Supplement of

Occurrence and source apportionment of perfluoroalkyl acids (PFAAs) in the atmosphere in China

Deming Han et al.

Correspondence to: Jinping Cheng (jpcheng@sjtu.edu.cn)

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Supplementary material

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NINETEEN pages: FIVE tables and FIVE figures, TWO sections.
### Table S1. Physical and chemical properties of target PFAAs compounds

| Component                              | Abbreviation | Molecular structure | Molecular weight | Bio–concentration factor | log\(K_{ow}\) \(^b\) | \(P_i\) (mmHg) \(^c\) |
|----------------------------------------|--------------|---------------------|------------------|--------------------------|----------------------|----------------------|
| Perfluoroalkane carboxylic acids (PFCAs) |              |                     |                  |                          |                      |                      |
| Perfluoropentanoic acid                | PFPeA        | C4F9COOH            | 263.98           | 1.00                     | 5.29                 | 7.9±0.4              |
| Perfluorohexanoic acid                 | PFHxA        | C5F11COOH           | 313.98           | 1.00                     | 5.97                 | 3.1±0.5              |
| Perfluoroheptanoic acid                | PFHpA        | C6F13COOH           | 363.97           | 1.00                     | 6.86                 | 0.5±0.6              |
| Perfluorooctanoic acid                 | PFOA         | C7F15COOH           | 413.97           | 1.90                     | 7.75                 | 0.3±0.7              |
| Perfluorononanoic acid                 | PFNA         | C8F17COOH           | 463.97           | 11.26                    | 8.64                 | 0.2±0.8              |
| Perfluorodecanoic acid                 | PFDA         | C9F19COOH           | 513.96           | 44.30                    | 9.53                 | 0.0±0.9              |
| Perfluoroundecanoic acid               | PFuDA        | C10F21COOH          | 563.96           | 128.19                   | 10.42                | 0.0±0.9              |
| Perfluorododecanoic acid               | PFDoA        | C11F23COOH          | 613.95           | 235.68                   | 11.31                | 0.0±1.0              |
| Perfluorotridecanoic acid              | PFTrDA       | C12F25COOH          | 663.95           | 474.19                   | 12.19                | 0.0±1.1              |
| Perfluorotetradecanoic acid            | PFTeDA       | C13F27COOH          | 713.95           | 1903.40                  | 13.08                | 0.0±1.2              |
| Perfluoroalkane sulfonic acids (PFSAs) |              |                     |                  |                          |                      |                      |
| Perfluorobutane sulfonic acid          | PFBS         | C4F9SO3H            | 299.98           | 1.00                     | 3.68                 | / \(^d\)              |
| Perfluorohexane sulfonic acid          | PFHxS        | C6F13SO3H           | 399.97           | 1.00                     | 5.25                 | /                    |
| Perfluorooctane sulfonic acid          | PFOS         | C8F17SO3H           | 499.97           | 1.00                     | 7.03                 | /                    |

\(^a\): Predicted data are generated using the Advanced Chemistry Development, Inc. (Canada), cited from (Yu, Liu et al. 2018);

\(^b\): Predicted octanol–water partitioning coefficients from individual PFAAs structure, cited from (Buck, Franklin et al. 2011, Yu, Liu et al. 2018);

\(^c\): Predicted pure compound vapor pressure, unit of mmHg at 298 K, cited from (Buck, Franklin et al. 2011, Yu, Liu et al. 2018);

\(^d\): “/” means lack of related data.
Table S2. The geographic information and annual temperature in different sampling sites of atmospheric PFAAs

| I.D. | Region        | Province | Type   | Location                        | Elevati on (m) | Monthly mean temperature (°C) | Gross Domestic Product (10^8 RMB) | Resident population (10^4) b | Crude plastic (10^4 tons) b |
|------|---------------|----------|--------|---------------------------------|---------------|-------------------------------|-----------------------------------|-------------------------------|-----------------------------|
| 1    | Northern of   | Beijing  | Urban  | Haidian District                | 31            | –5 – 24                       | 127.75                            | 2171                          | 28014.94                    |
| 2    | China, NC     | Tianjin  | Urban  | Jinnan District                 | 3.3           | –4 – 25                       | 332.42                            | 1557                          | 18549.19                    |
| 3    | Shanxi        | Rural    | Linshui County, Jincheng city   | 376           | –11 – 17                      | 79.47                            | 3702                            | 15528.42                    |
| 4    | Eastern of    | Shanghai | Urban  | Minhang District                | 4.5           | 5 – 28                        | 364.04                            | 2418                          | 30632.99                    |
| 5    | China, EC     | Zhejiang | Rural  | Yinzhou District, Ningbo City   | 4             | 4 – 23                        | 896.29                            | 5657                          | 51768.26                    |
| 6    | Jiangsu       | Urban    | Changzhou City                   | 5             | 2 – 26                        | 1175.39                           | 8209                            | 85869.76                    |
| 7    | Anhui         | Urban    | Yinquanym District, Fuyang City  | 30            | 2 – 27                        | 137.35                            | 6225                            | 27018                        |
| 8    | Fujian        | Urban    | Huian Country, Quanzhou City     | 30            | 12 – 26                       | 235.74                            | 3911                            | 32182.09                    |
| 10   | Jiangxi       | Urban    | Jiujiang City                    | 32.2          | 4 – 26                        | 25.46                             | 4622                            | 20006.31                    |
| 9    | Shandong      | Urban    | Laishan District, Yantai City    | 47            | –1 – 24                       | 710.42                            | 10006                           | 72634.15                    |
| 11   | Southern of   | Guangdong| Urban  | Nanshan District, Shenzhen City  | 7             | 15 – 26                       | 695.31                            | 11169                         | 89705.26                    |
| No. | Region                  | City          | District/County           | Meteorological Data    |
|-----|-------------------------|---------------|---------------------------|------------------------|
| 12  | China, SC               | Hainan        | Meilan District, Haikou City | 18 – 26, 19.67, 926, 4462.54 |
| 13  | Central of Hubei        | Urban         | Yunxi District, Shiyan City | 1 – 24, 191.86, 5902, 35478.09 |
| 14  | China, CC               | Henan         | Gaoxin District, Zhenzhou City | –2 – 26, 232.47, 9559, 44552.83 |
| 15  | Hunan                   | Urban         | Huaxin District, Hengyang City | 7 – 27, 48.4, 6860, 33902.96 |
| 16  | Northwestern of Xinjiang| Urban         | Tacheng City              | –14 – 18, 621.72, 2445, 10881.96 |
| 17  | China, NW               | Shaanxi       | Beilin District, Xi’an City | –1 – 24, 478.63, 3835, 21898.81 |
| 18  | Gansu                   | Urban         | Chengguang District, Lanzhou City | –7 – 19, 121.57, 2626, 7459.9 |
| 19  | Southwestern of Sichuan | Urban         | Shuangliu District, Chengdu City | 4 – 23, 214.94, 3789, 15901.68 |
| 20  | China, SW               | Yunnan        | Lanchang Country, Puer City | 3 – 19, 319.76, 4369, 23409.24 |
| 21  | Guizhou                 | Urban         | Xinren Country, Qiandongnan City | 6 – 22, 127.75, 2171, 28014.94 |
| 22  | Northeastern of Heilongjiang | Urban  | Beilin District, Suihua City | –22 – 19, 332.42, 1557, 18549.19 |
| 23  | China, NE               | Liaoning      | Rural                     | –12 – 21, 79.47, 3702, 15528.42 |

\[a:\] Meteorological data originated from China Meteorological Administration, http://www.cma.gov.cn/;

\[b:\] Data originated from China Statistic Yearbook 2018, National Bureau of Statistics China, http://www.stats.gov.cn/tjsj/ndsj/;
### Table S3. MS parameters, MDLs, LODs, LOQs values, recovery rates and blank values for individual compounds of PFAAs

| Analogues | Parent ions (m/z) | Daughter ions (m/z) | Declustering potential (V) | Collision energy (eV) | 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            | 1,2–13C₂–PFHxA    |
| PFHxA     | 313              | 269                 | -35                       | -36                   | 3.42              | 0.18          | 0.14         | 0.47          | 108±22           | 0.48±0.06          | 0.37±0.39            | 1,2–13C₂–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            | 1,2,3,4–13C₃–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            | 1,2,3,4–13C₄–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            | 1,2,3,4,5–13C₅–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            | 1,2–13C₃–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            | 1,2–13C₂–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            | 1,2–13C₂–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            | 1,2–13C₂–PFDa     |
| PFTeDA    | 713              | 669→169             | -65                       | -57                   | 5.94              | 0.14          | 0.31         | 1.03          | 97±21            | 0.12±0.14          | 0.06±0.13            | 1,2–13C₂–PFDa     |
| 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 | 18O2–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 | 18O2–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 | 1,2,3,4–13C4–PFOS |

**Internal Standards**

|        |        |        |      |        |        |        |        |        |                      |                      |
|--------|--------|--------|------|--------|--------|--------|--------|--------|----------------------|----------------------|
| 1,2–13C2–PFHxA | 315 | 270 | -75 | -41 | 3.40 | /    | /    | /    | /    | /    | /    | /    |
| 1,2,3,4–13C4–PFOA | 417 | 372 | -40 | -41 | 3.99 | /    | /    | /    | /    | /    | /    | /    |
| 1,2,3,4,5–13C5–PFNA | 468 | 423 | -84 | -52 | 4.34 | /    | /    | /    | /    | /    | /    | /    |
| 1,2–13C2–PFDA | 515 | 470 | -87 | -51 | 4.69 | /    | /    | /    | /    | /    | /    | /    |
| 1,2–13C2–PFUdA | 565 | 520 | -79 | -61 | 5.02 | /    | /    | /    | /    | /    | /    | /    |
| 1,2–13C2–PFDoA | 615 | 570 | -66 | -55 | 5.35 | /    | /    | /    | /    | /    | /    | /    |
| 18O2–PFHxS | 403 | 103 | -55 | 97  | 3.72 | /    | /    | /    | /    | /    | /    | /    |
| 1,2,3,4–13C4–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.
| 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.
Table S5. Correlation analysis of PFAAs in the atmosphere in China

|      | PFPeA | PFHxA | PFHpA | PFOA | PFNA | PFDA | PFUdA | PFDoA | PFTrDA | PFTcDA | PFBS | PFHxS |
|------|-------|-------|-------|------|------|------|-------|-------|--------|--------|------|-------|
| PFHxA| 0.70" |       |       |      |      |      |       |       |        |        |      |       |
| PFHpA| 0.12  | 0.31' |       |      |      |      |       |       |        |        |      |       |
| PFOA | 0.69" | 0.77" | 0.68" |      |      |      |       |       |        |        |      |       |
| PFNA | 0.66" | 0.66" | 0.65" | 0.70" |      |      |       |       |        |        |      |       |
| PFDA | 0.54" | 0.67" | 0.72" | 0.84" | 0.61"|      |       |       |        |        |      |       |
| PFUdA| 0.16  | 0.32  | 0.15  | 0.2  | 0.14 | 0.23 |       |       |        |        |      |       |
| PFDoA| 0.39' | 0.33  | 0.27  | 0.38 | 0.31 | 0.32 | 0.61" |       |        |        |      |       |
| PFTrDA| 0.53' | 0.48" | 0.3   | 0.42 | 0.44 | 0.51"| 0.65" | 0.62" |        |        |      |       |
| PFTcDA| 0.21  | 0.4   | 0.39' | 0.36' | 0.27 | 0.39 | 0.72" | 0.59" | 0.79" |        |      |       |
| PFBS | 0.68" | 0.26  | 0.15  | 0.15 | 0.39 | 0.14 | 0.23  | 0.28  | 0.18   |        |      |       |
| PFHxS| 0.57' | 0.69" | 0.27  | 0.42' | 0.57"| 0.64"| 0.3   | 0.43' | 0.54' | 0.38' | 0.28 |      |
| PFOS | 0.69" | 0.42' | 0.32  | 0.33 | 0.36 | 0.37' | 0.25  | 0.41' | 0.46' | 0.38  | 0.63"| 0.40' |

*: represent p <0.05;

**: represent p< 0.01.
Figure S1. Spatial distributions of 23 sampling sites of atmospheric PFAAs in China (including 20 urban sites, red circles; and three rural site, green triangles).
Figure S2. Temporal variations of PFAAs concentrations in selected four typical sites: Shanghai, Beijing, Xinjiang and Tianjin.
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)
Figure S4. The spatial distributions of fluoride related products manufacturers in Zhejiang site (a small village in Ningbo City)
Pu’er, Yunnan; Summer
Chengdu, Sichuan; Summer
Beijing; Summer
Suihua, Heilongjiang; Summer

Pu’er, Yunnan; Winter
Chengdu, Sichuan; Winter
Beijing; Winter
Suihua, Heilongjiang; Winter

Figure S5. The backward trajectories of air mass extracted by Hysplit trajectory model
Section S1. Sampling rate of XAD–PAS in this investigation

Sampling rate of XAD–PAS is a crucial factor to derive air concentrations from the amounts of chemicals accumulated in the XAD resin. Previous literature suggested the sampling rate of XAD–PAS of 3.5–4.5 m³/d for PFASs (Li, Vento et al. 2011, Liu, Zhang et al. 2015, Tian, Yao et al. 2018). However, the actual sampling rate is dynamically variable, and affected by several factors. In this study, a standard solution containing mass labeled 1,2,3,4–13C4–PFOA and 1,2,3,4–13C4–PFOS (20 ng/mL) was spiked directly onto the upper XAD resin in the Shanghai sampling site (Floor of 5-story building of School of Environmental Science and Engineering in Shanghai Jiao Tong University) for one month in April 2017, to account for analyte losses during sampling. The sampling rate was calculated as flowing formulas:

\[ R = -\ln\left(\frac{C_t}{C_0}\right) \times d \times A \times \left(\frac{K_{XAD}}{t}\right) = -\ln\left(\frac{C_t}{C_0}\right) \times V \times \left(\frac{K_{XAD}}{t}\right) \]  

(S1)

\[ \log K_{XAD} = 0.6366 \times \log \left(\frac{K_{OW}}{S_W/S_A}\right) \]  

(S2)

\[ S_A = \frac{P_L}{(RT)} \]  

(S3)

where \( C_t/C_0 \) represents the measured recoveries of 1,2,3,4–13C4–PFOA and 1,2,3,4–13C4–PFOS; \( V \) represents absorbent volume, 207.7 (cm³); \( K_{XAD} \) represent 13C8–PFOA partition coefficient between air and XAD; \( t \) represents sampling time, 30 d; \( K_{OW}, S_W, \) and \( S_A \), represent octanol–air partition coefficient (6.3), water solubility, and air solubility, respectively; \( P_L \) and \( R \) represent liquid vapor pressure and gas constant (8.314 J/(mol·K)), respectively.

The \( \log P_L, \) and \( \log S_W \) values was set as 1.3(Pa), and 0.24 (mg/L) in the present study.

The sampling rate of XAD–PAS was calculated as 3.2 m³/d in the selected geographical site. However, higher temperature and wind speed were suggested to have positive effect on sampler uptake efficiency, while negative effect on the sorption capacity. Although the sampling rate of PFAAs were proposed of site-specific under different meteorological conditions, we have not conduct the depuration compounds loss test in all the 23 sampling sites.

Since our calculated XAD–PAS rate value was very close to the recommended rate of 3.5–4.5 m³/d for PFAAs, the rate value of 3.2 m³/d was used in the present study.
Section S2. PMF analysis and uncertainty assessment

Positive matrix factorization (PMF) is considered an advanced algorithm among various receptor models, which has been successfully applied for source identification of environmental pollutants (Han, Fu et al. 2018; Han Fu et al. 2019). PMF has the following advantages: each data point is given an uncertainty–weighting; the factors in PMF are not necessarily orthogonal to each other and there is no non–negativity constraint with PMF. In the present study, PMF 5.0 (US EPA) was used to apportion the contributions of different sources to PFAAs in the atmosphere. The matrix X represents an ambient data set in which i represents the number of samples and j the number of chemical species. The goal of multivariate receptor modeling is to identify sources (p), the species profile (f) of each source and the amount of mass (g) contributed by each source to each individual sample as well as the residuals (eij), as following equation:

\[ X_{ij} = \sum_{k=1}^{p} g_{ik} f_{kj} + e_{ij} \]  \hspace{1cm} (S1)

The PMF solution minimizes the objective function Q based on these uncertainties (u):

\[ Q = \sum_{i=1}^{n} \sum_{j=1}^{m} \left[ \frac{x_{ij} - \sum_{k=1}^{p} g_{ik} f_{kj}}{u_{ij}} \right]^2 \]  \hspace{1cm} (S2)

The input data files of PMF consist of concentrations and uncertainty matrices, and the uncertainty data were calculated as Equation (S3) as suggested by PMF User Guide. The missing values were represented by average values, while measurements below MDL (method detection limit) were replaced by two times of the corresponding MDL values. The “weak” variables were down–weighted, while “bad” variables were omitted form the analysis process.

\[
\begin{align*}
\text{Unc}_{i} &= \frac{5}{2} \times \text{MDL}_{i} \quad \text{if } C_{i} \leq \text{MDL}_{i} \\
\text{Unc}_{i} &= \sqrt{(C_{i} \times \text{Error Fraction})^2 + \left(\frac{5}{2} \times \text{MDL}_{i}\right)^2} \quad \text{if } C_{i} > \text{MDL}_{i}
\end{align*}
\]  \hspace{1cm} (S3)

The model was run 20 times with 49 random seeds to determine the stability of goodness–of–fit values. If the number of sources is estimated properly, the theoretical Q value should be approximately the number of degrees of freedom or the total number of data points. Three to six factors were examined, and four factors were found to be the most appropriate
and most reasonably interpretable. $Q$ (True) is the goodness–of–fit parameter calculated including all points, while $Q$ (Robust) is the goodness–of–fit parameter calculated excluding points not fit by the model, $Q$ (Robust) and $Q$ (True) were 21672.9 and 25935, respectively, with $Q_{\text{true}}/Q_{\text{exp}}$ value of 12.56. Additionally, approximately 97% of the residuals calculated by PMF were within the range of −3 to 3, indicating a good fit of simulated results. The factor did not show oblique edges, suggesting there were little rotation for the solution. All these features implied the model simulation result was acceptable.
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