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Interactive Comment

## Interactive comment on "Source apportionment of the particulate PAHs at Seoul, Korea: impact of long range transport to a megacity" by J. Y. Lee and Y. P. Kim

### J. Y. Lee and Y. P. Kim

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Thanks for the review, we have accommodated most comments. We will try to present more clearly on the limitations on the modeling method and data we have used in the revised manuscript. Responses on the general comments are:

1. Validity of the modeling approach by using non-conserving low molecular compounds: We agree with the reviewer that it would be ideal if reactive species, i.e., low molecular PAH compounds be not included in the CMB modeling. However, if we do not include those low molecular compounds in the source profiles, the model performance might not be satisfactory such as the DF value far out of the suggested acceptable range. Thus, it is not feasible to run the model excluding the low molecular weight

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compounds in this study. We tried to minimize the effect of non-conserving compounds by normalizing the individual PAH compound concentration with benzo(e)pyrene. We will discuss this problem in detail in the revised manuscript.

2. The reason the source profile of Li et al. (2003) was used instead of Chen et al. (2004) for coal residential source: In addition to coal residential source profile, we have tried several source profiles, if available, for each source. It is desirable that accurate source profile obtained from that source area (for example, coal combustion source profiles obtained from a specific coal mine area in China) be used. However, the coal composition in China, for example, might vary significantly depending on the source area in China. Further, the reliability of the profiles from source regions might be in question. Thus, the primary selection criterion for the source profile in this study is the model performance. In other words, the source profile which gives the best model performance is selected. When using the profile of Chen et al., the major findings are the same with higher coal contributions (coal residential and coal power plant) and lower unidentified but with the worse statistical model performance indices. For example, the chi-square value (chi-square values in winter for the source profiles of Li et al. and Chen et al. are  $0.68 \pm 0.67$  and  $3.38 \pm 1.29$ , respectively) and the percent mass value (on the average the deviation is 10 for the Li et al.'s profile and 20 for the Chen et al.'s profile) show marked differences. Thus, in this study, the source profile of Li et al. (2003) is chosen. We will add a part on the selection criterion of the source profile and its consequences in the revised manuscript.

3. Some samples having DF value of 3: Out of 68 samples we have used in the modeling, three samples (Feb 27 and Dec. 18 and 23, 2003) have the DF value of 3 due the number of fitted sources being higher than other samples. The performance criterion for DF value set by USEPA was equal or higher than 5. However, as mentioned in the general comment 1, the number of PAH compounds applied in the modeling study were limited and, thus, we cannot satisfy the criterion for the three samples.

Also, responses on the specific comments are:

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1. Distinct markers for each source profile: The main reason for not observing nonlinearity problem during the CMB modeling, we believe, is the source profiles we have used are distinct. For example, the concentration of Phen is high in coal residential source profile, BghiP and Pyr for gasoline and diesel vehicles, and Chry for natural gas. Also, low molecular compounds are higher for coal power plant, middle molecular compounds for coal coke oven, and Flt and Pyr for biomass burning. We will add this part in the revised manuscript.

2. The internal standard being spiked after extraction: Internal standard (phenanthrene-D10) was used as the standard for the GC-MS analysis. That was the reason it was spiked after extraction. Extraction recovery was estimated by putting the standard solution to the blank filter. Detailed procedure was given in Lee et al. (2006), "Extraction recoveries were determined by spiking the PAHs standard solution in the pre-extracted QFFs (real filter samples in which PAHs were extracted). The same pre-treatment and analytical procedures had been repeated for the measurement of extraction recoveries. The reason of using the pre-extracted filter was to obtain extraction recovery." We will make this sampling and analysis part more clearly in the revised manuscript.

3. Is there any way to quantify the transported materials? Is there any transport from Japan? With our experimental setting, it is hard to directly identify the transport materials. We might use the ratio values of the concentrations of individual PAH compound and compared reported source specific (or emission specific) ratios. However, in our opinion, this approach is not that definitive. On the transport from Japan, during summer, some air parcel trajectories are from southeast, i.e., Japan. Some study results in southern part of Korea also indicated that emissions from Japanese volcanoes were observed during summertime. However, in Seoul which is in the middle part of Korean peninsula, it is not likely to observe the transport from Japan.

4. Pre-bake temperature of quartz filters: We followed the US EPA-TO13 method and,

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thus, we baked the filters in  $400^{\circ}C$ . Usually, the concentration levels of the field and lab blanks were below the method detection limits except Phen, BaA, BaP, and BghiP for those compounds, we accounted for the blank concentrations in the results.

5. Use of the light PAH compounds' data: It is closely related to the problem of 'the validity of the modeling approach by using non-conserving low molecular compounds' the reviewer raised earlier. Basically, these low molecular species are used to make the model performance satisfactory. Please note that even with the light PAHs, the model performance for summer was not as good as other seasons. We think that is mainly due to light PAHs as the reviewer pointed out. Another reason we did include the light PHAs is that though the concentrations of them in the gas phase were high, the concentration of them in the particulate phase were also high. If we exclude them in the data analysis procedure, we might miss some important emission sources in which light PAHs are important source identification compounds.

6. Typos in reference citing: We correct Zhang to Zheng and Li et al. (2005) to Li et al. (2003).

7. When using the source profile of Schauer et al. (2001): When using the Schauer et al.'s profile instead of Rogge et al. (1998) the model performance criteria values for both cases are in the acceptable range. Also, the results are comparable to each other. On the average, the biomass burning contribution was changed from 14% for Rogge et al.'s profile to 11% for Schauer et al.'s profile.

8. The value of 2 being higher: Sorry about it. When we upload the file, the Greek letter was removed during the process. As you pointed out, it is chi-square ( $\chi^2$ ).

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