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

Anonymous Referee #2

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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.

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. 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.

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.

Specific comments - The number of significant digits should be consistent throughout the manuscript.

Introduction - Line 24: PFASs include per- and polyfluoroalkyl substances and not only polyfluoroalkyl substances as stated in this line. - 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) - 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. - 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.

Material and Methods - Lines 85-86: Please add the number of sampling sites for each

of the seven divisions. - 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. - 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. - Line 126: There should be a reference to Table S3, which includes the mass transitions. - Line 134: Do the results refer to the linear isomer, e.g. of PFOS, or to the sum of all isomers? - 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.

Results and Discussion - How are results below MDL given in this table? Does "0" refer to values below MDL? Please include this information. - 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? - Table 1: It would be helpful to know, which "PFAAs" are included in the sum given in the fifth column. - Line 199: It would be interesting which type of manufacturers are included in figure S3 and which industries are not? - Line 201 to 209: Was this monthly variation stronger for specific sampling sites than for others? - 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. - 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? - 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?

Technical corrections - Line 15/16 "perfluorohexanoic" and "perfluoroheptanoic" have to be without "-" - Line 21: It has to be "fluorotelomer-based" instead of "fluoro-telomere-based" - Line 65: "deployed" instead of "depolyed" - 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. - Line 160: "which conducted in the landfill atmosphere in Tianjin" does not connect to the rest of the sentence. - Line 167: "neutral PFASs in Chinese air" instead of "neural PFASs in China air" - Line 189: "may be could attribute" is ungrammatical. - Line 318: correlations "to" each other

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.

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