

Interactive comment on “Occurrence and source apportionment of perfluoroalkyl acids (PFAAs) in the atmosphere in China” by Deming Han et al.

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A list of responses for comments from editors and reviewers

Dear Editors and Reviewers:

Thank you for your letter and for the reviewers' comments concerning our manuscript entitled "Occurrence and source apportionment of perfluoroalkyl acids (PFAAs) in the atmosphere in China" (Ref: acp–2019–676). These comments are valuable and very helpful for revising and improving our paper, as well as the important guiding significance to our researches. We have studied comments carefully and have made correction, the correction in the manuscript was marked–up with blue colour and underline (e.g. Revised Manuscript) which we hope meet with approval. The main corrections in

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the paper and the responds to the reviewer's comments are as flowing: Responds to the editors' and reviewers' comments:

Interactive comment on “Occurrence and source apportionment of perfluoroalkyl acids (PFAAs) in the atmosphere in China” by Deming Han et al.

Jing Ma jingma@shu.edu.cn Received and published: 16 August 2019

A national scale passive air sampling campaign was carried out in China, and 11 perfluoroalkyl acids compounds were determined in air samples during a whole year. The authors discussed concentration profiles, distributions and potential sources. The manuscript has the potential to add to the available body of evidence. I believe that the data are reliable and useful. In general, I recommend that the manuscript be accepted pending some minor revisions as outlined below. Response: Thank you for reviewer's appraisal of our manuscript. We appreciate reviewer's valuable comments for improving the manuscript.

Minor revisions: Query 1. There should be a space between numbers and units, like line 96 and line 130, –20°C. Response: Thanks for reviewer's suggestion. The format of number and units in lines 96 and 130 were revised in the revised manuscript, and all other were checked and revised throughout the revised manuscript.

Query 2. Line 29, the authors listed the environment like atmosphere, water, or snow, or in wildlife and even in the human body, however, the references cited seemed not match. Response: Considering reviewer's suggestion, the references cited were revised. It was changed as “PFAAs can be released to the surrounding environment during manufacturing and use of PFAAs containing products, which are ubiquitous in the environment (e.g., in the atmosphere, water, or snow) (Dreyer et al., 2009;Hu et al., 2016;Wang et al., 2017), in wildlife (Sedlak et al., 2017), and even in the human body (Cardenas et al., 2017;Tian et al., 2018).” in lines 30–33 in the revised manuscript.

Query 3. Line 32, the long–chain perfluoroalkyl carboxylic acids should be defined as

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C \geq 8. Response: To date, there are at least two classification for PFAAs, one for long-chain of C \geq 8 and short-chain of C \leq 7 (Dreyer et al. 2009; Liu et al. 2015), while the other for long-chain of C \geq 7 and short-chain of C \leq 6 (Liu et al. 2015; Jin et al. 2015; Tian et al. 2018). In the present study, to compare with the PFAAs variations which conducted in China recently reported, the long-chain of PFAAs was selected as C \geq 7. Hence, this query has not been revised in the revised manuscript.

Query 4. Line 74, the PFCAs analogues abbreviations listed in brackets should be given the full name, because some of them occurred at the first time. Response: All of full names and abbreviations of the 13 PFAAs analogues, were listed in Table S1 in the Supporting Materials. Considering reviewer's suggestion, the description of "The PFAAs standards used were Wellington Laboratories (Guelph, ON, Canada) PFAC-MXB standard materials, including C5-C14 PFCAs analogues (PFPeA, PFHxA, PFHpA, PFOA, PFNA, PFDA, PFUDA, PFDoA, PFTrDA, and PFTeDA), as well as C4, C6, and C8 PFSAs analogues (PFBS, PFHxS, and PFOS)." was changed to "The PFAAs standards used were Wellington Laboratories (Guelph, ON, Canada) PFAC-MXB standard materials, including C5-C14 PFCAs analogues (Perfluoropentanoic acid (PFPeA), Perfluorohexanoic acid (PFHxA), Perfluoroheptanoic acid (PFHpA), PFOA, Perfluorononanoic acid (PFNA), Perfluorodecanoic acid (PFDA), Perfluoroundecanoic acid (PFUDA), Perfluorododecanoic acid (PFDoA), Perfluorotridecanoic acid (PFTrDA), and Perfluorotetradecanoic acid (PFTeDA)), as well as C4, C6, and C8 PFSAs analogues (Perfluorobutane sulfonic acid (PFBS), PFHxS, and PFOS)." in lines 76-81 in the revised manuscript.

Query 5. Why did not the authors collect all the samples from urban area? Response: This research is a part of a large study aimed to investigate the occurrence and regional transportation of new emerging pollutants in China, in which one crucial pollutant was PFAAs. This investigation was implemented by Shanghai Jiao Tong University and Shanghai Academic of Environmental Science, and the sampling sites were selected based on the comprehensive effects of sampling geographical location, availability of

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volunteers, convenience of exchanging sorbent of XAD-PAS, and some other factors. Therefore, 20 urban sampling sites and 3 rural sampling sites were selected in this investigation ultimately.

Query 6. Please give the information on Amberlite XAD-2 resin. Response: Considering reviewer's suggestion, detailed information on XAD-2 was added in lines 99-100 in the revised manuscript, as "The particle size of XAD-2 is \sim 20-60 mesh, with water content of 20%-45%, its specific surface area \geq 430 m²/g, and the reference adsorption capacity \geq 35 mg/g."

Query 7. If the MDL was derived from three times SD of the field blank values, the authors should give the information about the field blanks and laboratory blanks. Which compounds were detected in those blanks? And in what level? Response: According to reviewer's suggestion, more information about field blanks and laboratory blanks was added in the revised manuscript and supporting materials. For instance, the description of "A total of 8 field blanks and 26 laboratory blanks were analyzed, and all the results were corrected according to the blank and recovery results.", was reworded as "A total of 8 field blanks and 26 laboratory blanks were analyzed, with individual blank values of N.D. (not detected)-1.1 pg/m³ and N.D.-1.3 pg/m³, respectively. All the results were corrected according to the blank and recovery results." in lines 149-150 in the revised manuscript.

Query 8. Did the authors use the matrix spike? Is there any matrix effect in passive air samples? Response: To control and assurance the PFAAs analysis quality, except for strictly pre-cleaning of XAD and HPLC-MS/MS experimental operation, we also conducted internal standards recovery experiment, field blank experiment, and laboratory blank experiment. Results showed that the mean spiked PFAAs recoveries ranged from 81% \pm 25% to 108% \pm 22%, the field blanks and laboratory blanks values were N.D.-1.1 and N.D.-1.3 pg/m³, respectively, and all the results were corrected according to the blank and recovery results. Considering all these above results and several reported researches, the matrix spike experiment was not used in this research.

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Special thanks to you for your good comments!

Reference Cardenas, A., Gold, D. R., Hauser, R., Kleinman, K. P., Hivert, M. F., Calafat, A. M., Ye, X., Webster, T. F., Horton, E. S., and Oken, E.: Plasma Concentrations of Per- and Polyfluoroalkyl Substances at Baseline and Associations with Glycemic Indicators and Diabetes Incidence among High-Risk Adults in the Diabetes Prevention Program Trial, *Environ Health Perspect*, 125, 107001, 2017. Hu, X. C., Andrews, D. Q., and Lindstrom, A. B.: Detection of Poly- and Perfluoroalkyl Substances (PFASs) in U.S. Drinking Water Linked to Industrial Sites, Military Fire Training Areas, and Wastewater Treatment Plants, *Environ Sci Technol Lett*, 3, 344-350, 2016. Karásková, P., Codling, G., Melymuk, L., and Klánová, J.: A critical assessment of passive air samplers for per- and polyfluoroalkyl substances, *Atmos Environ*, 185, 186-195, 2018. Sedlak, M. D., Benskin, J. P., Wong, A., Grace, R., and Greig, D. J.: Per- and polyfluoroalkyl substances (PFASs) in San Francisco Bay wildlife: Temporal trends, exposure pathways, and notable presence of precursor compounds, *Chemosphere*, 185, 1217-1226, 2017. Tian, Y., Zhou, Y., Miao, M., Wang, Z., Yuan, W., Liu, X., Wang, X., Wang, Z., Wen, S., and Liang, H.: Determinants of plasma concentrations of perfluoroalkyl and polyfluoroalkyl substances in pregnant women from a birth cohort in Shanghai, China, *Environment International*, 119, 165-173, 2018. Wang, Q. W., Yang, G. P., Zhang, Z. M., and Jian, S.: Perfluoroalkyl acids in surface sediments of the East China Sea, *Environ Pollut*, 231, 59-67, 2017. Dreyer, A., Kirchgeorg, T., Weinberg, I., Matthias, V. Particle-size distribution of airborne poly- and perfluorinated alkyl substances. *Chemosphere* 129, 142-149, 2015.

We tried our best to improve the manuscript and made some changes in the manuscript. These changes will not influence the content and framework of the paper. We appreciate for Editors/ Reviewers' warm work earnestly, and hope that the correction will meet with approval. Once again, thanks very much for your comments and suggestions.

Yours sincerely,

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Best regards!

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Please also note the supplement to this comment:

<https://www.atmos-chem-phys-discuss.net/acp-2019-676/acp-2019-676-SC2-supplement.pdf>

Interactive comment on *Atmos. Chem. Phys. Discuss.*, <https://doi.org/10.5194/acp-2019-676>, 2019.

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