

Interactive comment on “Characteristics and sources of carbonaceous aerosols from Shanghai, China” by J.-J. Cao et al.

J.-J. Cao et al.

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Anonymous Referee #2 Received and published: 31 August 2012 This study deals with the characterization and the analyses of carbonaceous components in atmospheric aerosols collected for 20 days in Shanghai, China. The collected and analyzed data are comprehensive and could serve as validation data in the future numerical model studies. Although the data analyses and discussion presented before section 3.4 are conventional ones, the source apportionment method presented in section 3.5 is an intriguing way. There is a possibility that this method can be applied to the data collected in other megacities.

Reply: We have replied the questions and revised the manuscript according to the comments of the reviewers.

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Specific comments 1. In the filter sampling of carbonaceous aerosols, especially in the short sampling time of 12 hr, the sampling artifact such as adsorption of organic gases onto the quartz-fiber filter media becomes relatively significant. It is necessary to evaluate the effect of such artifact on the measured OC conc., OC thermal profiles, and its effect on the final source apportionment presented in section 3.5.

Reply: There are positive and negative artifacts during our sampling, indeed. Because we used the mini-vol sampler (flow rate of 5 l min⁻¹) to collect the aerosol samples, the organic artifacts from sorption and evaporation should be minimal due to the low flow rate. Also it would be difficult to make a denuder system work effectively for a sampler of this type. For another reason, the total carbon would appear to be very high compared with any organic artifacts in the filters during our study, so we just corrected the data for blank filters to obtain estimates of contents of OC. It is also conventional by using blank filters to evaluate the OC sampling artifacts. The artifacts were less than 5% in our previous study (Cao et al., 2007), so the results of final source apportionment are acceptable. Cao J.J., et al., Spatial and seasonal distributions of carbonaceous aerosols over China, 112, D22S11, doi:10.1029/2006JD008205, 2007.

2. In the thermal analysis of EC, the contribution of carbonate (CaCO₃) sometimes occupies the certain fraction of EC₂ or EC₃. In this study, did the author remove the carbonate before the EC analysis? This problem becomes important in particular in Megacities in China where unintermittent construction works are going on anywhere. At least, the author can report Ca concentrations in the aerosol samples.

Reply: Yes, we removed the carbonate before the EC analysis. The most possible composition of the Ca-abundant particles is CaO or Ca(OH)₂, emitted from construction sites (Zhang et al., 1999), which didn't have impact on our carbon analysis. In addition, we also checked our Ca²⁺ data in these samples, the Ca²⁺ in most samples were zero and only six samples have concentration ranged from 0.04 - 9.7 mg m⁻³. Zhang D.Z., et al., Nitrate and sulfate in individual Asian dust-storm particles in Beijing China in spring of 1995 and 1996, Atmospheric Environment 33, 3213-3223, 1999.

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3. The estimated fractions of $\delta^{13}\text{C}_{\text{EC}_{\text{CC}}}$ and $\delta^{13}\text{C}_{\text{EC}_{\text{MV}}}$ to the measured $\delta^{13}\text{C}_{\text{EC}_{\text{AM}}}$ by equation 3 (i.e., X_{CC} and X_{MV} , respectively) may be sensitive to the assigned values of $\delta^{13}\text{C}_{\text{EC}_{\text{CC}}}$ and $\delta^{13}\text{C}_{\text{EC}_{\text{MV}}}$, because the assigned values are not largely different (-23.4 and -27‰). The author should provide the range of X_{CC} and X_{MV} when the assigned value of $\delta^{13}\text{C}_{\text{EC}_{\text{CC}}}$ and $\delta^{13}\text{C}_{\text{EC}_{\text{MV}}}$ varied within their reported error range. In the same manner, there should be some uncertainty range in the OC/EC ratios for CC and MV (2.7 and 1.1) adopted from a literature. The author should evaluate the effect of such uncertainty in the used parameters to the final result of source apportionment calculation.

Reply: The isotopic values from -23.4 and -27‰ were largely for coal combustion and motor vehicle sources. It is effective when using the isotopic mass balance model. We assigned the $-23.4 \pm 1.2\text{‰}$ for $\delta^{13}\text{C}_{\text{EC}_{\text{CC}}}$ and $-27 \pm 1.0\text{‰}$ for $\delta^{13}\text{C}_{\text{EC}_{\text{MV}}}$. The corresponding uncertainty were updated in the new text: The average percent contributions to EC calculated in this way were 53% (ranged from 52% to 57%) for CC and 47% (ranged from 42% to 48%) for MV. The reason for the use of OC/EC ratios (2.7 and 1.1) for CC and MV was due to lack of OC/EC ratios for source samples in Shanghai. In this study, we try to demonstrate a new method for source apportionment estimation by using the different carbon data. More accurate ratios from the study area will improve the precision of source contribution. Currently, it is hard to quantitatively evaluate the effect of such uncertainty in the used parameters to the final result of source apportionment calculation since there are no enough related data. We added a note about this point in the text: Since current OC/EC ratios were adapted from literature, more accurate ratios from Shanghai will improve the precision of source contribution.

4. Very low concentration of BaP compared to that of BeP (Table 3) seems strange to the reviewer even the decomposition rate of BaP is faster. Shanghai is one of the largest city in the world and accordingly the load of fossil-fuel originated carbonaceous aerosols may also be one of the heaviest. The author's interpretation, i. e., the potential impacts of non-local or aged aerosol, shall be suitable to the results obtained in the

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remote environment or at least in the suburbs, but not persuading when applied to the results in Shanghai unless further explanations are given. Can the author provide time-series graph of $\text{BeP}/(\text{BaP}+\text{BeP})$?

Reply: In this study, the sampling was conducted at Pudong, Shanghai which is closed to a park and far away from the major roads. Therefore, the impacts of fossil-fuel emissions from vehicles were not significant and low concentrations of BaP were observed. In previous study in Hong Kong (Zhang and Fang, 2000), the average concentration of BaP in six sampling sites was 0.5 ng m⁻³ which was close to the BaP concentration determined in our study. Feng et al (2006) found that the ratios of $\text{BeP}/(\text{BaP}+\text{BeP})$ in Shanghai were 0.73 in summer and 0.75 in winter which were similar to the result in our study (0.76). Moreover, Gu et al (2010) also found that the annual average of $\text{BeP}/(\text{BaP}+\text{BeP})$ ratio (2007 to 2008) in Shanghai was 0.63 which is close to our result. Moreover, the average wind speed in Pudong was around 1 m s⁻¹ during the sampling period, which was not favourable for the dispersion of air pollutants and increased the decomposition rate of BaP. As suggested by the reviewer, the time series of $\text{BeP}/(\text{BaP}+\text{BeP})$ was plotted and the ratios ranged from 0.66 to 0.85. In conclusion, the low concentration of BaP and the high ratio of $\text{BeP}/(\text{BaP}+\text{BeP})$ observed are common in Shanghai.

Fig. 1 Time series plot of $\text{BeP}/(\text{BaP}+\text{BeP})$

Zheng M. and Fang M.: Particle-associated Polycyclic Aromatic Hydrocarbons in the Atmosphere of Hong Kong, *Water, Air, and Soil Pollution* 117, 175-189, 2000.

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Fig. 1 Time series plot of BeP(Bal+BeP)

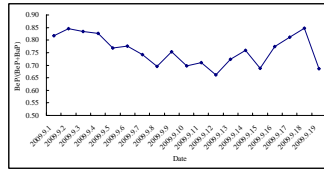


Fig. 1.

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