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Discussions

Interactive comment on "PM$_2$.5 pollution in a megacity of southwest China: source apportionment and implication" *by* J. Tao et al.

J. Tao et al.

schsu815@rcec.sinica.edu.tw

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In this manuscript, Tao et al. presented a study of PM2.5 at an urban site in Chengdu megacity in four seasons in 2011. PM2.5 and its chemical components were measured during the campaign. In addition, the likely chemical formations of ionic constituents have been studied by applying the ISORROPIA-II thermodynamic equilibrium model; and the contributions of several likely sources of PM2.5 have been identified by a widely used receptor model – PMF. Also, the performances of the two models have been evaluated compared with the measurement data. This manuscript addresses the scientific questions that are within the scope of ACP and it is certainly well suitable for this special issue – "Atmospheric impacts of Eastern Asia". These measurement

data in this study are complete, providing a valuable dataset for the study of PM2.5 in Chengdu megacity. Both of the measurement and modeling results are significant for PM2.5 control strategies in Chengdu megacity. However, my major concerns are some conclusions derived from those ratios, and the credibility of the PMF results since the number of samples used in each season is very limited (only around or less than 30).

Response: We sincerely thank for the comments and suggestions, which help us to improve the paper. Regarding the major concern, the required minimum sample numbers in applying the PMF model is 100, as emphasized in the manual of the PMF model (Norris et al., 2008). Our total samples are 117, most of which have high quality data significantly above detection limits for most chemical species. Seasonal results of source apportionment were mostly based on annual results, with additional analysis of seasonal characteristics. Moreover, the PMF results have been evaluated by comparing with the observed data particularly for several tracers of specific sources. Thus, seasonal results of source apportionment should mostly reflect the real situation. Also compared to many studies, the sample number is adequate theoretically. More importantly, as the referee stated, the PMF results have been evaluated by comparing with the observed data particularly for several tracers of specific sources. Besides, the Fpeak and Q values (Fig. S2) have been reported to support the performance of the model results, as suggested by another referee. As for other points, we have carefully responded point-by-point as follows.

Norris, G., Vedantham, R., Wade, K., Brown, S., Prouty, J., and Foley, C.: EPA positive matrix factorization (PMF) 3.0 fundamentals user guide, US Environmental Protection Agency, Office of Research and Development, Washington, DC, 2008.

My suggestions are as follows:

Specific comments:

1) P5160, L1-6: "High LG/OC ratio (0.029) and low LG/MN ratio (7.8) in winter ... By contrast, high LG/OC (0.034) and LG/MN (17) ratios ..." I had difficulty to understand

these. Please explain how you define the so-called "high" and "low". Are there any criteria? If so, please show them in the text. In addition, are those ratios widely applicable? Please clarify;

Response: Sorry for this confusion. The wording "high" or "low" is based on the comparison among the four seasons. In the revised manuscript, we have clarified the statements. Besides, when compared to those ratios of previous studies, we usually specified the values clearly, so that the readers (and the referees) would not be confused. In the revised manuscript, we revised to "Relatively high LG/OC ratio (0.029) and low LG/MN ratio (7.8) in winter compared to those in spring and summer may indicate dominance of wood burning (Szidat et al., 2009; Oliveira et al., 2007; Pio et al., 2008). By contrast, the highest LG/OC (0.034) and LG/MN (17) ratios in autumn over the sampling year could reveal combined sources of wood burning together with rice straw and other agricultural waste burning (Sheesley et al., 2003; Engling et al., 2009)." The LG/MN ratio has been recently proposed to identify particular wood types in ambient biomass burning aerosols (Engling et al., 2009; Schmidl et al., 2008). In general, low LG/MN ratios (typically $3 - 7$) are generated by softwood combustion. In contrast, burning of hardwoods and crop residues typically is associated with higher LG/MN ratios with a rather broad range of $10 - 83$. On the other hand, the LG/OC ratio has been widely applied for estimating the relative contribution of biomass burning to PM2.5 or OC (Harrison et al., 2012; Holden et al., 2011; Wang et al., 2007; Zhang et al., 2008; Zhang et al., 2010). These ratios have been widely used in some studies as listed above.

Engling, G., Lee, J. J., Tsai, Y.-W., Lung, S.-C. C., Chou, C. C.-K., and Chan, C.-Y.: Size-resolved anhydrosugar composition in smoke aerosol from controlled field burning of rice straw, Aerosol Science and Technology, 43, 662-672, 2009.

Harrison, R., Beddows, D., Hu, L., and Yin, J.: Comparison of methods for evaluation of wood smoke and estimation of UK ambient concentrations, Atmospheric Chemistry and Physics, 12, 8271-8283, 2012.

Holden, A. S., Sullivan, A. P., Munchak, L. A., Kreidenweis, S. M., Schichtel, B. A., Malm, W. C., and Collett Jr, J. L.: Determining contributions of biomass burning and other sources to fine particle contemporary carbon in the western United States, Atmospheric Environment, 45, 1986-1993, 2011.

Schmidl, C., Marr, I. L., Caseiro, A., Kotianová, P., Berner, A., Bauer, H., Kasper-Giebl, A., and Puxbaum, H.: Chemical characterisation of fine particle emissions from wood stove combustion of common woods growing in mid-European Alpine regions, Atmospheric Environment, 42, 126-141, 2008.

Wang, Q., Shao, M., Liu, Y., William, K., Paul, G., Li, X., Liu, Y., and Lu, S.: Impact of biomass burning on urban air quality estimated by organic tracers: Guangzhou and Beijing as cases, Atmospheric Environment, 41, 8380-8390, 2007.

Zhang, Z., Engling, G., Lin, C.-Y., Chou, C. C.-K., Lung, S.-C. C., Chang, S.-Y., Fan, S., Chan, C.-Y., and Zhang, Y.-H.: Chemical speciation, transport and contribution of biomass burning smoke to ambient aerosol in Guangzhou, a mega city of China, Atmospheric Environment, 44, 3187-3195, 2010.

2) P5160, L14-20: it is difficult to understand how you estimated the contributions of wood burning just from those ratios, please explain it clearly;

Response: We have clarified and changed the statement to "Using the mean concentration of LG (Table 2) divided by this characteristic ratio (none was available in China), we estimated an annual contribution of 3.1 μg/m3 (varying from 1.1 μg/m3 to 4.9 μg/m3 in the four seasons) from wood burning, which represents 18

3) Figure 7 is interesting. The six sources are derived from the PMF modeling results based on the analysis of certain tracers, so, apparently, those tracers should have good correlations with related sources, if not, the tracer would not be surrogated into the factor. However, the authors are using the good correlations to evaluate the model performance. It is not convincing.

Response: Along with the correlations with specific tracers, the performance of PMF modeled results have indeed been evaluated by the Fpeak and Q values though not shown in the original manuscript. In the revised manuscript, we have added the results in the methodology (Fig. S2), which is also responded to another referee's 4th comment.

Technical corrections:

1) P5150, L17-19: revise "PM2.5 is a complex mixture of sulfate, nitrate, ammonium, water, organic and element carbon, soil dust, trace elements : : :" to "PM2.5 is a complex mixture of sulfate (SO_4^{2-}), nitrate (NO_3^-), ammonium (NH_4^+), water (H_2O), organic and element carbon (OC and EC), soil dust, trace elements : : :" Since those ions are analyzed later, the full name of an abbreviation should be given. In addition, cite the related references for this conclusion;

Response: As suggested, we have revised, and cited relevant references (Seinfeld et al., 2004; Zhang et al., 2013).

Seinfeld, J.H., Carmichael, G.R., Arimoto, R., Conant, W.C., Brechtel, F.J., Bates, T.S., Cahill, T.A., Clarke, A.D., Doherty, S.J., Flatau, P.J.: ACE-ASIA-Regional climatic and atmospheric chemical effects of Asian dust and pollution. Bulletin of the American Meteorological Society, 85, 367-380, 2004.

Zhang, R., Jing, J., Tao, J., Hsu, S. C., Wang, G., Cao, J., Lee, C. S. L., Zhu, L., Chen, Z., Zhao, Y., and Shen, Z.: Chemical characterization and source apportionment of PM2.5 in Beijing: seasonal perspective, Atmos. Chem. Phys., 13, 7053-7074, doi: 10.5194/acp-13-7053-2013, 2013.

2) P5152, L4: please cite the original paper of the MODIS AOD product;

Response: (should be P5151). The reference (Remer et al., 2005) has been added.

Remer, L. A., Kaufman, Y., Tanré, D., Mattoo, S., Chu, D., Martins, J. V., Li, R.-R., Ichoku, C., Levy, R., and Kleidman, R.: The MODIS aerosol algorithm, products, and

validation, Journal of the atmospheric sciences, 62, 2005.

3) P5152, L7-9: please cite the reference for this conclusion;

Response: (should be P5151). As suggested, we have cited a reference (Tao et al., 2013) for this conclusion.

Tao, J., Zhang, L., Engling, G., Zhang, R., Yang, Y., Cao, J., Zhu, C., Wang, Q., and Luo, L.: Chemical composition of PM2.5 in an urban environment in Chengdu, China: Importance of springtime dust storms and biomass burning, Atmos. Res., 122, 270-283, doi:10.1016/j.atmosres.2012.11.004, 2013.

4) P5152, L24: What does "A total of 117 PM2.5 samples and 12 blank samples were collected : : :" mean? Is it "A total of 117 samples in which 12 of them are blank" or "117 valid samples and 12 blank invalid samples"? Please clarify;

Response: We have revised this sentence to "A total of 117 PM2.5 samples along with 12 blank samples —-" so that it is clearer.

5) P5158, L6-16: revise "EPA" to "Environmental Protection Agency (EPA)", and "HYSPLIT" to "HYbrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT)" The full name of an abbreviation should be given when it is firstly used. Please revise other similar errors;

Response: (should be P5157). As suggested, we have revised.

6) P5158, L15: please cite the original paper for the HYSPLIT model;

Response: We have added the following references.

Draxler, R.R. and Rolph, G.D., 2013. HYSPLIT (HYbrid Single-Particle Lagrangian Integrated Trajectory) Model access via NOAA ARL READY Website (http://www.arl.noaa.gov/HYSPLIT.php). NOAA Air Resources Laboratory, College Park, MD.

Rolph, G.D., 2013. Real-time Environmental Applications and Display sYstem (READY) Website (http://www.ready.noaa.gov). NOAA Air Resources Laboratory, College Park, MD.

7) P5158, L1-6: Since those values have already been shown in table 1, there is no need to write down all of them in the text;

Response: As suggested, we have removed these values from the text.

8) P5158, L1-10: some words (such as "even" in line 7 and "highest" in line 10) should be modified. There is no need to use "even" here. Apparently, Chengdu is not suffering the "highest" PM2.5 level compared with other cities based on table 1.

Response: As suggested, we have removed the wording "even" and revised that sentence to "Thus, Chengdu suffered from severe PM2.5 pollution problem around the world."

9) P5157, L19: there is no need to give a subtitle named "PM2.5 mass" here.

Response: As suggested, the subhead "PM2.5 mass" has been removed.

10) P5158, L20: (Fig. S1) should be (Fig. S2)?

Response: Thanks for the correction; it has been changed.

11) P5159, L10: "(Zdráhal et al., 2002)"?

Response: It has been corrected.

12) P5160, L6-9: please cite the reference for supporting this;

Response: We have added two references (Yan et al., 206; Edwards et al., 2003), as follows. However, we also revised the sentence to "These suggestions are in agreement with the fact that residents in Sichuan Basin used to utilize woods as energy source to generate heat in cold winter (Edwards et al., 2004; Yan et al., 2006)."

Yan, X., Ohara, T., and Akimoto H.: Bottom-up estimate of biomass burning in mainland

China. Atmos. Environ., 40, 5262–5273, 2006.

Edwards, R. D., Smith, K. R., Zhang, J., Ma,Y.: Implications of changes in household stoves and fuel use in China. Energy Policy, 32, 395–411, 2004.

13) P5160, L22-25: any references for supporting these conclusions?

Response: Two new references have been cited.

Cao, G., Zhang, X., and Zheng, F.: Inventory of black carbon and organic carbon emissions from China, Atmospheric Environment, 40, 6516-6527, 2006.

Zhao, B., Wang, P., Ma, J. Z., Zhu, S., Pozzer, A., and Li, W.: A high-resolution emission inventory of primary pollutants for the Huabei region, China, Atmospheric Chemistry and Physics, 12, 481-501, 2012.

14) P5161, L4: "Andreae et al., 2008" should be "Andreae and Merlet, 2001"?

Response: This reference has been missed in our original manuscript, and now has been added in the revised manuscript, as follows:

Andreae, M. O., and Rosenfeld, D.: Aerosol–cloud–precipitation interactions. Part 1. The nature and sources of cloud-active aerosols, Earth-Science Reviews, 89, 13–41, 2008.

15) P5162, L16-22: please cite the reference for setting those standards;

Response: A new reference has been cited.

World Health Organization. Regional Office for Europe. Air quality guidelines for Europe. No. 91. WHO Regional Office Europe, 2000.

16) P5164, L18-19: The full name of an abbreviation should be given when it is firstly used, e.g. SO2, NOx and NH3;

Response: Thanks for the suggestion; full names have been given.

17) P5165, L8: "Dan et al. (2004)" is missing in References;

Response: It has been added in the revised manuscript.

Dan, M., Zhuang, G., Li, X., Tao, H., and Zhuang, Y.: The characteristics of carbonaceous species and their sources in PM2.5 in Beijing, Atmospheric Environment, 38, 3443-3452, 2004.

18) P5166, L16: please cite the reference for this conclusion;

Response: Here four references (Querol et al., 2006; Ni et al., 2012; Yang et al., 2002; Machemer, 2004) have been cited originally in text. Moreover, we have added the following reference (Tian et al., 2012) in P5165, L16.

Tian, H., Zhao, D., Cheng, K., Lu, L., He, M., and Hao, J.: Anthropogenic atmospheric emissions of antimony and its spatial distribution characteristics in China, Environ. Sci. Technol., 46, 3973-3980, doi: 10.1021/es2041465, 2012.

19) P5169, L8: how you define "very well" here;

Response: We have removed the wording "very".

20) P5169, L11: actually, "10±10

Response: We have removed the wording "much".

21) P5169, L24-L28 and P5170, L1-7: where is the information from? Please cite the related references;

Response: We have added the Chengdu governmental website as the reference, from which we referred to a lot of information and reports (http://www.chengdu.gov.cn/), but none of which is available in journal articles. Thus, we also added a statement "—-, as summarized in Table 5, most of which were based on the official information and/or reports available at the Chengdu governmental website (http://www.chengdu.gov.cn/)"

22) P5179, L30: the year is missing;

Response: The year (2013) has been added.

23) P5188, revise "MODIS" to "Moderate-resolution Imaging Spectrometer (MODIS)". The full name of an abbreviation should be given when it is firstly used.

Response: (Should be 5187). It has been revised, as suggested.

Please also note the supplement to this comment:
http://www.atmos-chem-phys-discuss.net/14/C3591/2014/acpd-14-C3591-2014-supplement.pdf

Interactive comment on Atmos. Chem. Phys. Discuss., 14, 5147, 2014.