

## Interactive comment on "Measurement report: Seasonal, distribution and sources of organophosphate esters in $PM_{2.5}$ from an inland urban city in southwest China" by Hongling Yin et al.

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Thank you for your valuable comments and good advice on improving our manuscript. We are so sorry that the manuscript has some mistakes. The typos and wording of the manuscript, as well as the specific contents and references of the manuscript, have been revised as follows according to your comments. Specific comments on the manuscript 1. Introduction: line 30, the reference "Bacoloni, A. et al. 2008" was wrongly matched, since the referenced study measured water samples instead of air. Response: The reference "Bacoloni, A. et al. 2008" has been replaced by

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"Guo et all., 2016" and "Li et al., 2017". Guo, Z. M., Liu, D., Shen, K.J., Li, J. Yu, Z.Q. Zhang, G.: Concentration and seasonal variation of organophosphorus flame retardants in PM2.5 of Taiyuan City, China, Earth and environment (in chinese)., 44, 600-604. https://doi.org/10.14050/j.cnki.1672-9250.2016.06.002, 2016. Li, J., Xie, Z., Mi, W., Lai, S., Tian, C., Emeis, K.C.: Organophosphate esters in air, snow and seawater in the north atlantic and the arctic, Environ. Sci. Technol., 51, 6887-6896. https://doi.org/10.1021/acs.est.7b01289, 2017. 2. Introduction: line 32, the reference "Araki et al. 2014" didn't measured organisms, instead, they measured dust. Response: "Araki et al. 2014" was deleted. 3. Introduction: line 34, the reference "Matthews, et al., 1990, 1993". Both references are animal studies. Thus, stating "many scholars found that OPEs have negative effects on the human body. . ." is not appropriate. Response: Thank you very much for your advice .The word "organisms" was replaced by "human body". 4. Introduction: line 41, the reference " Covaci et al. 2007" focused on analytical method development instead of measurement reports, not sure if it is a good reference here. Response: It's regret that the manuscript has such a mistake. This reference was deleted and other references in the manuscript have been verified. 5. Introduction: line 53, change "14335" to "14,335". Response: Thank you very much for your advice in such a detail. It has been revised to "14,335". 6. Materials and Methods: line 72, (Sigma Aldrich, ? location? country?); Be consistent in the text in terms of listing instrument/chemical manufacturing info. Response: Sigma Aldrich is the reagent production company. The manufacturing information of instruments and reagents has been indicated in the manuscript, and the full text has been checked. 7. Results: line 124, "heavy or light polluted area" may be better. Response: Thanks for your advice. "polluted" has been revised to "pollution" in the manuscript. 8. Results: line 126-128, rephrase the sentence to make it more precise. Response: The sentence has been revised to: "These data were quite consistent with our previous study which studied the annual median concentration of OPEs in PM2.5 from December 2013 to October 2014 (Yin et al., 2015). Interestingly, the concentration of  $\Sigma$ 70PEs at the suburban site was similar to, or even higher than some

urban sites, which indicated more local sources of these compounds in the suburban area." 9. Results: line 136, "And they were lower than". Response: The grammatical problems in this manuscript have been carefully corrected. 10. Results: line 138, add a space before (Wang, T. et al.), Double check other places in the text to make the format consistent. Response: The typos of the manuscript have been proofread. 11. Results: section 3.3. "Seasonal and spatial variations of OPEs in PM2.5", starting line 186, there is a mis-match in Fig.2 with the context. Where are the seasonal variations presented in Fig.2? Only site variations were presented here. Response: We are so sorry for this mistake. Because the version we uploaded to the website is different from the first draft, the sequence numbers of figures have been adjusted. We forgot to change it here. Figure 1 refers to "levels and seasonal variation of  $\Sigma$ 70PEs at each sampling site". "Figure 2" in line 186 has been changed to "Figure 1". 12. Results: line 227, delete first "the". "Considering" instead of "Considered". Response: Thank you for your valuable comments. They were revised as suggested. 13. Results: line 228, 229, lowercase "the third ring road". Response: Thanks a lot. The "third Ring Road" has been revised to "the third ring road". 14. Results: line 229, maybe "the uniform patterns of OPEs levels and distribution across the city is understandable"? Response: This sentence has been revised to "the similar level and profile of OPEs across the city was understandable". 15. Results: line 229, delete "But". Response: "But" has been deleted. 16. Results: line 232, "shoemaking industrial parks are located in the suburbs". Response: This sentence has been revised to "...shoemaking industrial parks are located in the suburbs". Thank you for your advice. 17. Results: line 233, "high levels". Response: It was revised as suggested. 18. Results: line 235, delete "to the individual OPE". Response: "to the individual OPE" has been deleted. Thank you for pointing out this problem. 19. Results: line 257, 258, " their gas-particle distributions determine their concentrations in PM2.5". Response: Thank you for your correction. "...distributions determines" has been revised to "distributions determine". 20. Results: line 266, is it "Fig.4 showed" or "Fig.5 showed"? Response: We are so sorry for this mistake. Line 266 should be "Figure 5 showed" and it was revised in the

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manuscript. 21. Results: line 275, delete "so". Response: "So" has been deleted. Thanks for your advice. 22. Results: line 282, add "The correlations between" before actually listing pairs of OPE monomers. Response: "The relationships between" has been added. 23. Results: line 284, delete second "was". Response: The second "was" has been deleted. Thank you for reminding us of this mistake 24. Results: section 3.4.3 "Correlation analysis of OPEs and PM2.5 concentrations", you mentioned Fig. S2, in which you used Pearson correlation tests. Why not spearman's rank correlation tests as you used in Figure 5? Response: As we know, Pearson evaluates the linear relationship between the two variables, while Spearman evaluates the monotonic relationship between the two variables. According to the results of other literature (Wong et al., 2018) and our hypothesis, we think that PM2.5 concentration is linearly related to the content of OPEs. So we carried out Pearson correlation tests in Fig. S2 according to the hypothesis. The results showed that the correlation was very poor, which was totally different from what we expected. In order to emphasize the difference of correlation between OPEs/other pollutants and PM2.5 concentration, Pearson correlation test result was used. 25. Results: line 291, add "found" after "was". Response: "found" has been added. 26. Results: line 315, "different uses". Response: "us" has been revised to "uses" 27. Results: line 338,339, add a reference to the statement "Chengdu's wind has always been. . .". Response: Thanks for your advice. A website was added. "Chengdu is a city located in the interior of China" has been added to illustrate that its wind intensity is smaller than coastal cities. iijLhttps://baike.baidu.com/item/%E6%88%90%E9%83%BD/128473?fr=aladdiniijL 28. Conclusions and Implications: line 372, "compared to the levels of OPEs in other cities". Response: Line 372 has been changed to "compared to the levels of OPEs in other cities" 29. Conclusions and Implications, line 390, maybe change "not easy to degrade" to "persistent"? What do you mean by "have a high con-tent"?, change the wording to clarify. Response: The sentence in line 390 has been changed to "the chlorinated phosphate, especially TCPP and TCEP, which are highly toxic and persistent in the environment, have high concentrations in this

study." 30. Reference: line 486-488, where the reference was cited? Cannot locate it in the text "Tang, R., Keming, M.A., Zhang, Y., Mao, Q.: Health risk assessment of heavy metals of street dust in Beijing, Acta. Scientiae. Circumstantiae., 32, 2006-2015, https://doi.org/10.13671/j.hjkxxb.2012.08.029, 2012." Response: It has been deleted. 31. Reference: what is the novelty in this paper compared with your reference paper in Chinese (Line 512-514) "Yin, H.L., Li, S.P., Ye, Z.X., Yang, Y.C., Liang, J.F., You, J.J.: Pollution Level and Sources of 513 Organic Phosphorus Esters in Airborne PM2.5 in Chengdu City, Environ. Sci. (in chinese), 36, 3566-3572, https://doi.org/10.13227/j.hjkx.2015.10.003, 2015." Response: The article we published earlier is a report of our experiment results from only two sampling sites. The purpose of that paper was to report the pollution level and distribution of the atmospheric OPEs at urban and suburban sites. Interestingly, we found the seasonal variations of OPEs were significantly different from PM2.5 concentrations and PM2.5bound PAHs, etc.. So we carried out a more detailed experiment with six sampling sites in the second year. In this paper, except for reporting the level and seasonal variations of OPEs at six sites, we paid more attention to investigate the relationships and correlations among the target compounds or with influence factors and illustrate the potential sources of OPEs in PM2.5. For example, whether different functional areas affect the distributions of atmospheric OPEs, correlations of OPEs with environmental factors (vapor pressure, boiling points, etc.), correlations of OPEs with PM2.5 concentrations, correlations of OPEs in PM2.5 and soil, correlations of OPEs in indoor and outdoor air were all discussed. These differences are the innovation of this paper. 32. Reference: line 515-517, reference "Zhang, Q. H., Yang, W. N., Ngo, H. H., Guo, W. S., Jin, P. K., Dzakpasu, M.: Current status of urban wastewater treatment plants in China, Environ. Int., 92-93, 11-22, https://doi.org/10.1016/j.envint.2016.03.024, 2016" might not be a good reference to be used here. Response: Thanks for your advice. It has been deleted. 33. Figure 2: where is the seasonal variations? As only site variation is presented here. Response: There are some errors in the arrangement of the sequence number of the figure. Figure 1 refers to "levels and seasonal variation

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of  $\Sigma$ 70PEs at each sampling site", and Figure 2 refers to "the proportion of individual OPE in the  $\Sigma$ 7OPEs at each point". Figure 2 in the text should actually be Figure 1. 34. Figure 4: line 542, be consistent with your notations/subscripts in the manuscript, PM2.5 or PM2.5. Same issue in line 544 etc. Response: Thanks for your advice. All "PM2.5" appearing in the manuscript has been replaced by "PM2.5". 35. Figure 5: Line 544, Should be "Spearman's rank correlation coefficients". Double check other places to be consistent. Response: It has been revised to "Spearman's rank correlation coefficients". We have checked other places throughout the manuscript. 36. Table 1: line 549, "orientation" of what? wind direction? If so, may want to use a different term since suburb and downtown probably do not quite fit. Response: "Orientation" refers to the direction of the city, not the wind direction. It has been replaced with "sampling sites". 37. In Figure 5 "Spearman's ranks correlation coefficients between the concentrations of individual OPEs in PM2.5 samples" and Figure S2 "Scatter plot of OPEs and PM2.5", spearman's rank tests and Pearson's correlation coefficients were used. Could you explain more about the selection of two different correlation tests? Response: Pearson evaluates the linear relationship between the two variables, while Spearman evaluates the monotonic relationship between the two variables. According to the results of other literatures and our hypothesis, we selected the different test method. In addition, when choosing which of the two test methods to use, firstly we would use the data distribution map to determine whether the data was normal distribution or non normal distribution. If it was normal distribution, Pearson's correlation coefficients were used. If not, Spearman's ranks correlation coefficients were used.

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