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Atmospheric Chemistry and Physics Discussions

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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Dear Reviewers #2:

Thank you for your 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 the paper and the responds to the your comments are as flowing:

Interactive comment on "Occurrence and source apportionment of perfluoroalkyl acids (PFAAs) in the atmosphere in China" by Deming Han et al. Anonymous Referee #2 Received and published: 9 September 2019

General comments. This study provides a nationwide dataset of PFAAs in the Chinese atmosphere. It included 23 sampling locations at which XAD-PAS were deployed for one year and samples were taken approximately every month. The results were evaluated with regard to tempo-spatial variations and sources attribution was done using correlations, Hysplit backward trajectories and a PMF receptor models. As most available studies on PFAAs in the atmosphere are derived from single or only a few sampling sites, this nationwide study is of interest to the international community to better understand the atmospheric distribution of PFAAs. Additionally, China is a country of specific interest as large parts of the PFAS production were shifted from countries in Western Europe, the US and Japan to China and other Asian countries. Response: Thanks for your appraisal for ou manuscript. We appreciate your valuable comments for improving our manuscript.

Major comment: Query (1). A major query refers to the description and discussion of the used sampling technique. In different parts of the manuscript, it is stated that XAD-PAS collects representative portions of both the particle and the gas phase (line 62, line 191). However, it is reported in other publications that XAD-PAS collects primarily the gas phase (Lai et al., 2018; Melymuk et al., 2014). This difference should be discussed somewhere in the manuscript. Response: Thanks for the reviewer's good suggestion. As reported by previous researches (Melymuk et al., 2014; Lai et al., 2018), XAD could sample solely gas-phase pollutants, while PUF is able to accumulate both gas-phase and particle associated semi-volatile pollutants, although the particles sampled with a low accuracy and variable sampling rates. However, due to the XAD-PAS sampler design,the atmospheric aerosols bound PFAAs could moved into the sampler. Especially for the aerosol size distributions of particle bound PFASs varied with individual specie, e.g. the airborne PFASs, PFOA was predominantly (>70%) observed in small

size fraction (<0.14 μm) (Dreyer et al., 2015). In fact, Okeme et al., (2016) employed XAD-Pocket PAS to sample gaseous and particulate SVOCs in indoor environment, with finding not consistent with previous result that XAD-PAS could sample gas phase pollutant singly. And suggested that the sample efficiency of XAD sorbent sampler for gaseous and particulate phases pollutants need further investigations. Considering reviewer's suggestion, this differences were discussed and added, detailed as following: (1). The description of "However, recent field studies have confirmed their occurrence in gaseous phase (Lai et al., 2018;Cassandra et al., 2018;Ahrens et al., 2013)." in lines 57-58 in the original manuscript, was changed to "However, recent field studies have confirmed their occurrence in gaseous phase (Cassandra et al., 2018;Ahrens et al., 2013), e.g. Fang et al., (2018) found the total concentrations of C2, C4–C10 PFCAs and C6 and C8 PFSAs in the gas phase were 0.076–4.0 pg/m3 in the air above the Bohai and Yellow Seas, China." in lines 61-64 in the revised manuscript. (2). The description of "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. We should keep in mind that the unimpeded movement of particle bound PFAAs would be captured during sampling using XAD-PAS, which cannot differentiate PFAAs between gas and particle phases. Despite some research suggest the sampling efficiency of gas and particle phase PFAAs were similar (Karásková et al., 2018). In the present study, the two phases PFAAs sampled by XAD-PAS were treated as the whole atmosphere PFAAs concentration." was added in lines 106-111 in the revised manuscript. (3). "Fortunately, a number of reports showed that the XAD (a styrene–divinylbenzene copolymer) impregnated sorbent based passive air sampler (SIP–PAS) and XAD based PAS (XAD–PAS), were proven to be an ideal alternative sampling tool for monitoring PFAAs in a wide region, which was suggested to collect a representative sample of both gas and particle phases (Lai et al., 2018;Pavlína et al., 2018)." in lines 60-63 in the original manuscript, was changed to "Fortunately, a number of reports showed that the XAD (a styrene–divinylbenzene copolymer) impregnated sorbent based passive air sampler (SIP–PAS) and XAD based PAS (XAD–PAS), were proven to be an

ideal alternative sampling tool for monitoring PFAAs in a wide region. Despite several publications suggested XAD-PAS collects primarily gaseous PFAAs (Melymuk et al., 2014; Lai et al., 2018) in the ambient, current findings were not consistent. Due to the unimpeded movements of particles into the sampler, XAD–PAS was indicated to collect a representative sample of both gas and particle phases (Ahrens et al., 2013; Okeme et al., 2016; Karásková et al., 2018). Moreover, the dominant sorbent for fluorinated compounds was reported as XAD resin in the XAD impregnated SIP–PAS, instead of PUF themselves (Krogseth et al., 2013)." in lines 65-73 in the revised manuscript.

Query (2). Moreover, the comparison of the reported concentrations with measurements in other regions in section 3.1 can be skewed because of different sampling techniques and sampling media. If a comparison like this is done, the differences between the sampling techniques and their possible effects on the results should be discussed in a paragraph. Response: According to reviewer's suggestion, the limitation of direct comparison between PFAAs concentration and other measurements was discussed and added in the revised manuscript, as following: "Although there existed inherent differences of PFAAs levels between regions, the impacts from differences in sampling techniques and sorbents between XAD-PAS and SIP-PAS could not be neglected. As indicated by previous researches, XAD has much higher sorptive capacity of PFASs than PUF, wind speed and temperature displayed different degrees of impact on their sampling capacity among different regions. Additionally, UV radiation has the potential to degradate PFAAs due to O3, OHÂů, and other atmospheric oxidants during sampling. " in lines 202-206 in the revised manuscript.

Query (3). The manuscript is well structured and the reader can easily follow the drain of thoughts. However, it still contains several typing and grammar errors. Some are addressed in the section "technical corrections", but this is not exhaustive. Further proofreading by a native speaker would improve the manuscript. Response: Thanks for the reviewer's hard work on reviewing our manuscript. According to reviewer's suggestion, we have sent the revised manuscript to a professional English language editing service

provider in science. This revised manuscript was revised carefully and checked line by line, numerous grammaticalÂămistakes and errors were corrected. For example, “to investigate their occurences” in line 12 in the original manuscript, was reworded as “to investigate their occurrences” in line 13 in the revised manuscript; “was reported ranks as” in line 167 in the original manuscript, was changed to “was reported to rank as” in line 188 in the revised manuscript.

Query (4). Specific comments - The number of significant digits should be consistent throughout the manuscript. Response: According to reviewer’s suggestion, the number of significant digits of concentrations were revised, and kept consistent throughout the manuscript.

Introduction Query (5). -Line 24: PFASs include per- and polyfluoroalkyl substances and not only polyfluoroalkyl substances as stated in this line. Response: As suggested by reviewer, the description of line 24 in the original manuscript was changed to “Perfluoroalkyl acids (PFAAs) are one class of ionic polyfluoroalkyl substances (PFASs), which have excellent characteristics in terms of chemical and thermal stability, high surface activity, and water and oil repulsion (Lindstrom et al., 2011;Wang et al., 2014).” in line 26 in the revised manuscript.

Query (6). -Line 32: In the PFAS community, usually the definition of Buck et al. (2011) is used to differentiate between short- and long-chain homologues. According to this, long-chain PFCAs possess 8 or more carbon atoms (7 perfluorinated carbon atoms plus the carboxy group). Response: According to reviewer’s suggestion, the classification of long-chain and short-chain PFAAs homologues were revised based on study of Buck et al. (2011). The description of “Of the PFAAs, the long–chain (C $\geq$7) perfluoroalkyl carboxylic acids (PFCAs) and (C $\geq$6) perfluoroalkyl sulfonic acids (PFSAs) are more toxic and bio–accumulative than their short–chain analogues (Konstantinos et al., 2010).” in line 32 in the original manuscript, was reworded as “Of the PFAAs, the long–chain (C $\geq$8) perfluoroalkyl carboxylic acids (PFCAs) and (C $\geq$7) perfluoroalkyl sulfonic acids (PFSAs) are more toxic and bio–accumulative than their short–chain analogues

(Buck et al., 2011).” in lines 35-36 in the revised manuscript. Also, the corresponding result was revised, e.g. “To the contrary, a recent measurement found the long chain (C $\geq$ 8) PFCAs were much higher which conducted in the landfill atmosphere in Tianjin, China (Tian et al., 2018).” in lines 159-161 in the original manuscript, was changed to “Similarly, a recent PFAAs measurement conducted in the landfill atmosphere in Tianjin, China (Tian et al., 2018), found the long chain PFCAs were much higher than the short species.” in lines 182-183 in the revised manuscript.

Query (7). -Line 35/36: In May this year, the Parties to the Stockholm Convention adopted the listing of PFOA to Annex A. It would be good to add this new development to the text. Response: According to reviewer’s suggestion, the description of “This especially applies to perfluorooctanoic acid (PFOA) and perfluorohexane sulfonate (PFHxS) for which have been regulated in numerous countries, while perfluorooctane sulfonate (PFOS) have been added to Annex B of the Stockholm Convention in 2009 (Johansson et al., 2008).” in lines 34-36 in the original manuscript, was reworded as “This especially applies to perfluorooctanoic acid (PFOA), perfluorooctane sulfonate (PFOS) and perfluorohexane sulfonate (PFHxS), in which PFOS and PFOA have been added to Annex B and Annex A of the Stockholm Convention in 2009 and 2019, respectively, while PFHxS was under review by the Persistent Organic Pollutants Review Committee (Johansson et al., 2008; UNEP Stockholm Convention, 2019).” in lines 36-40 in the revised manuscript.

Query (8). -Line 63: Your references “Pavlina K et al., 2018” and “Karaskova P et al., 2018”, used later in the manuscript, is in fact the same publication. Please change it to “Karaskova P et al, 2018” in the whole manuscript, as Karaskova (not Pavlina) is the family name of the author. Response: Thanks for the reviewer’s hard work on reviewing our manuscript. The reference of “Pavlina K et al., 2018” was changed to “Karaskova P et al., 2018” in the revised manuscript, and the reference of “Pavlina K et al., 2018” was deleted in the revised manuscript.

Material and Methods Query (9). -Lines 85-86: Please add the number of sampling

sites for each of the seven divisions. Response: Considering reviewer's suggestion, the description of "These sampling sites were divided into seven administrative divisions: norther of China (NC), southern of 86 China (SC), central of China (CC), eastern of China (EC), northwest of China (NW), northeast of China (NE), and southwest of China (SW)." in lines 85-87 in the original manuscript, was reworded as "These sampling sites were divided into seven administrative divisions: norther China (NC, n=3 sites), southern China (SC, n=2), central China (CC, n=3), eastern China (EC, n=7), northwest of China (NW, n=3), northeast of China (NE, n=2), and southwest of China (SW, n=3)." in lines 98-100 in the revised manuscript.

Query (10). -Line 87: It would be helpful for the reader to understand from Figure S1 which sampling site belongs to which region (NC, SC etc.). This information could be given in the map itself or in the figure caption. Response: Considering reviewer's suggestion, the information of each sampling site belonging to which region was added in Figure S1 the revised manuscript. Detailed revision was as following: Figure R1. Revised figure S1 in the manuscript, the upper for the original one, the bottom for the revised figure.

Query (11). -Line 121: Usually, "A" refers to the aqueous phase and "B" to the organic solvent, not the other way round. It would avoid misunderstandings if this was turned around. Response: Considering reviewer's suggestion, the description of "The gradient elution program of the mobile phase A (methanol) and B (5 mmol/L aqueous ammonium acetate) was 20% A + 80% B at the start, 95% A + 5% B at 8 min, 100% a at 13 min, 20% A + 80% B at 14 min, and was maintained for 6 min." in lines 120-122 in the original manuscript, was reworded as "The gradient elution program of the mobile phase A (5 mmol/L aqueous ammonium acetate) and B (methanol) was 80% A + 20% B at the start, 5% A + 95% B at 8 min, 100% a at 13 min, 80% A + 20% B at 14 min, and was maintained for 6 min." in lines 138-140 in the revised manuscript.

Query (12). -Line 126: There should be a reference to Table S3, which includes the mass transitions. Response: Considering reviewer's suggestion, references of "

Karásková et al., 2018" and "Liu et al., 2015" were added to this table in the revised manuscript.

Query (13). -Line 134: Do the results refer to the linear isomer, e.g. of PFOS, or to the sum of all isomers? Response: Thanks for reviewer's good suggestion, this result refer to the liner isomer.

Query (14). -Line 138: Please add the information, which PFAAs could be detected in which type of blanks and with which standard deviations, either in the text or in Table S3. Response: Considering reviewer's suggestion, the detailed information of filed blanks and laboratory blanks was added in the revised Table S3 in the revised manuscript. Table R1 (Table S3). MS parameters, MDLs, LODs, LOQs values, recovery rates and blank values for individual compounds of PFAAs

Results and Discussion Query (15). - How are results below MDL given in this table? Does "0" refer to values below MDL? Please include this information. Response: Thank very much for reviewer's suggestion. The measured abundances of PFAAs which below MDL was marked as "0" in the original manuscript. Considering reviewer's suggestion and some statistics standard used, we have modified these values to "BDL" in Table S4 in the revised Supporting Materials, as following: Table R2 (Table S4). The measured abundances of PFAAs in this studyïijĹn=268ïijĽ

Query (16). - For some of the results, the median value is below the MDL given in Table S3 (e.g. for PFTeDA). How did you calculate these median values? Response: Thanks for the reviewer's suggestion. WeÂăvalueÂăthisÂăsuggestionÂăvery highly, checked these results carefully and found the MDL of PFTeDA was 0.14 rather 0.41, and revised this mistake. For the statistics analysis of measured concentrations, the results of BDL were replaced by 1/2 of the corresponding MDL values. Considering reviewer's suggestion, the description of "Statistical analyses were carried out by SPSS Statistics 22 (IBM Inc. US) and SigmaPlot 14.0 (Systat Software, US)." in line 147 in the original manuscript, was changed to "Statistical analyses were carried out by SPSS

Statistics 22 (IBM Inc. US), and the values of 1/2 MDL were used to replace these measured results of BDL. The statistics figures were depicted using technical software of SigmaPlot 14.0 (Systat Software, US)." in lines 166-168 in this revised manuscript.

Query (17). -Table 1: It would be helpful to know, which "PFAAs" are included in the sum given in the fifth column. Response: Considering reviewer's suggestion, more detailed information on species PFAAs was added in the note of "b"of PFAAs in Table 1, it was reworded as "b: represent the total concentration ranges of PFCAs and PFSAs; mean concentrations of the total PFCAs and PFSAs;" in Table 1 in the revised manuscript.

Query (18). -Line 199: It would be interesting which type of manufacturers are included in figure S3 and which industries are not? Response: Considering reviewer's suggestion, more detailed information on the fluoride manufacturer was added to the caption of Figure S3 in the revised Supporting Materials, "Figure S3. The spatial distributions of fluoride related products manufacturers in China (note that part of fluoride related industries were not included in this figure) and the different geographical conditions" "Figure S3. The spatial distributions of fluoride related products manufacturers in China and the different geographical conditions (note that the fluoride related manufacturers including textiles, crude plastic, paint coating, packaging materials, while part of fluoride related industries were not included in this figure)"

Query (19). -Line 201 to 209: Was this monthly variation stronger for specific sampling sites than for others? Response: The monthly variations of PFAAs varied based on site environments and local geographical conditions, the monthly variations of PFAAs in each site differed more or less. As shown in Figure S2, Beijing, Tianjin, and Xinjiang sites shared a similar monthly PFAAs variations, while Shanghai displayed a much different trend, which was controlled by local sources emissions as well.

Query (20). -Line 272 to 274: It would be helpful for the reader to get a short explanation (1-2 sentences) why the air mass origins shown in figure S5 were a driving

factor for PFAA variation. Response: Considering reviewer's suggestion, an explanation of the air mass origins in Table S5 was added in the manuscript, as"As illustrated in Figure S5, the 48 hours back trajectories were generally associated with air masses originating from the surrounding areas of the sampling locations, the trajectories which overlapped with urban areas in Zhejiang, Jiangxi and Shanghai, which confirmed that the air mass origins was a driving factor for PFAAs variation. " in lines 298-301 in the revised manuscript.

Query (21). -Line 300: The production of PFOA to use it as emulsifier in PTFE manufacturing is also an important direct source in China, isn't it? Response: We strongly agree with reviewer's suggestion, since PFOA is widely used in the manufacturing of polytetrafluoroethylene (PTFE), perfluorinated ethylene propolymer (FEP), and perfluoroalkoxy polymers (PFA). Considering reviewer's suggestion, the description of "PFOA was considered as the marker for the emulsification of plastics, rubber products, flame retardants for textiles, paper surface treatments, and fire foams (Liu et al., 2015;Konstantinos et al., 2010)." in lines 299-300 in the original manuscript, was reworded as "PFOA was considered as the marker for the emulsification of plastics, rubber products, flame retardants for textiles, paper surface treatments, fire foams and PTFE emulsifiers (Liu et al., 2015;Konstantinos et al., 2010)." in lines 326-237 in the revised manuscript.

Query (22). -Line 331: You state in the conclusion that the measured PFAAs were "several times to several magnitudes higher" than other urban atmosphere levels. This is not that obvious when reading 3.1 and looking at table 1. For example, the values reported for Brno are in a similar range as the results from this study, if I understand it correctly? Response: Considering reviewer's suggestion, this description of this conclusion was modified, "Results indicated that the measured PFAAs were several times to several magnitudes higher than other urban atmosphere levels, and much higher abundances existed in winter seasons compared with in the summer." in lines 330-332 in the original manuscript, was changed to "Results indicated that the measured PFAAs in the present study were several times to several magnitudes higher than the levels

conducted in most other urban locations, while far lower than the measurements implemented at point sources. In which, the C5–C14 PFCAs analogues occupied 79.6% of the total PFAAs variations, PFOA, PFHxA and PFOS ranked the top three species. Additionally," in lines 357-361 in the revised manuscript.

Technical corrections Query (23). - Line 15/16 "perfluorohexanoic" and "perfluoroheptanoic" have to be without "-" Response: According to reviewer's suggestion, the "perfluoro–hexanoic acid (PFHxA)", and "perfluoro–heptanoic acid (PFPeA)" was revised as "perfluoro–hexanoic acid (PFHxA)" and "perfluoro–heptanoic acid (PFPeA)" in the revised manuscript, respectively.

Query (24). -Line 21: It has to be "fluorotelomer-based" instead of "fluoro-telomere based" Response: Considering reviewer's suggestion, "fluoro-telomere based" in line 21 in the original manuscript was reworded as "fluorotelomer-based" in line 22 in the revised manuscript.

Query (25). - Line 65: "deployed" instead of "depolyed" Response: Considering reviewer's suggestion, "depolyed" in line 65 in the original manuscript was reworded as "deployed" in line 74 in the revised manuscript.

Query (26). -Lines 85-86. I think it has to be "north of China (NC)" or "northern China (NC)" instead of "northern of China (NC)". This also applies to the other regions. Response: Considering reviewer's suggestion, these related description were modified. For example, in the section of "2.2 Sample Collection", "These sampling sites were divided into seven administrative divisions: norther of China (NC), southern of China (SC), central of China (CC), eastern of China (EC), northwest of China (NW), northeast of China (NE), and southwest of China (SW)." in line 85-87 in the original manuscript, was changed to "These sampling sites were divided into seven administrative divisions: norther China (NC, n=3 sites), southern China (SC, n=2), central China (CC, n=3), eastern China (EC, n=7), northwest of China (NW, n=3), northeast of China (NE, n=2), and southwest of China (SW, n=3)." in lines 98-100 in the revised manuscript. In ab-

stract, the description of "Spatially, the content of PFAAs displayed a declining gradient trend of central areas > eastern areas > western areas, and Henan contributed as the largest proportion of PFAAs." in lines 17-19 in the original manuscript, was changed to "Spatially, the content of PFAAs displayed a declining gradient trend of central China> northern China> eastern China> northeast of China> southwest of China> northwest of China> southern China areas, and Henan contributed as the largest proportion of PFAAs." in lines 18-20 in the revised manuscript.

Query (27). -Line 160: "which conducted in the landfill atmosphere in Tianjin" does not connect to the rest of the sentence. Response: Considering reviewer's suggestion, "To the contrary, a recent measurement found the long chain ($C \geq 8$) PFCAs were much higher which conducted in the landfill atmosphere in Tianjin, China (Tian et al., 2018)." in lines 159-161 in the original manuscript, was changed to "Similarly, a recent PFAAs measurement conducted in the landfill atmosphere in Tianjin, China (Tian et al., 2018), found the long chain PFCAs were much higher than the short species." in lines 182-183 in the revised manuscript.

Query (28). -Line 167: "neutral PFASs in Chinese air" instead of "neural PFASs in China air" Response: Considering reviewer's suggestion, "Meanwhile, one major variation of PFOA precursor, 8:2 FTOH, was reported ranks as the highest concentration among neural PFASs in China air" in lines 166-167 in the original manuscript, was changed to "Meanwhile, one major variation of PFOA precursor, 8:2 FTOH, was reported to rank as the highest concentration among neural PFASs in air of China" in lines 189-190 in the revised manuscript.

Query (29). -Line 189: "may be could attribute" is ungrammatical. Response: Considering reviewer's suggestion, "The winter maxima abundance of PFAAs may be could attribute to the stagnant atmospheric conditions," in lines 189-190 in the original manuscript, was changed to "The winter maxima abundance of PFAAs could be attribute to the stagnant atmospheric conditions," in lines 215-216 in the revised manuscript.

Query (30). -Line 318: correlations “to” each other Response: Considering reviewer’s suggestion, the description of “In addition, these four analogues showed apparent positive correlations each other (r =0.59–0.79, p<0.01).” in lines 317-318 in the original manuscript, was changed to “In addition, these four analogues showed apparent positive correlations to each other (r =0.59–0.79, p<0.01).” in lines 344-345 in the revised manuscript.

References Buck, R.C., Franklin, J., Berger, U., Conder, J.M., Cousins, I.T., de Voogt, P., van Leeuwen, S.P., 2011. Perfluoroalkyl and polyfluoroalkyl substances in the environment: terminology, classification, and origins. Integr. Environ. Assess. Manag. 7(4), 513-541. doi:10.1002/ieam.258. Lai, F., Rauert, C., Gobelius, L., Ahrens, L. (2018) A critical review on passive sampling in air and water for per- and polyfluoroalkyl substances (PFASs). TrAC Trends in Analytical Chemistry. https://doi.org/10.1016/j.trac.2018.11.009 Loewen, M., Wania, F., Wang, F., Tomy, G., 2008. Altitudinal Transect of Atmospheric and Aqueous Fluorinated Organic Compounds in Western Canada. Environ. Sci. Technol. 42(7), 2374-2379. https://doi.org/10.1021/es702276c. Response: Thanks for the reviewer’s hard work on reviewing our manuscript.

Special thanks to you for your careful reading and good comments!

Reference Dreyer, A., Kirchgeorg, T., Weinberg, I., Matthias, V. Particle-size distribution of airborne poly- and perfluorinated alkyl substances. ChemosphereÂă129, 142-149, 2015. Fang, X., Wang, Q., Zhao, Z., Tang, J., Tian, C., Yao, Y., Yu, J., and Sun, H.: Distribution and dry deposition of alternative and legacy perfluoroalkyl and polyfluoroalkyl substances in the air above the Bohai and Yellow Seas, China, Atmos Environ, 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. Lai, F. Y., Rauert, C., Gobelius, L., and Ahrens, L.: A critical review on passive sampling in air and water for per- and polyfluoroalkyl substances (PFASs), TrAC Trends in Analytical Chemistry, Available online 23 Nov. 2018. Pavlína, K., Garry,

C., Lisa, M., and Jana, K.: A critical assessment of passive air samplers for per- and polyfluoroalkyl substances, Atmos Environ, 185, 186-195, 2018.

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,

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-SC3-supplement.pdf

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

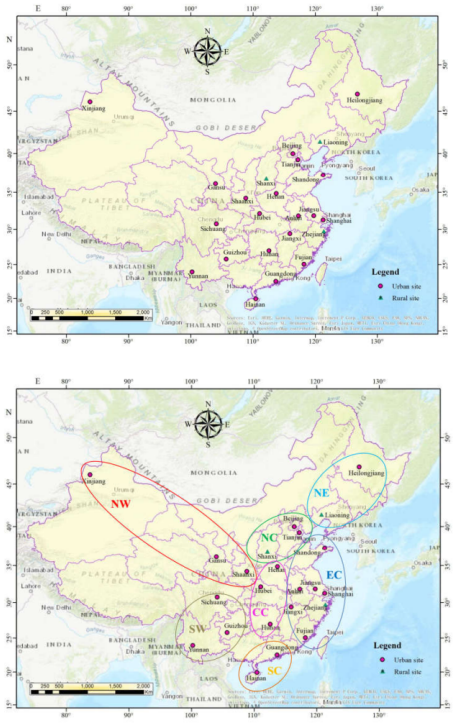

Figure R1. Revised figure S1 in the manuscript, the upper for the original one, the bottom for the revised figure.

Fig. 1.

Table R2 (Table S3). MS parameters, MDLs, LODs, LOQs values, recovery rates and blank values for individual compounds of PFAAs

Analogues	Parent ions (m/z)	Daughter ion (m/z)	Declustering potential (V) [a]	Collision energy (eV) [b]	Retention time (s)	MDLs (pg/m³)	LODs (pg/m³)	LOQs (pg/m³)	Recovery rate (%)	Filed bank (pg/m³)	Laboratory blank (pg/m³)	Internal Standards
PFCAs												
PFPeA	263	219	-40	–34	3.16	0.41	0.31	1.05	96±17	0.41±0.14	0.22±0.17	1,2-$^{13}C_2$-PFHxA
PFHxA	313	269	-35	–36	3.42	0.18	0.14	0.47	108±22	0.48±0.06	0.37±0.39	1,2-$^{13}C_2$-PFHxA
PFHpA	363	319→169	-55	–28	3.70	0.22	0.16	0.55	93±16	0.62±0.07	0.22±0.32	1,2,3,4-$^{13}C_4$-PFOA
PFOA	413	369→169	-45	–39	3.99	0.33	0.26	0.87	91±13	0.93±0.11	0.41±0.29	1,2,3,4-$^{13}C_4$-PFOA
PFNA	463	419→219	-40	–44	4.32	0.61	0.46	1.53	89±17	0.57±0.20	0.20±0.25	1,2,3,4,5-$^{13}C_5$-PFNA
PFDA	513	469→219	-50	–47	4.67	0.56	0.42	1.39	93±11	0.35±0.19	0.28±0.22	1,2-$^{13}C_2$-PFDA
PFUdA	563	519→269	-45	–61	5.02	0.28	0.21	0.70	88±16	0.31±0.09	0.31±0.13	1,2-$^{13}C_2$-PFUdA
PFDoA	613	569→169	-45	–65	5.35	0.28	0.21	0.70	94±18	0.44±0.09	0.15±0.18	1,2-$^{13}C_2$-PFDoA
PFTrDA	663	619→169	-50	–59	5.64	0.34	0.26	0.87	102±17	0.09±0.11	0.05±0.11	1,2-$^{13}C_2$-PFDoA
PFTeDA	713	669→169	-65	–57	5.94	0.14	0.31	1.03	97±21	0.12±0.14	0.06±0.13	1,2-$^{13}C_2$-PFDoA
PFSAs												
PFBS	299	80→99	-45	–64	3.19	0.25	0.20	0.66	81±25	0.11±0.08	0.27±0.46	$^{18}O_2$-PFHxS
PFHxS	399	80→99	-55	–87	3.70	0.16	0.12	0.40	86±13	0.16±0.05	0.42±0.27	$^{18}O_2$-PFHxS
PFOS	499	80→99	-55	–98	4.31	0.24	0.19	0.63	95±15	0.75±0.08	0.54±0.61	1,2,3,4-$^{13}C_4$-PFOS
Internal Standards												
1,2-$^{13}C_2$-PFHxA	315	270	-75	–41	3.40	/	/	/	/	/	/	/
1,2,3,4-$^{13}C_4$-PFOA	417	372	-40	–41	3.99	/	/	/	/	/	/	/
1,2,3,4,5-$^{13}C_5$-PFNA	468	423	-84	–52	4.34	/	/	/	/	/	/	/
1,2-$^{13}C_2$-PFDA	515	470	-87	–51	4.69	/	/	/	/	/	/	/
1,2-$^{13}C_2$-PFUdA	565	520	-79	–61	5.02	/	/	/	/	/	/	/
1,2-$^{13}C_2$-PFDoA	615	570	-66	–55	5.35	/	/	/	/	/	/	/
$^{18}O_2$-PFHxS	403	103	-55	97	3.72	/	/	/	/	/	/	/
1,2,3,4-$^{13}C_4$-PFOS	503	80	-80	97	4.31	/	/	/	/	/	/	/

[a]: cited from Karásková et al., 2018.
[b]: cited from Karásková et al., 2018 and Liu et al., 2015.

Fig. 2.

Table R3 (Table S4). The measured abundances of PFAAs in this study（n=268）

Analogues	Detection frequency (%)	Average value (pg/m³)	Standard deviation (pg/m³)	Minimum value (pg/m³)	Maximum value (pg/m³)	Median value (pg/m³)
PFCAs						
PFPeA	84.8	4.96	4.77	BDL	35.2	3.55
PFHxA	92.1	5.36	7.17	BDL	79.7	3.73
PFHpA	94.7	3.42	3.71	BDL	28.9	2.39
PFOA	100	8.19	8.03	0.36	70.4	6.24
PFNA	96.6	3.07	2.77	BDL	22.7	2.52
PFDA	96.2	4.13	3.74	BDL	30.5	3.36
PFUdA	75.6	1.24	1.32	BDL	6.72	0.86
PFDoA	63.5	0.56	0.50	BDL	3.18	0.45
PFTrDA	37.3	0.58	0.56	BDL	3.57	0.47
PFTeDA	41.7	0.19	0.25	BDL	2.25	0.11
PFSAs						
PFBS	62.2	1.96	1.85	BDL	9.39	1.37
PFHxS	71.6	0.99	1.38	BDL	13.2	0.56
PFOS	100	5.20	4.30	0.34	25.5	3.87

BDL: below detection limit.

Fig. 3.