ranged between 30 to 180 s, depending on particle loading”

Page 20979, Line 23: Define API.

Page 20981, Line 8: “. . . with CORREPONDING API DAILY AVERAGES OF 71, 298, . . .”

Page 20981, Lines 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.

Page 20981, Line 24: typo. “shangai” (missed capital letter)

Page 20981, Line 24: For those who are not familiar with China geography, are Jiangshu and Anhui names of Chinese cities, regions, counties?

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.

Page 20982, Line 18: You are actually referring to Figure 4

Page 20982, Line 19 and 26: Typo: SO42- is an anion, not a cation.

Page 20983, Lines 15-16: “Such particles ranged in diameter from 200nm to 1.5 μm.”

Page 20983, Line 28: you are actually referring to Figure 5d

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

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

Page 20984, Line 20: you are actually referring to Figure 5f

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).”

Page 20987, Line 16: remove quotes from tar ball

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Page 20987, Lines 16 and 18: you are actually referring to Figure S3

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.”

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.

Page 20989, Line 23: “. . . (see the section 3.4).”

Page 20989, Lines 24-25: “and exhibits a smooth water uptake, increasing THE PARTICLE SIZES”

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

Page 20990, Line 8: “. . .TEM-EDX showed THAT many of the AMMONIUM ammonia sulphate grains. . .”

Page 20991, Line 5: “. . . and eventually make them efficient SCATTERS OF SOLAR RADIATION AND CCN.”

Page 20991, Lines 15-16: “The attached sulphates could AFFECT THE HYGROSCOPICITY OF THE PARTICLES, FAVORING THE ACTIVATION TO CCN.”

Page 20992, Lines 2-3: “. . .which IS characteristic of H2SO4 particles. . .”

Page 20992, Line 9: “. . . AMMONIUM sulphate species. . .”

Page 20992, Line 24: typo: “. . . (R2 of 0.74),. . .”

Page 20992, Line 26: use southeast instead of east south

Page 20993, Lines 8-9: “. . .regarding the INCREASED CONCENTRATION of sea salt particles. . .”

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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.”

References:

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.

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.

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.

Ramanathan, V. and Carmichael, G.: Global and regional climate changes due to black carbon, Nature Geoscience, 1, 221-227, 2008.

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