Itemized Response to Reviewer's Comments

Ms. Ref. No.: egusphere-2024-4147

Title: Characteristics, main sources, health risks of $PM_{2.5}$-bound perfluoroalkyl acids in Zhengzhou, central China: From seasonal variation perspective

RESPONSE TO REVIEWERS

Reviewer Comments:

RESPONSE: We sincerely thank the valuable and constructive inputs of the reviewer on our manuscript. We believe that we have adequately addressed all comments and thus the current version has been greatly improved with those valuable comments. In the revised manuscript, all the modifications were highlighted in red.

General Comments:

While the data presented represent a tremendous undertaking in terms of samples collected, quality assurance, and depth of analysis, the manuscript readability could greatly benefit from an in-depth editorial review. There are numerous instances where it is either unclear or ambiguous what the authors are trying to convey (ex. lines 48 – 49 "PFAAs levels in the atmosphere have attracted adequate attention due to people breathe second by second." the phrase "systemative investigation" in line 109, and "The polypropylene tubes were used." in lines 168 – 169.) Analytically speaking, it is evident that the authors have taken careful consideration in their sampling and data analysis, with numerous blanks, quality assurance checks, and data validation methods. It is also appreciated that methods for calculating MDLs were provided, and gives evidence that the researchers were thorough in their investigation.

Response: Thank you for your valuable and constructive comments. Detailed revisions have been listed below.

Specific Comments:

Comment 1: In Figure 1, it is unclear whether the reported data is averaged across all seasons or is for a single season. Line 192 suggests the highest average concentration was 181.63 pg m^{-3}, but PFBA, PFOA, and PFOS all appear to have concentrations above 200 pg m^{-3} in the figure. Please clarify.

Response: The data in the manuscript (46.68 – 181.63 pg·m^{-3}) refers to the average PFAA concentrations for each season and the data in Figure 1 refers to the PFAA concentrations for the four seasons. These sentences have been rephrased in this manuscript.

Lines 193 – 201 (New Version): The PFAA average concentrations ranged from 46.68 to 181.63 pg·m^{-3} in Fig. 1 across four seasons. However, the increased airflow during pump operation enhanced the adsorption of gaseous PFAA on quartz filters (Turpin et al., 1994; McMurdo et al., 2008; Ahrens et al., 2012; Chang et al., 2024), which may lead to a slight overestimated of PFAA values in this study. The PFAA average concentrations were comparable to levels observed in Chengdu (150 pg·m^{-3}) (Fang et al., 2019), but significantly higher than those

recorded in Shenzhen (8.80 pg·m^{-3}) (Liu et al., 2015a) and the average concentration in China (39.84 pg·m^{-3}) (Han et al., 2019).

Line 215 (New Version): Fig. 1. Box diagram of 17 PFAA concentrations in $PM_{2.5}$ across four seasons.

Reference:

Fang, S., Li, C., Zhu, L., Yin, H., Yang, Y., Ye, Z., et al., 2019. Spatiotemporal distribution and isomer profiles of perfluoroalkyl acids in airborne particulate matter in Chengdu City, China. Sci. Total. Environ. 689, 1235-1243. http://dx.doi.org/10.1016/j.scitotenv.2019.06.498

Han, D., Ma, Y., Huang, C., Zhang, X., Xu, H., Zhou, Y., et al., 2019. Occurrence and source apportionment of perfluoroalkyl acids (PFAAs) in the atmosphere in China. Atmos. Chem. Phys. 19, 14107-14117. http://dx.doi.org/10.5194/acp-19-14107-2019

Liu, B., Zhang, H., Yao, D., Li, J., Xie, L., Wang, X., et al., 2015a. Perfluorinated compounds (PFCs) in the atmosphere of Shenzhen, China: Spatial distribution, sources and health risk assessment. Chemos. 138, 511-518. http://dx.doi.org/10.1016/j.chemosphere.2015.07.012

Comment 2: In Figure 2, it may be appropriate to increase the spacing between the text and the markers in the legend, if possible, to increase the readability of the figure.

Response: The spacing between the text and the markers in the legend has been increased to improve readability.

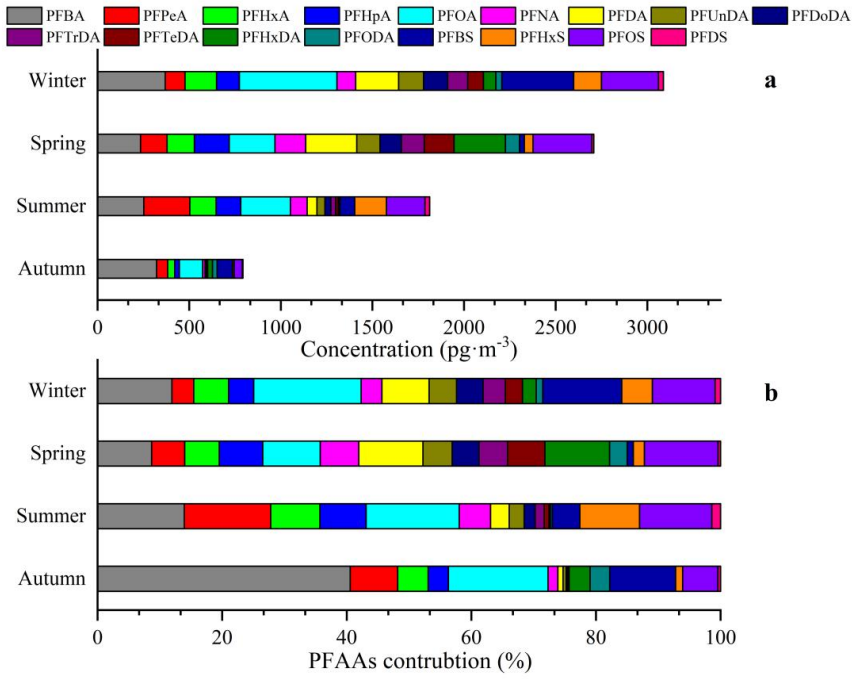

Fig. 2. PFAA concentrations characteristics across four seasons

Comment 3: While there are several mentions that this information can be used

to better shape policy to control PFAA levels in $PM_{2.5,}$ it is unclear what such policies could entail. Lines 298 – 303 suggest that management of local emissions is one way to control PFAAs, but it would be helpful to provide potential methods for emission reductions in industrial applications.

Response: In this manuscript suggestions for reducing PFAA pollution levels have been added, with the perspectives of PFAA proportion and air mass transport pathways.

Lines 242 – 245 (New Version): Long-chain PFAAs (e.g., PFOA and PFOS) were major pollutants and require replacement with short-chain alternatives (e.g., PFBS and PFPeA) or non-fluorinated substitutes such as silicon-based emulsifiers.

Lines 309 – 314 (New Version): For example, regulate PFAA emissions from textile and electroplating industries along southern urban in spring, collaborate with northwestern provinces to curb coal combustion in key transport cities in winter, establish pollution-blocking monitoring networks at northwestern entry points (e.g., Jiaozuo city and Jiyuan city) and leveraging the Taihang Mountains and Loess Plateau to intercept pollutants, in summer and autumn.

Comment 4: In line 159 of the supplement, "ED" is referred to as the burst time. It may be more appropriate to refer to this as the "exposure duration". I am assuming the authors use 72 as a general value for life expectancy. Also in line 159, the EF, exposure frequency is 350 days/year. Is this to suggest a two-week annual vacation? If so, perhaps it would be beneficial to more explicitly list the assumptions made, and why.

Response: The "ED" has been amended and more detailed information of EF has been added in this supplement.

Lines 158 – 160 (New Version): EF is the annual exposure frequency (350 $days \cdot year^{-1}$, without the time of two-week annual vacation), ED is exposure duration (72 a).

Comment 5: In line 226, “the researches have identified PFHxDA as…” is unclear. Perhaps “Previous studies have identified PFHxDA as…” would be better-suited here.

Response: This sentence has been rephrased.

Lines 232 – 234 (New Version): Previous studies have identified PFHxDA as a degradation byproduct of substances based on FTOHs (Ellis et al., 2004; Loewen et al., 2005).

Reference:

Ellis, D.A., Martin, J.W., De Silva, A.O., Mabury, S.A., Hurley, M.D., Andersen, M.P.S., et al., 2004. Degradation of fluorotelomer alcohols: A likely atmospheric source of perfluorinated carboxylic acids. Envion. Sci. Technol. 38, 3316-3321. http://dx.doi.org/10.1021/es049860w

Loewen, M., Halldorson, T., Wang, F.Y., Tomy, G., 2005. Fluorotelomer carboxylic acids and PFOS in rainwater from an urban center in Canada. Envion. Sci. Technol. 39, 2944-2951. http://dx.doi.org/10.1021/es048635b

Comment 6: In line 323, "The research indicated hat" should be "the research indicated that"

Response: This sentence has been revised.

Lines 336 – 339 (New Version): The research indicated that three primary kinds of chemicals related to PFOS-namely perfluorooctane sulfonates, substances containing these compounds and polymers were widely useded in industrial production (Xie et al., 2013).

Reference:

Xie, S., Wang, T., Liu, S., Jones, K.C., Sweetman, A.J., Lu, Y., 2013. Industrial source identification and emission estimation of perfluorooctane sulfonate n hina. Environ. Int. 52, 1-8. http://dx.doi.org/10.1016/j.envint.2012.11.004