Anonymous Referee #2

The subject manuscript compares the size distributions of n-Akanes, PAH, Hopanes and during different seasons at three contrasting sites in East Asia. The experimental methods and data analysis tools are reasonable well documented in the literature and as the authors state on page 13862, lines 5-8 that similar studies have been conducted in the costal cities of China but this is the fist study to examine inland sites. Although the manuscript does present interesting data for three very different locations, the manuscript does not present advances in methods, data analysis, or new insights into atmospheric science. As a result the study is more of local interest and I do not believe that the manuscript will be or broad interest to the readership of ACP. The authors need to better highlight the novelty and advances associated with the paper before publication in ACP. In addition, I have some concerns about the manuscript that need to be addressed before the manuscript should be considered for publication.

Response: We appreciate the reviewer's very valuable comments and suggestions. Many studies have focused on the aerosol pollution in East Asia, especially in China, because the air pollution from China can be transported into the North Pacific region and even into North America (Wilkening et al., 2000; Vancuren and Cahill, 2002; Aldhous, 2005). Much more coal and biomass are burned in China with minimal emission controls in some cases, adding more absorbing soot and organic aerosol to the Asia and Pacific atmospheres (Huebert et al., 2003), which makes the air pollution in China different from those in other countries in the world. Moreover, satellite observations showed that the level of nitrogen dioxide in the atmosphere over eastern China has exceeded those in Europe and North America (Akimoto, 2003), together with high levels of gaseous (Wittrock et al., 2006) and solid-phase carbonyls (Wang et al., 2009) , suggesting the atmospheric environment in East Asia may be changing. The current study demonstrated that benzo(b)fluoranthene is the most abundant PAH in the aerosols due to coal burning for house heating and cooking although the number of cars has sharply increased recently. Such a PAH composition is different from that in Europe and North America, where benzo(ghi)perylene is the most abundant particulate PAH mainly due to vehicle emission. The current study, to be our knowledge, is the first time to simultaneously measure the size distribution of organic aerosols from inland, mountain and marine atmospheres over East Asia. The results are very helpful for readers to better understand the current status of air pollution in East Asia and the impact on size distribution of aerosols by source properties, meteorological conditions and other atmospheric processes such as evaporation, condensation and photo-oxidation. Therefore, we believe our results are of not only local/regional but also global importance.

General Comments

1) The numbers of samples used in the manuscript are very small and no information is provided concerning the representativeness of the measurements for the locations studied.

Response: Actually, the number of samples used in this paper is N=135, which is composed of 6 sets (nine samples for each set, thereinafter) for the urban site in winter and spring, 4 sets for the mountaintop in winter and summer and 5 sets for the marine site in spring. Consistent results have been obtained in the current study. Therefore, we think the representiveness of the measurement is good enough, although the sample number is not very big. Similar levels of sample number and sampling duration were also applied by some other studies for the size distribution measurement of organic aerosols, for example, Venkataraman et al., 1994; Kavouras and Stephanou, 2002 and Kleeman et al., 2008.

2) The statistical aspects of the data analysis are virtually non-existent in the manuscript. Throughout the manuscript, the authors make statements about peaks in the size distribution and peaks in the distribution of compound concentrations and ratios that are not supported by the data given the reported uncertainties. Likewise, the authors present considerable comparisons of GMD but provide not information of the uncertainties obtaining these results from the measurements.

Response: We disagree with the comment above. Standard deviations were given for all the data, which can be taken as one of the statistical aspects. Moreover, relative standard deviations of the diagnostic ratios and GMDs at each site are in general less than 10% for the major biomarkers, i.e., n-alkanes and PAHs (see Table 3 and Table 6 in the revised paper). Similar statistic analysis is also applied by others (e.g. Kavouras and Stephanou, 2002). Thus we believe the data reported by this study are accurate and the related discussions are reasonable.

3) The manuscript tends to infer an understanding of the sources of organic carbon in many locations but the presented analysis only addresses three categories of tracers and is not sufficient to infer results about organic carbon sources or distributions. Such analysis is possible with the measured data but is not presented and

comparison to source profiles that include organic carbon since organic carbon was not measured in this study or at least not reported.
Response: Unlike biomarkers, organic carbon does not give the fingerprint of organic aerosols, thus we did not measure the organic carbon and saved the filter aliquots for the measurement of water-soluble organics. In this study we only focused on the three classes of biomarkers to discuss their sources and size distributions. We identified their sources at the three sites by comparison with different source profiles, for examples, the molecular compositions of *n*-alkanes, PAHs and hopanes in different sources.

4) The three samples location are actually very far from each other and the local sources impacting these sites as well as the long range transported sites could also be quite different. To this end, the pairwise comparison of the data from the sites seems inappropriate in the context of sources. In this context, I question the conclusion stated at the end of the abstract that the differences are solely due to atmospheric processing.
Response: We did not find any point source around the three sampling sites, thus the local source impact is insignificant. As mentioned above, we identified the sources of the biomarkers at the three locations by comparison the molecular compositions of the compounds in the samples with those in different sources and further discussed the differences in the size distributions. Similar measurements were also conducted by others for the comparison of primary and secondary organic aerosol in urban, marine and forest environments (Kavouras and Stephanou, 2002). Therefore we do not think the comparison made in the current study is inappropriate. However, we agree with the reviewer that differences in the size distributions are not solely dependent on the atmospheric process such as repartitioning and coagulation. Other factors such as increased hygroscopicity and differences in sources among the three sites may also be attributable to the difference in GMDs. We modified the related statement.

Specific Comments
1) Title – I am not sure that the term "n-hydrocarbons" is appropriate. I think this should be n-alkanes. I am not familiar with the terms n-hydrocarbons.
Response: We changed the title "n-hydrocarbons " as "*n*-alkanes".

2) Page 13862, lines 18-26- The duration of sample collection was never specified. How long were samples collected? It seems that they were collected for several if not more days. What has been done to demonstrate that the long sample times did not impact the measurements?
Response: Sampling information including the duration and the sample numbers had already been presented in the ACPD version by Table 1. As reported by Watson et al (2008), long-term sampling is necessary, because the approach can minimize the artifact from field blanks. Moreover, the long-term sampling applied in the current study is also necessary in order to obtain enough materials, especially at the mountain and marine sites.

3) Page 13863, line 28 – The statement that "no serious contamination was found in field blanks," is very subjective. A more quantitative presentation of the blanks and other QA/QC should be presented.
Response: Suggestion taken. We added more discussion on the QA/QC in the experiment section.

4) Page 13864, lines 4-5 - I see no reason that the authors should "advertise" a forthcoming paper. How is the fact that measurements are not presented here important the current paper?
Response: We deleted this sentence.

5) Page 13865, lines 3-5 – The assessment of pollution in Baoji is very anecdotal. Can the authors support this claim? Are the authors referring to PM, organic aerosol, or the compounds measured in this study.
Response: We deleted the related discussion here.

6) Figures 2, 3, and 4 – Error bars should be presented to see if the conclusions stated by the authors are robust
Response: Suggestion taken. We changed the format of the figure with error bars representing the standard deviation.

Reference:

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