3# Referee's comments:

Characterization of light-absorbing carbon particles at three altitudes in East Asian outflow by transmission electron microscopy Zhu et al.,

The morphology, microstructure, and composition of the submicron fraction of individual light-absorbing carbon (LAC) particles were studied in three different altitudes in low troposphere during the ACE-Asia. Although the LAC particles is one kind of particles in atmospheric science communication, the study is not very surprised. Most of the study concern the technique to know the soot and LAC particle but not much atmospheric science or chemistry. In light of completely investigating physical properties of the absorbing carbon particles, the paper can be published the atmospheric chemistry and physics.

The low magnification TEM images in Figure 1 show that most of these particles are organic particles and soot. I have many questions about the samples.

(1) Does the aerosol during the sampling period is very unique? As one of the reviewer' comment, they did found many particles were mixture of sulfate, soot, organics. In particular, sulfate particles were dominated at the Mt. Tai in the upper altitude (Science of the TotalEnvironment 430: 217–222 and Atmos. Chem. Phys. 2011, 11, (22), 11733-11744.). Such kind of particles are expected to exist in the troposphere over the Yellow sea. Ramana et al., found the extent of black carbon induced warming in East Asia is dependent on the concentration of sulphate and organic aerosols which coated on soot particles in East Asian outflow. From the reviewing similar papers, most study found spherical organic particles were nearby the sources, for example, Li et al., 2012 found similar spherical organic particles in the dry haze layer induced by coal combustion for industrial or house-hold heating. Posfai et al., 2004 and Adachi et al. 2011 found spherical organic particles (tar ball) nearby the biomass burning sources. Therefore, the author need to explain why the sample in this study was different with others in previous studies. Because the authors only collected sample during one flight time, I suppose that the sample should be associated with one dry cold front. Or the collect pathway nearby some large sources?

Response:
We found sulphate particles besides the soot and organic carbon. However, in this work, we only focused on carbonaceous aerosols, although we agree the sulphate or coating on the carbonaceous aerosols would change their radiative forcing. We did not find any coated soot. We agree with that soot can be coated in the atmosphere, however, it is not necessary to be coated and was not observed in the cases described. Aggregation (not coating) with minor sulfate is common in some samples, but depends upon the air mass. Unpublished time-resolved data from an elevated ground site (1100 m) on Jeju Island

show an increase in aggregation of dust and black carbon with sulfate before the same frontal passage as encountered between flights RF-13 and RF-14 and then dropping off in the cooler air behind the front. There was no convincing evidence of significant aggregation of brown carbon with sulfate in the low electron-dose automated SEM results (the SEM methods used are different from the TEM methods described here).

(2) The authors show light-absorbing carbon (LAC) particles include black carbon, brown carbon, and humic –like substances. In the TEM study, it is difficult to determine the light-absorbing carbon, from the study of Alexander, et al., 2008. Based on the conclusion of Alexander, et al., 2008, the study almost supposed all of these visible organic particles were light-absorbing carbon. In the atmosphere, there are brown carbon, black carbon, and colorless organic carbon (Andreae et al.,2006). Could the authors explain where the colorless organic carbon (they should be there) occurred in the samples? Do the author can directly use the previous conclusion from Alexander, et al., 2008? If in other places, scientists found similar spherical organic particle, can they claim them as the light-absorbing carbon? Therefore, the standard used in this study should be provided.

Response:
The optical properties of the different carbonaceous aerosols cannot be determined directly by TEM images, however, they may be determined by electron energy-loss spectroscopy of the TEM (as shown in the previous work of Alexander et al, 2008). Alexander's work showed that the larger amorphous spheres in that air mass were brown carbon and made no statement about the optical properties of other carbons. The current manuscript focusses on determine the morphology, structure and size distribution of soot and amorphous carbons at different altitudes. The complex issue of optical property determination will be the topic of a different paper.

Results and Discussion

(3) P32954 "Typical EDS indicate that the spherical soot contains less silicon and oxygen than that of non-spherical ones." The EDS only can give the semi-quantitive data. Based on the minor silicon in individual particles, the comparison is not significant. Therefore, it is not enough to make the conclusion. About silicon in the individual particles, some references were needed here.

Response:
Even though EDX in this case analysis may be semi-quantitative, we see at least a factor of 5 differences in the relative Si/C ration in the two types of soot. We have modified the manuscript to put in these numbers.

(4) P32955 Line 9-10, In section 2, Experimental, the authors suggest that the lower altitude was influenced by local pollution including ships and fishing boats. However, line 9-10, the soot was proposed from biomass burning. How to explain the contradiction?

Response:
There were small spherule soot and large spherule soot found in altitude of 120 m. It is well-known that the potassium is a characteristic feature of aerosols from biomass burning. So, it is no doubt that the small spherule soot with potassium is from the biomass burning. In section 3.3, we supposed the large spherule soot without potassium found only in altitude of 120 m were from the local pollution including ships and fishing boats. We have modified the text to make this clearer.

(5) P32957 Line 33-25, this is very interesting method to know 3-D particle shape. Unfortunately, the authors didn't introduce the detailed method. The detailed process or methods should be provided in the section 2.

Response:
Image intensity analysis was performed on some particles to obtain information on the 3-D particle shape, we transferred part of the method from discussion into the section 2 and added more details about the method used.

(6) P32959 line 1-10, the authors observed the spherical organic aggregation in the samples, showed in Figure 8e. I don't agree with the explanation. From the Figure 8e, these spherical particles should connect each other instead of physical overloading aggregation. Why did the author think such aggregation could not exist in the atmosphere.

Please see the references, they did found similar particles in the air.

Response:
We see how these statements could be confusing so we have deleted the last two sentences of the paragraph to make this clearer.

(7)P32947 Line 17 CNN-CCN

Response:
Thanks. We changed it in manuscript.

(8)P32949 Line 2 Due to

Response:
Thanks. We changed it in manuscript.

(9)P32961 line 17 120 m

Response:
Thanks. We changed it in manuscript.