Anonymous Referee #1

General comments

This paper reports particulate-phase concentrations of n-alkanes, PAHs, and hopanes from three sites in China and Japan. Total concentrations and size-resolved ones are presented for different seasons and disparate environments: urban, mountain, and marine. The paper is clearly written, and the results are clearly presented. The analysis is interesting and informative. An exploration of diagnostic ratios within each major category of organic compound helps draw a contrast between influential sources at the different sites. The paper also contrasts GMDs and size distributions at each site and concludes that differences are due to coagulation and repartitioning of species during long-range transport. However, it overlooks the possible role of water content in influencing size distributions. The downwind marine environment is likely more humid, and oxidation of the organic compounds–both those analyzed within this work and others not analyzed here–during transport may have increased the particles' hygroscopicity. Because such data from East Asia are relatively newer and because the region's sources are different, I would like to see a discussion of how these results compare with similar measurements in Europe and North America. The large influence of coal in East Asia is likely to produce interesting contrasts. A few minor revisions, suggested below, will help strengthen the paper.
Response: We thank the reviewer's comments and agree with him/her that the hygroscopic growth of aerosols during the long-range transport from inland China to the North Pacific may also be responsible for the differences in the aerosol sizes. The related discussions are revised. As suggested by the reviewer, a comparison with similar measurement in Europe and North America was supplemented to identify the difference in the sources of PAHs. We added a new table (Table 4, see below) to compare the source profiles of PAHs with those in the ambient aerosols, and found that benzo(ghi)perylene (BghiP) is the most abundant particulate PAH in Europe and US mainly due to vehicle emission. Whereas benzo(b/k)fluoranthene (BbkF) is the most abundant solid-phase PAH in China and even in the North Pacific because coal burning for house heating and cooking is the major source in the country, although the number of mobile vehicles has increased sharply in the recent years. This results are consistent with the previous studies conducted for fourteen Chinese cities (Wang et al., 2006; Wang et al., 2007). The related revision can be seen in the paper section 3.1.2. Moreover, based on the molecular composition, we also found that particulate hopanes in inland Chinese regions like Baoji city are mostly originated from coal burning, in contrast to those in eastern China, where hopanes are mostly derived from vehicle exhausts (Wang et al., 2009). Such a difference in the molecular compositions of PAHs and hopanes is more significant in winter when burning coal for house heating (Wang et al., 2007). The related discussion is also revised and added into the section 3.1.3.

Specific comments

1. (Title) The title reads "n-hydrocarbons" but really the paper only analyzes n-alkanes, so the title could be more specific.
Response: Suggestion taken. We already changed the title.

2. (p. 13864, line 22) Since this section emphasizes differences in C23-C27 versus C31, it would be useful to know what are the major sources of smaller versus larger n-alkanes, or at least how n-alkane speciation profiles differ between major sources. Such information might add further insight to identifying the dominant sources at each location. The paper later analyzes the carbon preference index, but this addresses the ratio of odd to even n-alkanes and not the broader speciation.
Response: Suggestion taken. We added one sentence into the related text "*n*-Alkanes originated from plant wax are dominated by high molecular weight (HMW) ones (>C_{25}) (Hays et al., 2005), in contrast to *n*-alkanes derived from fossil fuel combustion, which are dominated by low molecular weight (HMW) ones (<C_{25}) (Rogge et al., 1993; Simoneit et al., 2004).".

3. (p. 13870, lines 7-8) As mentioned in the general comments above, a simple analysis of GMDs does not rule out the possibility for increased water content of particles in the marine atmosphere. The argument for enhanced repartitioning of organics to explain larger GMDs would be strengthened if it examined changes in the ratios of individual species at different sites relative to their vapor pressures.
Response: We agree that increased water content of particles in the marine atmosphere may also contribute to the larger GMDs compared to those in the urban and mountain atmosphere. Related discussions are modified. We have examined changes in the ratios of individual species at different sites relative to their vapor pressures, and found it is not so helpful when explaining the larger GMDs. Instead, we used the concentration ratios of fluoranthene and pyrene to benzo(ghi)perylene in each stage to elucidate the larger GMDs mainly caused by the repartitioning of the pollutants. As shown in the new figure (i.e. Figure 7a-b, see below), the difference between the ratios of Flu/BghiP and

Pyr/BghiP increase as the increase of particle sizes, suggesting Flu migrates toward larger particle faster than does Pyr. Gas-particle partitioning of pollutants depends on the vapor pressure of each species and the surface area of particles. Vapor pressure (p=1.23 × 10^{-3} Pa at 25°C) (Finlayson-Pitts and Pitts Jr., 2000) of Flu is larger than that (p=6.0 × 10^{-4} Pa) of Pyr, thus Flu repartitioning toward larger particle faster than pyre. The new figures and related discussion is also supplemented in the revised paper, section 3.2.2).

Technical corrections

4. (p. 13863, line 20) Define BSTFA.

Response: Suggestion taken.

5. (p. 13864, line 15) "Air pollution" is always singular, not plural.

Response: Suggestion taken.

Reference

Finlayson-Pitts, B. J. and Pitts Jr., J. N., *Chemistry of the Upper and Lower Atmosphere*, Academic Press, San Diego, 2000.

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Wang, G., Kawamura, K., Xie, M., Hu, S., Cao, J., An, Z., Watson, J. G. and Chow, J. C.: Organic molecular compositions and size distributions of Chinese summer and autumn aerosols from Nanjing: Characteristic haze event caused by wheat straw burning, Environmental Science & Technology, 43, 6493-6499, 2009.

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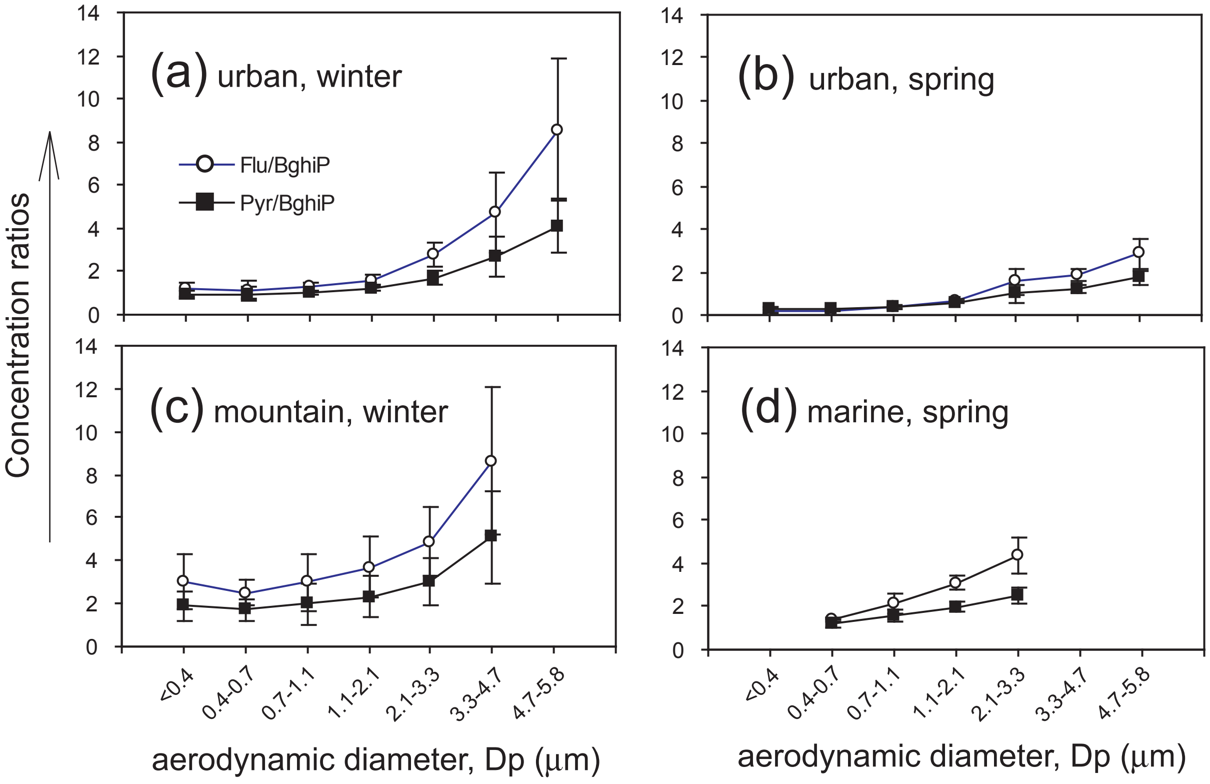
Table 4. Comparison of compositions of major PAHs with those measured in US and Europe

Location	Site description	Particle size	Season	BbkF/BeP	IP/BeP	BghiP/BeP	References
I. East Asia							
Baoji, China	Urban	TSP	Winter	3.8 ± 0.4	1.3 ± 0.1	1.2 ± 0.0	This study
		TSP	Spring	4.0 ± 0.1	1.7 ± 0.2	1.5 ± 0.1	
Mt. Tai, China	Mountain	TSP	Winter	4.5 ± 0.2	0.7 ± 0.2	0.8 ± 0.2	This study
		PM_9	Summer	1.6	1.1	1.0	
Okinawa Is., Japan	Marine	TSP	Spring	7.8 ± 0.6	1.3 ± 0.1	0.9 ± 0.1	This study
II. US and Europe							
Sacramento, US	Urban	$PM_{1.8}$	Winter	2.3	0.4	5.6	Kleeman et al., 2008
Los Angeles, US	Urban	TSP	Annual	1.0	1.3	3.5	Finlayson-Pitts and Pitts Jr., 2000
London, UK	Urban	TSP	Autumn	0.3	0.9	1.7	Finlayson-Pitts and Pitts Jr., 2000

Figure Caption

Figure 7. Relative concentration of fluoranthene (Flu) and pyrene (Pyr) to that of benzo(ghi)perylene (BghiP) in each stage at the urban, mountain and marine sites during the same season: (a) and (c) for winter and (b) and (d) for spring (Data points showed here only for the size ranges where BghiP can be detected. Error bar represents the standard deviation)

Figure 7



Concentration ratios
(a) urban, winter
Flu/BghiP
Pyr/BghiP
(b) urban, spring
(c) mountain, winter
(d) marine, spring
0
2
4
6
8
10
12
14
<0.4
0.4-0.7
0.7-1.1
1.1-2.1
2.1-3.3
3.3-4.7
4.7-5.8
aerodynamic diameter, Dp (μm)