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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Impact of air pollution control measures and regional transport on carbonaceous aerosols in fine particulate matter in urban Beijing, China: Insights gained from long-term measurement" *by* Dongsheng Ji et al.

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General Comments This research is to investigate the OC and EC hourly concentration variation in Beijing, China from 2013 to 2018. Based on data, authors discussed the relationship between OC and EC, and with major air pollutants and sources via inter-annual, seasonal, weekly and diurnal variations. Finally NWR and PSCF were employed to evaluate the local and regional anthropogenic sources. In general, the

data of this manuscript are informative and this paper ïňĄts the scope of ACP. Please modify the manuscript based on the following comment before ACP publication.

Response: We thank the reviewer for the constructive comments and suggestions. According to the reviewer's suggestions, we have done our best to revise our manuscript.

Comments:

1.Section 3.1, page 9, Table 2 contains a lot of data, and the source of data should be acknowledged. I could not see the importance of Table 2 in the main text, and suggest that Table 2 can be shifted to supporting information.

Response: We thank the reviewer for the comment. As suggested, Table 2 has been moved to the supplementary material.

2. Page 10, line 237, it should be "Table 3", not Table 2. In Table 3, what are the meaning for TOT and TOT? Please explain both of them by note in the table. (Same with Table 4).

Response: Thanks for pointing this out. As the original Table 2 has been removed, the original Table 3 is Table 2 now. In addition, the notes for the abbreviations TOT, TOR and EA have been added as footnotes of Tables 2 and 3 in the revised manuscript.

3. Page 10, lines 254-255, what criteria did authors classify PM2.5 different air quality levels as excellent, good, slightly polluted, moderately polluted, heavily polluted and severely polluted? Please specify in the text.

Response: Thanks for the comment. The criteria used to classify the air quality have been added in the revised manuscript. Air quality as Excellent, good, lightly polluted (LP), moderately polluted (MP), heavily polluted (HP) and severely polluted (SP) are based on the daily average PM2.5 concentration, i.e., excellent (0<PM2.5$\leq$35 μg/m3), good (35<PM2.5$\leq$75 μg/m3), lightly polluted (LP, 75<PM2.5$\leq$115 μg/m3), moderately polluted (MP, 115<PM2.5$\leq$150 μg/m3), heavily polluted (HP, 150<PM2.5$\leq$250 μg/m3) and severely polluted (SP, PM2.5>250 μg/m3), respectively.

4. Page 10, line 254, in Fig. 2, “White Block” label is referring to PM2.5 in both two ïňĄgures?

Response: Thanks for the comment. It is true that the “white block” refers to the PM2.5 concentration in the top sub-figure in Fig. 2, while it means percentage of composition in PM2.5 excluding OC and EC in the bottom sub-figure in Fig. 2. The legends in Fig. 2 have been revised for better clarity in the revised manuscript.

5. Section 3.2, page 11, Fig. 4 can be part of Fig. 3. Does Fig. 4 contain special information? If yes, some more discussion related to Fig. 4 should be added. Otherwise I recommended to remove this ïňĄgure.

Response: As suggested, the original Fig. 4 has been removed in the revised manuscript as it presents partially repetitive information of Fig. 3.

6. Page 11, I found quite a lot of data in Fig. 3 including OC-EC during 2002-2012, SO2, NO2, ïňĄre spots, please specify the data sources of all these data.

Response: Thank you for pointing this out. As advised, the sources of the data, which were not included in this study, have been added in section 2 and in lines 304-305 (He et al., 2001; Zhao et al., 2013; Ji et al., 2016; Lang et al., 2017; Tao et al., 2017) in the revised manuscript.

References:

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Ji, D. S., Zhang, J. K., He, J., Wang, X. J., Pang, B., Liu, Z. R., Wang, L. L., and Wang, Y. S.: Characteristics of atmospheric organic and elemental carbon aerosols in urban Beijing, China, Atmos. Environ., 125, 293-306, 2016.

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2000-2015. Aerosol Air Qual. Res., 17, 412-425, 2017.

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Zhao, P., Dong, F., and Yang, Y.: Characteristics of carbonaceous aerosol in the region of Beijing, Tianjin, and Hebei, China, Atmos. Environ., 71, 389-398, 2013.

7. Page 12, lines 302-305, authors claims that biomass burning could contribution a lot to OC and EC, please be noted that ïňĄre spots in 2012 were highest, please add some discussion in this section.

Response: Thanks for this comment. More discussion has been added in line 327-344 in the revised version: “In Fig. 3, it can be seen that the annual average EC concentration and the fire spot counts exhibit a rather similar variation from 2004 to 2017, except in the year 2012, which suggests that the EC levels are somewhat correlated with the biomass burning; this might indicate that biomass burning contributed somewhat to the EC levels. The reduction in fire spot counts from 2014 to 2017, which resulted from efforts to control the agricultural field residue burning since 2013, helped to reduce the EC concentrations to some extent, but the low EC levels during 2014-2017 are likely mostly due to the implementation of the clean air act. With regard to the anomaly in the year 2012, based on the MODIS data for this year, a very non-uniform distribution of fire spots in the BTH region was observed, with a distinct decrease of fire spot counts in Beijing, but higher fire spot counts in the southern Hebei Province; this may be ascribed to the fact that the policy of Banning Straw Burning in Summer and Autumn was executed to different degrees in the whole region, with better implementation in the Beijing area and worse action in the other parts. (http://www.beijing.gov.cn/zfxxgk/110029/qtwj22/2012-12/11/content_357114.shtml). In addition, for the years from 2002 to 2017, the highest

precipitation volume in Beijing was recorded in 2012, i.e., 733.2 mm, and the rainy days mainly occurred in the intensive straw burning periods, accounting for 76.4% of all rainy days in 2012. The frequent wet scavenging might have suppressed the EC concentrations during the intensive straw burning periods, so that the annual EC level for 2012 was comparable to those recorded from 2011 onward."

8. Section 3.3, pages 12-14, authors separately discussed monthly and seasonal variations. Actually, I found quite a lot of data explanations are similar for both monthly and seasonal variation. Is it possible for authors to combine both to simplify the discussion?

Response: Thank you for pointing this out. We have simplified the discussion in lines 355-411 in the revised manuscript and the revised text is as follows: "Fig. S1 shows the monthly mean OC and EC concentrations at our study site for the whole 5-year period. Similar variations are observed with generally higher mean OC and EC levels in the cold season (from November to March next year when the centralized urban residential heating is provided) and lower ones in the warm season (from April to October). The highest average OC and EC concentrations were 24.1 $\pm$ 18.7 μg/m3 in December 2016 and 9.3 $\pm$ 8.5 μg/m3 in December 2015, respectively. However, the lowest OC and EC levels were not observed in the warm months; they were 5.0 $\pm$ 4.6 μg/m3 in January, 2018 and 1.5 $\pm$ 1.7 μg/m3 in December, 2017, respectively; this was associated with both frequent occurrence of cold air mass and the implementation of a winter radical pollution control action plan (Chen and Chen, 2019) in Beijing from November, 2017. Overall, the increased fuel consumption for domestic heating in addition to unfavorable meteorological conditions (lower mixing layer height, temperature inversion and calm wind) in the colder months is considered to lead to higher OC and EC levels (Ji et al., 2014). In addition, the lower air temperature in the cold months led to shifting the gas-particle equilibrium of semi-volatile organic compounds (SVOCs) into the particle phase, leading to higher OC levels. In the cold months, the cold start of vehicles (5.64 million vehicles in Beijing at the end of 2017) also increased the emission of OC. In the warm season, lower OC and EC levels were observed, which could be attributed

to the following factors: no extra energy consumed for domestic heating, strong wet scavenging by frequent precipitation occurring in these months, and more unstable atmospheric conditions favorable for pollutant dispersion; in addition, during this period, the monthly mean OC and EC concentrations generally decreased from year to year. In contrast, for the cold season, the monthly mean OC and EC concentrations did not show a clear decreasing trend from year to year except for October. In addition to the more intensive energy consumption in the cold season, the EC and OC levels could be also be enhanced strongly by regional transport and stagnant meteorology leading to ground surface accumulation in the autumn and winter (Wang et al., 2019; Yi et al., 2019), which would counteract the efficacy of the energy structure change in the Beijing-Tianjin-Hebei region of the past few years. It is worth pointing out that, on a year to year basis, the monthly average OC and EC concentrations in the cold seasons of 2017 and 2018 were generally lower than those in 2016, demonstrating to some extent the effectiveness of the execution of the radical pollution control measures for cities on the air pollution in the Beijing-Tianjin-Hebei region. The interquartile ranges of OC and EC in the warm months were narrower than in the cold months, indicating that there was more substantial variation in concentration in the latter months. The larger variation in the colder months could be caused by the cyclic accumulation and scavenging processes. In this region, due to the cyclic accumulation and scavenging process, the concentration of particulate matter increases rapidly when the air mass back trajectories change from the northwest and north to the southwest and south over successive days in Beijing; in contrast, the concentration of particulate matter declines sharply when a cold front causes the shift of back trajectories from the southwest and south to the north and northwest (Ji et al., 2012). The successive accumulation processes are closely associated with unfavorable meteorological conditions, which gives rise to higher OC and EC concentrations, while more scavenging of aerosols by cold fronts leads to lower levels. As to the seasonality in OC and EC, similar seasonal variations are observed in the various years with generally higher mean concentrations in autumn and winter and lower levels in spring and summer (Fig. 4). Remarkably, the OC and

EC concentrations in the autumn and winter of 2017 were lower than those in the previous years. This was due to the combined effect of controlling anthropogenic emissions strictly and favorable meteorological conditions. Since September 2017, a series of the most stringent measures within the Action Plan on Prevention and Control of Air Pollution was implemented to improve the air quality; these measures included restricting industrial production by shutting down thousands of polluting plants, suspending the work of iron and steel plants in 28 major cities and limiting the use of vehicles and reducing coal consumption as a heating source in northern China. In addition, the air quality improvement in the autumn and winter of 2017 was closely tied to frequent cold fronts accompanied by strong winds, which was favorable for dispersing the pollutants. The average OC and EC concentrations in the winter were 1.69 and 1.14, 2.17 and 1.93, 1.49 and 2.14, 2.41 and 2.29 and 0.80 and 0.88 times higher than those in the summer for 2013, 2014, 2015, 2016 and 2017, respectively. The difference in the ratios for 2017 was due to the series of the most stringent measures taking effect and favorable meteorology. The Beijing municipal government in particular has made great efforts to replace coal by natural gases and electricity-powered facilities. Besides, new energy vehicles are increasingly used to replace the gasoline vehicles."

Reference: Chen, H. and Chen, W.: Potential impact of shifting coal to gas and electricity for building sectors in 28 major northern cities of China, Appl. Energ., 236, 1049-1061, 2019.

9. Section 3.4, page 14, lines 365-367, EC concentrations increased starting from 17:00 because of evening rush hours, I am curious why morning rush hour did not result in the increase of EC?

Response: Thank you for this comment. As indicated in the manuscript, whereby 'other times' stands for 'non-night times', "At other times, both the higher PBL height and lower traffic intensity resulted in lower EC concentrations". It has been reported that morning peaks of OC and EC levels were higher than those during the nighttime in urban areas in the US (Rattigan et al., 2010, Kang et al., 2010), where there was

no strong traffic emission of OC and EC during the nighttime. However, as regulated by the Beijing Traffic management Bureau (http://www.bjjtgl.gov.cn/zhuanti/10weihao/), HDV and HDDT are allowed to enter the urban area inside the 5th Ring Road from 0:00 to 06:00 (local Time) in Beijing. In addition to the nocturnal PBL effect, these high emitters contribute significantly to the high levels of OC and EC from midnight to the early morning; the OC and EC originating from the morning rush hour is not sufficiently dominant to form peaks during the 6:00-8:00 period.

References: Kang, C. M., Koutrakis P., and Suh, H. H.: Hourly measurements of fine particulate sulfate and carbon aerosols at the Harvard-U.S. Environmental Protection Agency Supersite in Boston, J. Air Waste Manage., 60:11, 1327-1334, 2010.

Rattigan, O. V., Felton, H. D., Bae, M. S., Schwab, J. J., and Demerjian, K. L.: Multi-year hourly PM2.5 carbon measurements in New York: Diurnal, day of week and seasonal patterns, Atmos. Environ., 44(16), 2043-2053, 2010.

10. Section 3.5, similar to the above comment No. 6, please specify the data source of gaseous pollutants.

Response: Thank you for pointing this out. We have added the following text for the data sources of the gaseous pollutants in section 2.2 in the revised manuscript: "The analyzers/monitors for O3, CO, SO2, NOx and PM2.5, and their precision, detection limits and calibration methods have been described in detail elsewhere (Ji et al., 2014). Briefly, O3 was measured using an ultraviolet photometric analyzer (model 49i, Thermo Fisher Scientific (Thermo), USA), CO with a gas filter correlation nondispersive infrared method analyzer (model 48i, Thermo, USA), SO2 using a pulsed-fluorescence analyzer (model 43i, Thermo, USA), NO-NO2-NOx with a chemiluminescence analyzer (model 42, Thermo, USA) and PM2.5 using a US Environmental Protection Agency Federal Equivalent Method analyzer of PM2.5 (SHARP 5030, Thermo, USA)."

11. Page 18, 2nd paragraph, authors discussed the relationship between ozone and OC. It is interesting to ïňĄnd that O3 at 50ug/m3 represented the highest OC, and OC

increased with ozone for O3 concentration above 100ug/m3. More discussion of the potential reasons will deïňĄnitely enhance the quality of manuscript.

Response: Thank you for pointing this out. We have added the following relevant discussion and explanation on the relationship between ozone and OC in lines 536-562 in the revised manuscript: “Emissions of primary air pollutants lead through multiple pathways to the formation of ozone and secondary organic carbon (SOC) (Seinfeld and Pandis, 1998), both of which are the principal components of photochemical smog. The relationship between OC and O3 is of use for understanding their variation and formation. The OC concentrations were highest for an O3 concentration of 50 μg/m3, which is approximately the average O3 concentration in Beijing in winter (Cheng et al., 2018). During the period of an O3 concentration of 50 μg/m3, low atmospheric temperature (9.4$\pm$9.9 °C), relatively high RH (59.2$\pm$23.7 %), lower WS (1.1$\pm$0.8 m/s) and higher NOx concentrations (72.7$\pm$57.5 ppb) were observed and a lower mixed layer height was recorded in winter (Tang et al., 2016), which were favorable for accumulation and formation of OC. A relatively lower temperature is beneficial for condensation/absorption of SVOCs into existing particles (Ji et al., 2019), which would then experience further chemical reactions to generate secondary organic aerosol (SOA). Note that a low temperature does not significantly reduce SOA formation rates (Huang et al., 2014) in the winter. In addition, processes including aqueous-phase oxidation and NO3-radical-initiated nocturnal chemistry may contribute to or even dominate SOA formation during winter (Hallquist et al., 2009; Rollins et al., 2012; Huang et al., 2014). Hence, the above factors gave rise to the higher OC concentration at an O3 concentration of 50 μg/m3 particularly in winter. In addition, scattering and absorbing effects of aerosols that were trapped in the lower mixed layer height led to less solar radiation reaching the ground and further restrained the O3 formation in the cold season (Xing et al., 2017; Wang et al., 2016b). OC declined when O3 concentrations increased from 50 to 100 μg/m3. Usually moderate O3 concentrations accompanying lower OC concentrations are caused by increasing T (19.5$\pm$8.3 °C), increasing WS (2.0$\pm$1.3 m/s) and less titration of relatively lower observed NO concentrations (6.4$\pm$14.6 ppb). It can also

be seen that there was a concurrent increasing trend of OC and ozone when the O3 concentration was above 100 μg/m3, which generally occurred in the warmer season. Besides the impact of meteorological conditions, such a trend might not be dominated by gas-to-particle partitioning of low-volatility organic compounds but by the oxidation of VOCs driven by hydroxyl radicals to generate both SOC and O3 with relatively long lifetimes (>12 h; Wood et al., 2010)”

References:

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Wood, E. C., Canagaratna, M. R., Herndon, S. C., Onasch, T. B., Kolb, C. E., Worsnop, D. R., Kroll, J. H., Knighton, W. B., Seila, R., Zavala, M., Molina, L. T., DeCarlo, P. F., Jimenez, J. L., Weinheimer, A. J., Knapp, D. J., Jobson, B. T., Stutz, J., Kuster, W. C., Williams, E. J.: Investigation of the correlation between odd oxygen and secondary organic aerosol in Mexico City and Houston. Atmos. Chem. Phys. 18(10), 8947-8968, 2010.

12. Section 3.6, page 19, line 493, no Fig. 14 and 15 are in the whole manuscript.

Response: Our apologies for this error. It has been corrected and all the joint probability data are presented in the right panels of Figs. 7 and 8 in the revised manuscript.

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