**1# Referee's comments:**

This paper describes the characterisation of carbon particles collected at three different altitudes during one research flight as part the ACE-Asia project. Single aerosol particles are analysed using transmission electron microscopy imaging. Particle chemical composition is determined using electron dispersion spectrometry.
The authors showed that aerosol particle sizes and morphologies can vary greatly at different altitudes. In addition to soot particles a high percentage of brown carbon particles were measured. These carbon particles contributed the most to aerosol volume (relative to the soot fraction). The authors highlight that the physical and chemical properties of LAC can vary greatly with altitude and that care should be taken when interpreting measurements from ground based stations. This paper is well written and I think that it is suitable for publication in atmospheric chemistry and physics. However, I have some minor comments below.

Comments:
Experimental:
Please include a short description of the main aerosol inlet on the aircraft? Flow rates, inlet diameters. Was the flow isokinetic? What was the measured RH in the sampling line? What was the sampling time? Did the sampling time change depending on different altitudes (to account for different aerosol concentrations?)

Response:

The inlet used on the NCAR C-130 was a low-turbulence inlet (LTI) described by Huebert et al. (2004) and was controlled by a dedicated operator. The inlet tip was operated isokinetically, but at a research speed of about 100 m/s, avoiding downstream losses in the larger sizes is always a challenge, hence the inlet design that was used. The LTI was optimized to collect particles with diameters up to about 10 µm and in fact tended to enhance the largest sizes. For particles in the size ranges described in this paper, the inlet appeared to reproduce results from wing-mounted probes. The absolute concentrations of brown carbon and dust are calculated from polycarbonate filter samples identical to those discussed by Huebert et al. (2004) and used to characterize the LTI. Ambient RH was measured outside the cabin but would have varied within the inlet. The sample from the MBL had high ambient RH. Sampling time for level legs was determined by the needs of various investigators on the NCAR C-130 and not decided by preset criteria. The sampling time changed on different altitudes, as shown in Table 2, 18, 26 and 18 min were used for 120, 450 and 1500 m altitudes.

Huebert, B.J., S.G. Howell, D. Covert, T. Bertram, A. Clarke, J.R. Anderson, B.G. Lafleur, W.R. Seebaugh, J.C. Wilson, D. Gesler, B. Blomquist, and J. Fox, PELTI: Measuring the passing efficiency of an airborne low turbulence aerosol inlet, Aerosol Science and Technology, 38 (8), 803-826, 2004.

Are there other published results from measurements taken alongside the impactors (aerosol chemistry, BC) that can be used to give a more global description of this flight.

The authors mention how LAC can affect CCN concentrations and light scattering measurements. Were CCN measurements operating alondside the impactors and do CCN properties vary with altitude.
Samples are representative on one day of sampling. How do the authors think these measurements represent longer time periods or certain air mass sources?

Response:

Results from the previous flight, RF-13 on April 23, and some average data for multiple flights during ACE-Asia are discussed by Clarke et al. (2004). Data from a variety of instruments, including bulk optical measurements, are described. RF-13 was the leading moist, warm side of a cold front and RF-14 on the trailing dry, cool side. Strong variation in a wide range of parameters with altitude was encountered on all the flights. However, the range of black carbon and brown carbon types observed by SEM were similar. SEM images of LAC from RF-13 are included in Clarke et al. (2004).

Size distributions and mixtures of dust and black carbon aerosol in Asian outflow: Physiochemistry and optical properties, Clarke, A. D.; Shinozuka, Y.; Kapustin, V. N.; Howell, S.; Huebert, B.; Doherty, S.; Anderson, T.; Covert, D.; Anderson, J.; Hua, X.; Moore, K. G., II; McNaughton, C.; Carmichael, G.; Weber, R., J. Geophys. Res., 109, doi: 10.1029/2003JD00437 (2004)

There is a large amount of unpublished data that suggest that the variation with altitude observed for RF14 is typical.

Results and Discussion:
Since the focus of this manuscript is describing the physical and chemical properties of organic particles, a discussion of their contribution to the overall aerosol particle population should be included in order to provide a better idea of their importance in the atmosphere.

Response:

From SEM analysis, the number concentrations of brown carbon are about, 0.3 $x10^5$, $1x10^5$ and $4x10^5$ particles per standard liter of air at altitudes of 120, 450 and 1500m, respectively. On the other hand, the number concentrations of silicate-bearing dust particles (aluminosilicates, such as clay and feldspar and quartz) are about $2.1x10^3$, $7.5x10^3$ and $4.5x10^3$ particles per standard liter of air at altitudes of 120, 450 and 1500m, respectively. So, from the point of view of number concentrations, the brown carbon makes a large contribution to the overall aerosols particle population.

For the volume concentrations of brown carbon, they are 0.4 x $10^3$, 1 x $10^3$, and 2.3 x $10^3$ $\mu m^3$ per standard liter at altitudes of 120, 450 and 1500m, respectively. The volume concentrations of silicate-bearing dust particles are 5.5x $10^3$, 34 $x10^3$ and 24 $x10^3$ $\mu m^3$ per standard liter at altitudes of 120, 450 and 1500m, respectively.

The dust is dominant by volume because of the larger particle diameters, but even considering the size difference the brown carbon is a significant fraction of total aerosol mass.

Notes: 120m is the marine boundary layer. 450 m is the dust layer. 1500 m is the layer that contains less dust but more pollution.

What was the fraction of aerosol particles that contained soot compared to the total aerosol concentration measured, how did this fraction change as a function of altitude?

Response:
The number concentrations of soot spherules (not soot particles) are about, $2.8x10^4$, $7.1x10^4$ and $3.8x10^4$ particles per standard liter of air at altitudes of 120, 450 and 1500m, respectively.
As a fraction of the total number concentration of brown carbon, soot spherules and silicate-bearing dust particles in the samples (there are some other less abundant particle types), the fraction of soot spherules in population are about 46%, 38% and 9% at altitudes of 120, 450 and 1500m, respectively.
However, the total volume concentrations of soot spherules is 2, 20, 20 $\mu m^3$ per standard liter. The volume fraction is $3 \times 10^{-2}$% and $6 \times 10^{-2}$% and $6 \times 10^{-3}$% at 120, 450 and 1500 m.

Is the relative abundance shown in Figure 5 for the total aerosol composition or just aerosol particles containing soot?

Response:
This is just the particles containing soot.

P32955, Line 9: In section 3.3 the authors suggest different sources of the organic carbon. However, they do not discuss other types of aerosol particles collected on the grids. A short description of the other types of aerosol particles on the grid and the overall contribution of LAC particles would allow better source apportionment.

Response:
In our response to the above comments, we gave the number concentration of brown carbon, soot and silicate-bearing dust particles at three altitudes. We add a short description about it in the manuscript.

Figure 5 and elsewhere in the manuscript, how is the aerosol particle diameter defined?

Response:
This is a good question which becomes important for aggregates of soot spherules. The outer boundary of a spherule is identified by the outer most graphene sheet belonging to

each spherule. The diameter is then taken as the distance between the outer boundary on opposite sides of the spherule. The manuscript has been changed to clarify this point.

Figure 6, please describe line profiles a bit better.

Response:
The line profile is the image intensity plotted as a function of position along a line in the image. In TEM images, the intensity will be highest in the vacuum area (no sample), and will drop on the samples because the electron beam was blocked somehow (by absorption, reflection or diffraction). For the first approximation of thin specimen in our case, the intensity at each point of the TEM images is linearly proportional to the mass thickness of the specimen.

We made changes in the manuscript and explained the line profile procedure more clearly on page 11, Section 3.2, 2$^{nd}$ paragraph.

P32953, line 19: Is it possible that the silicon is coming from the window in the EDS detection system?

Response:
We do not think the Si is coming from the detector.

P32953, line 21: What is the fractal dimension of a sphere using your method, and how does your fractal dimensions compare with other studies (Adachi et al., 2010, Xue et al., 2009).

Response:
We deleted the part of fractal dimension.

P32954, line 12: Has the EDS been calibrated so to be able to make quantitative conclusions on the measurements?

Response:
Typical EDS (Fig. 3e and f) show that the atomic percentages of carbon, oxygen and silicon in spherical soot are 99.5, 0.3 and 0.2, respectively and those values in nonspherical soot are 94.7, 4.1 and 1.2, respectively. The spherical soot contains less silicon and oxygen than that of non-spherical ones. We made changes in the manuscript.

P32956, Line 15: “ in different operating conditions”..

Response:
Thanks, “operation” changed into “operating”.

P32960, Line 1: " ..are from tens of particles". If Figure9a is representative of more than 11 points, how are the 11 points calculated? Are they averaged over a range of particle thickness? If so can you include error bars? (Figure 9a and b).

Response:
In Figure 9a, there are 11 data points and in Figure 9b, there are 14 data points. We made changes in the manuscript.

P32964, Line2: what was the composition, size, and morphology of aerosol particles that the soot was internally mixed with?

Response:
From our samples study, in most case, the soot was attached to the amorphous carbon spheres. The composition, size and morphology of the amorphous carbon spheres were discussed in the manuscript. We shorten our conclusions as suggested by one reviewer and delete part of the non-important conclusions.

Minor comments
P32951,Line 6: insert comma, "..for stage 3, 0.05 to 0.3 microns"

Response:
Thanks. We changed it in manuscript.

P32951, Line 10: Reword sentence and define SEM.

Response:
Thanks. The sentence from line 9-12, changed to: We performed scanning electron microscopy (SEM) observations of single-stage filter samples for which absolute number concentration over the entire size range up to about 10 microns was determined. The results presented here are primarily from stage 3. Based on these observations, the size fraction on the stage 3 has the largest number concentration.

P32951, Line 14:"..equipped with a thin window light elemental...

Response:
Thanks. While, we think it is better to keep element, not elemental, because the detector here is for the detecting elements including heavy and light elements.

P32952, Line 23: For altitudes.

Response:
There is no "altitudes" in line 23, the one in Line 22, We changed it to : "For the altitude"

P32959, Line16: "Was used to investigate"

Response:

Thanks. We changed it in manuscript.

P32959, Line 27: "..and has a linear relationship"

Response:
Thanks. The sentence of Line 26 and 27, was reworded: As shown in Fig. 9a, the $ln\ (I_0/I_t)$ versus $t$ curve is linear over a wide size range (here $t$ is the measured diameter of the carbonaceous spheres) and has a slope of $\alpha$.

P32960, Line 20: Sometimes "to" is used to define a range and other times "-" is used. Please be consistent and use the same terminology.

Response:
Thanks, we modified it through the manuscript and used the "-".

P32960, Line 25: "The small quantity of large spheres above 500 nm contributes about 18
Alot of percentages are used to describe the aerosol particle population in this section. However, no errors are reported. The authors mentions that "about 70

Response:
A lot of percentages (i.e. 70%) were used to give a general sense of the range that most of the particles falls in. This 70% is a roughly estimation, can range from 65-75%.

P32961, Line 16: 120 m

Response:
Thanks. We changed it in manuscript.

P32962, Line 26: These larger particles will have much more important consequences on aerosol light scattering and CCN. Why do you not discuss them further?

Response:
The large cenospheres are in very low concentration as measured by SEM and their contribution to bulk optical properties would be correspondingly low. There are higher concentration of carbon cenospheres in other flights and in those cases their optical effects may be significant.