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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Chemical characterization and source apportionment of $PM_2.5 in Beijing: seasonal perspective" by R. Zhang et al.

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Response to Reviewer 1

The study analyzed data of PM2.5 and its chemical components collected in Beijing using various methods. The data covered one month time in each season and the study focused on discussing seasonality of PM2.5 and major components. The paper is generally well organized and only need some minor revisions, as listed below.

R: We sincerely appreciate for the positive comments and suggestions. Also we respond the following queries point-by-point carefully; please refer to them.

P9964, L26 and P9965, L22: The resolution for PSCF analysis is 0.5 degree by 0.5 degree, but the back trajectory resolution is 1 degree by 1 degree. Shouldn't the grid size for back trajectory be much smaller than the PSCF grid in order to obtain a reasonable accuracy in PSCF analysis?

R: The trajectory model calculation yields the hourly endpoint, which is in turn used and can therefore sufficiently satisfy the required endpoints in each cell of the 0.5 degree by 0.5 degree grid (and even finer resolution) of the PSCF approach. Such application is often adopted, even with finer resolution (0.3 degree by 0.3 degree) (e.g., Hafner and Hites, 2003; Hwang and Hopke, 2007; Han et al., 2008; Heo et al., 2009).

Cited references:

Hafner, W.D., and Hites, R.A. (2003). Potential sources of pesticides, PCBs, and PAHs to the atmosphere of the Great Lakes. Environ. Sci. Technol. 37, 3764-3773.

Hwang, I., and Hopke, P.K. (2007). Estimation of source apportionment and potential source locations of PM2.5 at a west coastal IMPROVE site. Atmospheric Environment 41, 506–518.

Han, Y.J., Kim, T.S., and Kim, H. (2008). Ionic constituents and source analysis of PM2.5 in three Korean cities. Atmospheric Environment 42, 4735–4746

Heo, J.-B., Hopke, P.K., and Yi, S.-M. (2009). Source apportionment of PM2.5 in Seoul, Korea. Atmos. Chem. Phys., 9, 4957–4971.

P9967, L9-12: If you have precipitation data, you should compare precipitation amount in this particular summer month with previous seasonal average data. You will then know if it is the less than usual precipitation that preventing the summer minimum. Then you can qualitatively discuss the impact of photochemistry, which also needs temperature data to support your hypothesis (e.g., if it was higher T than typical season mean).

R: As suggested, we show the climatology of temperature, precipitation, relative hu-

midity and wind speed in Beijing in the newly added Figure S2; also we compare the temperature and precipitation of 2009 (the sampling year) with their respective climatology. The precipitation in July 2009 is slightly higher than that of the climatology, while the temperature in the summer of 2009 is obviously higher than those of the climatology. Also we have added two relevant statements, as follows:

This suggestion can be supported by the fact that temperatures through February to October 2009 in Beijing are higher than the climatology by $\sim$1 °C or higher (Figure S2), while precipitation in July 2009 (i.e., 197 mm) is even slightly larger than the climatology (185 mm), demonstrating that the photochemical effect might overwhelm the precipitation scavenging effect for the fine aerosol pollutants.

One might further conclude that in Beijing, photochemistry plays a more vital role in the sulfate aerosol formation and variability than the change in precursor SO2 emission as well as rain scavenging process.

P9967, L21-24, and P9968 L13-14: you attributed the lower winter time sulfate (compared to 2003 data) to effective emission control, but such as a decrease in sulfate was not found in other seasons. You also mentioned the increased vehicular emission may offset the control measure for NOx. I think the discussion in this paragraph should be improved. Firstly, you only had one month data which might not represent the season average. Secondly, the meteorology data for this month might be significantly different from the seasonal average of those years you compared. In order to identify the exact causes of the observed differences between different years and between sulfate and nitrate, you need to discuss meteorological data, and if possible to provide the emission data to support you conclusions.

R: The decrease in SO2 emission in China is evident in the recent years (since 2007 to 2010) (Itahashi et al., 2010). However, temperature in summer 2009 is hotter than the climatology, which is thus favorable for the sulfate aerosol formation. Accordingly, the relative lower sulfate concentration could be seen in winter, but not in summer.

Therefore we have added a conclusion as given in the former response.

Regarding the NOx, we have revised to “The overall trend of NOx emission in China is increasing primarily due to persistently increasing energy demand for industrial development and transportation, though control measures for NOx emissions have been implemented in power plants (Zhao et al., 2013).”, as suggested by the reviewer. For the SO2 and NOx emissions in China, they can be found in the newly cited three references, as follows:

Itahashi, S., Uno, I., Yumimoto, K., Irie, H., Osada, K., Ogata, K., Fukushima, H., Wang, Z. and Ohara, T.: Interannual variation in the fine-mode MODIS aerosol optical depth and its relationship to the changes in sulfur dioxide emissions in China between 2000 and 2010, Atmos. Chem. Phys., 12, 2631–2640, 2012.

Zhang, Q., Streets, D. G., Carmichael, G. R., He, K. B., Huo, H., Kannari, A., Klimont, Z., Park, I. S., Reddy, S., Fu, J. S., Chen, D., Duan, L., Lei, Y., Wang, L. T., and Yao, Z. L.: Asian emissions in 2006 for the NASA INTEX-B mission, Atmos. Chem. Phys., 9, 5131–5153, 2009.

Zhao, B., Wang, S. X., Xu, J. Y., Fu, K., Klimont, Z., Hao, J. M., He, K. B., Cofala, J., and Amann, M.: NOx emissions in China: historical trends and future perspectives, Atmos. Chem. Phys. Discuss., 13, 16047–16112, 2013.

P9968, L25-26: Do you have an explanation why the seasonality in your study was different from earlier studies?

R: Such seasonality is closely related to harvesting in the autumn and cultivation in the spring, which has been described by Duan et al. (2004). Accordingly, we revised to “Such seasonality was also found by some previous studies (He et al., 2001; Zheng et al., 2005), but distinct from other previous studies, in which winter often had the highest concentration (Duan et al., 2006; Wang et al., 2005). Specifically, the highest levoglucosan, which is suggested to serve as an excellent tracer of biomass burning pollutants

relatively to K+, has been measured in autumn (He et al., 2006), although no spring sample was measured in that study." along with the addition of the aforementioned reference. The newly cited two references are as follows:

Duan, F. K., Liu, X., Yu, T., and Cachier H.: Identification and estimate of biomass burning contribution to the urban aerosol organic carbon concentrations in Beijing, Atmos. Environ., 38, 1275–1282, 2004.

He, L. Y., Hu, M., Huang, X. F., Zhang, Y. H., Tang, X. Y.: Seasonal pollution characteristics of organic compounds in atmospheric fine particles in Beijing, Sci. Total Environ., 359, 167– 176, 2006.

P9979, discussion on Fig 11: Are the patterns shown in Figure 11 consistent with the current emission inventory for every species? The results/discussions have potential to improve the existing emission inventory for places with discrepancies. The trajectory resolution seems too rough though for this purpose. Also, you chose average concentrations as the "polluted" standard (P9965, L16). If you choose higher level, will the results significantly change your conclusion?

R: As guessed by the reviewer, the spatial patterns of the PSCF result for each species (sulfate, nitrate, ammonium, OC, EC, and dust) are relatively consistent with those of their respective corresponding species' emissions such as SO2, NOx, NH3, OC and EC except dust. The reasoning for the discrepancy for dust is because Talimakan Desert is a very important dust source in China while for the dust transported to Beijing is primarily originated from northern/northwestern regions of China, rather than Talimakan Desert. We have added related statements in the revised manuscript. The newly cited references are as follows.

Hsu et al. (2003) have compared the results using the mean and the 75th percentile concentrations as a threshold and demonstrated that overall, the two thresholds can achieved consistent results though the 75th percentile concentration would underestimate the moderate sources but has better performance for strong sources. They there-

fore suggested using mean concentration as the criterion in performing the PSCF approach, which has often been adopted in the community. Besides, we also performed a test experiment by increasing the threshold to 1.5 times of the mean concentration; the results didn't change too much.

Fu, T.-M., Cao, J. J., Zhang, X. Y., Lee, S. C., Zhang, Q., Han, Y. M., Qu, W. J., Han, Z., Zhang, R., Wang, Y. X., Chen, D. and Henze, D. K.: Carbonaceous aerosols in China: top-down constraints on primary sources and estimation of secondary contribution, Atmos. Chem. Phys., 12, 2725–2746, 2012.

Huang, X., Song, Y., Li, M., Li, J., Huo, Q., Cai, X., Zhu, T., Hu, M., and Zhang, H.: A high-resolution ammonia emission inventory in China, Global Biogeochem. Cycles, 26, GB1030, doi:10.1029/2011GB004161, 2012.

Laurent, B., Marticorena, B., Bergametti, G., Chazette, P., Maignan, F., and Schmechtig, C.: Simulation of the mineral dust emission frequencies from desert areas of China and Mongolia using an aerodynamic roughness length map derived from the POLDER//ADEOS 1 surface products, J. Geophys. Res., 110, D18S04, doi:10.1029/2004JD005013, 2005.

Editorial comments:

P9957, L28: Change "to improve slowly" to "to improve little, or even become worse".

R: Changed to "to improve little".

P9974: you should move the sentence on lines 8-10 to the beginning of line 6 (first explain Table 2 then discuss data in Table 2).

R: Moved.

P9974, L14-15: change "higher" to "highest" and "lower" to "lowest".

R: Amended.

P9975: L10-15: rewrite this sentence.

R: As suggested, we have rewritten this sentence as “Coal combustion is the predominant source of fine aerosols over China (Yao et al., 2009), which has resulted in severe air pollution problem not only locally, but also regionally and globally; for instance, it alone contributes more than 10% (268 Mg) of the global anthropogenic mercury emission (2319 Mt) annually (Pirrone et al., 2010). Besides, coal fly ash could be one of the main contributors of aerosol Pb in China as they contain abundant Pb (Zhang et al., 2009b).”; please refer to the revised manuscript

P9977, L23: delete the second “based”.

R: Deleted.

Response to Reviewer 2

General comments:

This paper shows the detailed chemical composition and seasonality of particulate matter (PM2.5) in Beijing of China based on sampling. The numerous approaches are used for characterizing aerosol possible sources. The result of his paper is useful for assessing the regional aerosol environment effect of PM in the mega of China. Overall, this is a good work.

R: Thanks for the positive comments and recommendation for publication.

Specific comments:

1. In figure 6, the correlation coefficient mark ‘r’ should be change for ‘R’;

R: Changed, as suggested.

2. In page 9960, line 18, the pore size of Teflon filter is 2 μm, please the authors assure it. If 2 μm pore used, it may influence the sampling efficient of PM2.5, especially small particles in size < 2μm.

R: In general, our sampling protocol is consistent with that of the US EPA speciated particle sampling guideline, written by Chow and Watson (1998). The PTFE filter of power size 2 μm is recommended in the standard sampling method of PM2.5 and is therefore used widely by the community long time (e.g., Chow et al., 1993; Chow and Watson, 1998; He et al., 2001; Gibson et al., 2013), which does not influence the sampling efficiency of PM2.5.

Cited references:

Chow, J. C., Watson, J. G., Lowenthal, D. H., Solomon, P A., Magliano, K. L., Ziman S D., & Willard Richards L. (1993). PM10 and PM2.5 compositions in California’s San Joaquin Valley. Aerosol Sciencc and Technology 18, 105-128.

Chow JC, Watson JG (1998). Guideline on Speciated Particulate Monitoring, 291 page (http://epa.gov/ttnamti1/files/ambient/pm25/spec/drispec.pdf)

Gibson M.D. (2013). Identifying the sources driving observed PM2.5 variability over Halifax, Nova Scotia, during BORTAS-B. Atmos. Chem. Phys. Discuss., 13, 4491–4533.

He, K. B., Yang, F. M., Ma, Y. L., Zhang, Q., Yao, X., Chan, C. K., Cadle, S., Chan, T. and Mulawa P. (2001). The Characteristics of PM2.5 in Beijing, China, Atmos. Environ., 35, 4959-4970.

Please also note the supplement to this comment:
http://www.atmos-chem-phys-discuss.net/13/C3917/2013/acpd-13-C3917-2013-supplement.pdf

Interactive comment on Atmos. Chem. Phys. Discuss., 13, 9953, 2013.