Supplementary information for

Fate of perfluoroalkyl substances in the Third Pole proglacial lake basins

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Content

Supplementary Note

Supplementary Note S1 The HPLC-MS/MS procedure for PFAS analysis.

The analytical procedure was previously reported in a number of studies [1, 2], as summarized below. The sample (10 μL) was injected onto an Acclaim 120 C18 column (5 μm, 100 Å, 150 mm × 4.6 mm, ThermoFisher Scientific, USA). Ten mM of ammoniom acetate (pH 4, A) and acetonitrile (B) were used as the mobile phases. The flow rate was set at 1 mL min^{-1}. The mobile phase gradient condition was started at 10% B, held constant for 1.5 min, increased to 95% by 4 min, held constant until 8 min, returned to the initial condition by 8.5 min, and then balanced for 1.5 min. The mass spectrometer (MS) was operated in the negative ion ESI-MS/MS, using the multiple reaction monitoring mode. The MS-related parameters were set as follows: sheath gas, 40 units; aux gas, 12 units; source voltage, 2500 V; vaporizer temperature, 350 °C; capillary temperature, 400 °C; and scan time, 0.01 s.

Supplementary Note S2 Sensitivity analysis for QWASI model

Through sensitivity analysis, it is possible to determine the most sensitive components of the model. The purpose of this analysis is to measure the sensitivity of parameters, forcing functions, or sub-models to state variables of greatest interest. In the practical modeling, sensitivity analysis is carried out by changing parameters, forcing functions, and sub-models and observing their corresponding effects on the selected state variables. Here, sensitivity analysis was only conducted for the fifty-six parameters. The sensitivity coefficient (*S*) was determined by changing the parameter by ±10%:

$$S = \frac{Y_{1.1} - Y_{0.9}}{0.2 \times Y}$$

where $Y_{1.1}$ and $Y_{0.9}$ represent the estimated concentrations when the tested parameter was changed at +10% and -10%, respectively. Parameters with a higher absolute sensitivity coefficient are more sensitive, with results presented in Supplementary Fig. S7.

Supplementary Table

Supplementary Table S1 Concentrations (pg g^{-1} dw) of PFAS in QY lake sediment samples.

Year	PFBA	PFPeA	PFHxA	PFHpA	PFOA	PFNA	PFDA	PFUdA	PFDoA	PFTrDA	PFBS	PFHxS	PFOS
2009	92.2	35.4	<LOD	10.9	145.9	17.4	19.7	32.8	8.50	<LOD	149.0	<LOD	141.4
2007	80.0	9.00	7.90	<LOD	146.5	23.2	26.4	33.9	18.1	4.80	159.7	<LOD	95.3
2005	44.5	13.2	<LOD	4.40	149.1	31.7	26.3	27.3	<LOD	<LOD	194.7	12.3	77.1
2003	28.4	15.9	6.00	<LOD	9.70	24.5	21.2	20.4	5.70	<LOD	95.9	<LOD	49.4
2002	236.1	15.0	<LOD	<LOD	141.2	29.1	8.10	21.2	6.60	<LOD	71.2	<LOD	73.1
2000	60.6	20.6	6.90	<LOD	168.9	24.8	14.5	29.0	8.50	<LOD	46.6	13.1	80.6
1999	34.0	20.7	<LOD	2.60	147.4	13.8	7.50	18.8	<LOD	<LOD	96.3	<LOD	122.5
1997	70.9	9.80	<LOD	<LOD	152.4	15.3	7.90	<LOD	<LOD	<LOD	178.3	12.7	109.1
1994	54.7	19.4	<LOD	2.90	145.1	12.4	8.50	10.0	<LOD	<LOD	101.5	12.9	61.7
1992	216.1	17.6	3.80	<LOD	16.2	12.9	4.80	9.90	<LOD	<LOD	61.0	12.9	63.1
1990	64.2	27.3	4.00	4.80	132.3	<LOD	<LOD	9.00	<LOD	<LOD	101.5	11.1	55.1
1988	32.6	19.7	<LOD	<LOD	141.3	6.90	<LOD	<LOD	<LOD	<LOD	120.5	10.0	74.2
1986	72.7	27.1	4.00	<LOD	117.7	<LOD	<LOD	<LOD	<LOD	<LOD	64.3	14.4	47.5
1983	237.6	23.2	<LOD	<LOD	3.30	<LOD	<LOD	<LOD	<LOD	<LOD	57.8	<LOD	46.2
1981	127.7	<LOD	10.1	<LOD	144.6	<LOD	<LOD	<LOD	<LOD	<LOD	215.1	21.4	36.6
1979	46.7	12.3	6.80	<LOD	146.9	<LOD	<LOD	<LOD	<LOD	<LOD	123.0	<LOD	113.7
1976	81.0	6.10	<LOD	0.60	103.2	<LOD	<LOD	<LOD	<LOD	<LOD	94.4	10.8	93.8
1973	45.3	14.2	4.50	<LOD	119.5	<LOD	<LOD	<LOD	<LOD	<LOD	89.4	8.20	69.2
1970	41.3	24.7	<LOD	<LOD	144.1	<LOD	<LOD	<LOD	<LOD	<LOD	53.9	<LOD	19.9
1967	133.2	9.20	<LOD	1.50	6.20	<LOD	<LOD	<LOD	<LOD	<LOD	33.1	<LOD	24.6
1963	32.0	8.70	<LOD	5.80	129.2	<LOD	<LOD	<LOD	<LOD	<LOD	86.6	<LOD	18.7
1961	107.2	21.0	<LOD	<LOD	5.60	4.70	<LOD	<LOD	<LOD	<LOD	155.3	<LOD	21.3
1958	120.8	20.1	<LOD	3.90	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	108.5	<LOD	14.7

Note: LOD means the limit of detection.

Supplementary Table S2 Concentrations (pg g^{-1} dw) of PFAS in PM lake sediment samples.

Year	PFBA	PFPeA	PFHxA	PFHpA	PFOA	PFNA	PFDA	PFUdA	PFDoA	PFTrDA	PFBS	PFHxS	PFOS
2013	72.1	60.3	12.9	14.5	115.3	12.3	5.90	<LOD	<LOD	<LOD	24.1	14.8	24.0
2011	65.2	119.5	29.1	23.3	65.7	9.80	<LOD	<LOD	<LOD	<LOD	125.4	61.4	190.5
2008	49.7	92.6	34.3	31.7	69.2	<LOD	<LOD	<LOD	<LOD	<LOD	146.0	25.9	272.6
2004	173.8	132.1	24.9	26.6	13.8	<LOD	<LOD	<LOD	<LOD	<LOD	308.2	26.5	361.5
2000	162.6	14.3	11.8	39.6	4.60	<LOD	<LOD	<LOD	<LOD	<LOD	382.0	42.0	222.6
1997	41.4	19.7	34.0	9.10	72.1	<LOD	<LOD	<LOD	<LOD	<LOD	197.7	38.0	296.2
1994	213.4	28.7	40.9	24.1	8.40	<LOD	<LOD	<LOD	<LOD	<LOD	222.1	22.9	336.3
1990	86.2	31.1	44.1	21.0	58.3	<LOD	<LOD	<LOD	<LOD	<LOD	185.4	21.5	274.1
1986	128.3	21.0	26.1	<LOD	7.20	8.50	<LOD	<LOD	<LOD	<LOD	253.8	12.8	156.0
1983	160.8	38.0	18.3	2.40	7.40	<LOD	<LOD	<LOD	<LOD	<LOD	201.5	26.5	338.6
1980	57.3	25.4	5.40	5.90	60.2	<LOD	<LOD	<LOD	<LOD	<LOD	227.8	3.20	273.2
1978	21.3	44.7	25.0	13.6	72.9	<LOD	<LOD	<LOD	<LOD	<LOD	194.4	13.0	224.0
1977	116.6	26.8	26.7	5.70	56.6	<LOD	<LOD	<LOD	<LOD	<LOD	70.0	64.3	213.7
1973	471.0	16.4	15.1	9.10	6.40	<LOD	<LOD	<LOD	<LOD	<LOD	174.0	<LOD	227.9
1969	40.0	10.7	32.2	28.5	69.3	<LOD	<LOD	<LOD	<LOD	<LOD	167.3	94.7	87.1
1967	65.7	6.50	15.3	13.9	85.1	<LOD	<LOD	<LOD	<LOD	<LOD	115.1	103.1	82.0
1966	74.5	24.0	19.2	9.50	86.5	<LOD	<LOD	<LOD	<LOD	<LOD	93.2	126.1	75.1
1963	31.6	14.0	22.6	18.0	80.4	<LOD	<LOD	<LOD	<LOD	<LOD	92.5	186.0	93.3
1961	15.1	11.3	8.00	<LOD	83.2	<LOD	<LOD	<LOD	<LOD	<LOD	87.4	182.4	87.5
1958	166.3	8.10	12.3	9.40	76.2	<LOD	<LOD	<LOD	<LOD	<LOD	73.6	144.6	53.4

Note: LOD means the limit of detection.

Supplementary Table S3 Component proportion and detected rate of individual PFAS in sediment samples from the QY and PM lakes.

Analytes		Component proportion		Detected rate	
		QY	PM	QY	PM
PFCAs	PFBA	21.3%	16.6%	100.0%	100%
	PFPeA	4.0%	5.6%	95.7%	100%
	PFHxA	0.6%	3.4%	39.1%	100%
	PFHpA	0.4%	2.3%	52.2%	90%
	PFOA	24.9%	8.3%	95.7%	100%
	PFNA	2.2%	0.2%	52.2%	15%
	PFDA	1.5%	0.1%	43.5%	5%
	PFUdA	2.2%	0.0%	43.5%	0%
	PFDoA	0.5%	0.0%	13.0%	0%
	PFTrDA	0.0%	0.0%	4.3%	0%
	Sum	**57.6%**	**36.5%**	—	—
PFSAs	PFBS	25.4%	25.1%	100.0%	100%
	PFHxS	1.4%	9.1%	47.8%	95%
	PFOS	15.6%	29.2%	100.0%	100%
	Sum	**42.4%**	**63.5%**	—	—

Supplementary Table S4 Comparison of PFAS deposition fluxes in the sediment core and snow core on the Tibetan Plateau (ng m^{-2} y^{-1}).

Study regions	The Tibetan Plateau			
	The QY Lake Sediment core	The PM Lake Sediment core	Mt. Zuoqiupu Snow core	Mt. Muztagata Snow core
Time	1958-2009	1958-2013	1996-2007	1980-1999
PFBA	19.5-283	18.1-424	21.8-62.1	ND
PFPeA	3.7-31.8	5.6-106	32.0-46.4	20.0 - 90.0
PFHxA	<LOD-8.6	4.9-55.0	6.3-44.8	7.43-63.4
PFHpA	<LOD-9.8	1.7-35.7	<LOD	<LOD
PFOA	1.1-169	4.1-162	27.3-188	15.1-154
PFNA	<LOD-34.9	<LOD-11.1	18.1-22.3	2.68-26.1
PFDA	<LOD-29.0	<LOD-5.3	30.2-56.3	2.14-31.7
PFUdA	<LOD-37.3	<LOD-5.3	ND	1.92-6.32
PFDoA	<LOD-20.0	<LOD	ND	3.81-21.0
PFTrDA	<LOD-5.3	<LOD-2.3	ND	ND
PFBS	23.2-176	21.7-428	ND	ND
PFHxS	<LOD-17.1	2.9-261	ND	ND
PFOS	10.3-127	21.6-493	<LOD	20.8-195
Sum	150-725	326-1345	136-420	74.9-588
Reference	This study	This study	Wang et al. (2014) [3]	Wang et al. (2014) [3]

Note: ND means not detected, LOD means the limit of detection.

Supplementary Table S5 The first three principal components (factors) and their factor loadings in sediment core from the QY and PM lakes.

Indicator	QY Lake			PM Lake		
	PC 1	PC 2	PC 3	PC 1	PC 2	PC 3
PFAS flux	**0.854**	0.445	0.144	0.299	-0.229	**0.895**
PFAS concentration	0.600	**0.658**	0.029	0.200	**0.802**	0.194
Sediment rate	0.927	0.023	0.212	0.069	-0.699	0.627
Temperature	0.643	0.479	-0.475	-0.162	**0.845**	0.072
Precipitation	-0.136	0.061	**0.926**	-0.676	0.110	**0.524**
TOC	-0.498	**0.686**	0.109	0.320	**0.520**	-0.420
IC	**0.804**	-0.511	0.117	**0.320**	**0.612**	0.577
Ti flux	**0.762**	-0.068	0.249	**0.408**	-0.697	-0.122
Rb/Sr	-0.680	0.430	0.198	-0.099	-0.921	-0.161

Supplementary Table S6 Coefficients (B_i) and mean percent contribution of factor scores in the QY and the PM lakes, as calculated based on PCA and MLRA.

Principal component	Process	Coefficients (B_i)		Mean percent contribution	
		QY Lake	PM Lake	QY Lake	PM Lake
PC1	Glacial meltwater	0.854	0.299	59%	21%
PC2	Lake sediment sequestration	0.445	0.229	31%	16%
PC3	Precipitation	0.144	0.895	10%	63%

Supplementary Table S7 The properties (dry weight, wet weight, water content, sediment density, porosity, IC, and TOC) of the PM Lake sediment core samples.

Depth (cm)	Dry weight (g)	Wet weight (g)	Water content (%)	Sediment density (g cm^{-3})	Porosity (%)	IC (%)	TOC (%)
0.5	11.99	16.86	28.85	0.63	25.64		
1.0	10.78	18.24	40.86	0.57	39.30	2.92	1.67
1.5	11.21	17.5	35.93	0.59	33.15	2.50	0.96
2.0	9.29	18.82	50.65	0.49	50.29	2.73	0.83
2.5	9.72	18.5	47.44	0.51	46.29	2.32	0.70
3.0	9.63	17.53	45.05	0.51	41.64	2.40	0.74
3.5	11.31	17.12	43.11	0.6	30.67	2.35	0.17
4.0	8.72	14.05	37.92	0.46	28.10	2.33	0.83
4.5	8.68	19.65	55.82	0.46	57.83	2.21	0.835
5.0	12.06	22.49	46.39	0.64	55.03	2.60	1.27
5.5	10.37	19.63	47.18	0.55	48.85	2.63	0.54
6.0	13.21	21.36	38.12	0.7	42.93	2.78	0.28
6.5	9.47	13.56	30.15	0.5	21.57	2.35	0.64
7.0	12.85	16.68	22.99	0.68	20.22	2.30	0.61
7.5	12.16	23	47.12	0.64	57.15	2.12	0.70
8.0	11.38	20.21	43.66	0.6	46.53	2.04	0.42
8.5	8.36	14.69	43.06	0.44	33.34	2.09	0.50
9.0	12.57	20.48	38.63	0.66	41.72	2.15	0.57
9.5	11.15	17.76	37.21	0.59	34.86	2.10	0.64
10.0	7.96	18.07	55.93	0.42	53.29	2.18	0.45

Supplementary Table S8 The properties (dry weight, wet weight, water content, sediment density, porosity, IC, and TOC) of the QY Lake sediment core samples.

Depth (cm)	**Dry weight (g)**	**Wet weight (g)**	**Water content (%)**	**Sediment density ($g\ cm^{-3}$)**	**Porosity (%)**	**IC (%)**	**TOC (%)**
1.0	9.40	15.93	40.97	0.30	20.53	2.73	0.93
1.5	11.3	18.61	39.29	0.36	23.00	3.23	0.77
2.0	13.36	22.77	41.34	0.42	29.61	2.77	0.93
2.5	9.55	15.81	39.59	0.3	19.69	3.27	0.87
3.0	11.83	20.42	42.06	0.37	27.02	3.05	0.91
3.5	6.23	10.39	40.06	0.20	13.09	2.56	0.95
4.0	8.83	15.22	42.01	0.28	20.11	2.42	0.98
4.5	13.84	23.82	41.88	0.44	31.38	2.56	0.94
5.0	8.26	14.35	42.44	0.26	19.15	2.44	1.09
5.5	9.46	16.09	41.24	0.30	20.87	3.03	0.91
6.0	11.95	21.21	43.66	0.38	29.13	2.96	0.92
6.5	6.62	11.46	42.24	0.21	15.22	2.42	1.02
7.0	10.91	19.82	44.98	0.34	28.04	2.29	0.95
7.5	10.22	17.76	42.43	0.32	23.70	2.49	0.85
8.0	8.95	14.97	40.23	0.28	18.94	2.66	0.85
8.5	12.18	20.97	41.94	0.38	27.66	2.55	0.93
9.0	11.26	19.75	43.00	0.35	26.71	2.29	0.93
9.5	11.12	20.59	46.00	0.35	29.79	2.01	1.05
10.0	9.58	17.02	43.73	0.30	23.41	2.08	0.99
10.5	16.81	28.46	40.94	0.53	36.65	2.83	0.84
11.0	9.23	15.09	38.82	0.29	18.43	2.94	0.83
11.5	11.25	18.46	39.08	0.35	22.69	2.94	0.81
12.0	11.36	19.08	40.48	0.36	24.29	2.68	0.81

Supplementary Table S9 Recovery standard (RS) mix used for HPLC-MS/MS detection.

analyte	acronym	formula	supplier (purity)	precursor/product ion [m/z]
Perfluoro-n-[1,2,3,4-$^{13}C_4$] butanoic acid	[$^{13}C_4$]-PFBA	[2,3,4-$^{13}C_3$]F_7^{13}COOH	Well. Lab. (>98%)	216.8/171.8
Perfluoro-n-[1,2-13C2] hexanoic acid	[13C2]-PFHxA	C_4F_9[2-13C]F_2^{13}COOH	Well. Lab. (>98%)	314.9/ 269.9
Perfluoro-n-[1,2,3,4-$^{13}C_4$] octanoic acid	[$^{13}C_4$]-PFOA	C_4F_9[2,3,4-$^{13}C_3$]F_6^{13}COOH	Well. Lab. (>98%)	416.9/ 371.8
Perfluoro-1-[1,2,3,4-^{13}C4] octane sulfonate	[$^{13}C_4$]-PFOS	C_4F_9[1,2,3,4-$^{13}C_4$]$F_8SO_2O^-$	Well. Lab. (>98%)	502.9/ 79.5
Perfluoro-n-[1,2,3,4,5-$^{13}C_5$] nonanoic acid	[$^{13}C_5$]-PFNA	C_4F_9[2,3,4,5-$^{13}C_4$]F_8^{13}COOH	Well. Lab. (>98%)	467.9/ 423.0
Perfluoro-n-[1,2-$^{13}C_2$] decanoic acid	[$^{13}C_2$]-PFDA	$C_8F_{17}{}^{13}CF_2{}^{13}COOH$	Well. Lab. (>98%)	514.9/ 469.8
Perfluoro-n-[1,2-$^{13}C_2$] undecanoic acid	[$^{13}C_2$]-PFUnA	$C_9F_{19}{}^{13}CF_2{}^{13}COOH$	Well. Lab. (>98%)	564.9/ 519.8

Supplementary Table S10 Target analysts of this study

Analyte	Acronym	Formula	Supplier (purity)	Precursor/product ion [m/z]
Perfluorobutane sulfonic acid	PFBS	$C_4F_9SO_2O^-$	Fluka (97%)	298.9/ 79.8
Perfluorohexane sulfonic acid	PFHxS	$C_6F_{13}SO_2O^-$	Fluka (98%)	398.9/ 79.8
Perfluorooctane sulfonic acid	PFOS	$C_8F_{17}SO_2O^-$	Well. Lab.[a] (>98%)	498.9/ 79.7
Perfluorobutanoic acid	PFBA	C_3F_7COOH	ABCR (99%)	112.9/ 168.7
Perfluoropentanoic acid	PFPeA	C_4F_9COOH	Alfa Aesar (98%)	262.8/ 218.9
Perfluorohexanoic acid	PFHxA	$C_5F_{11}COOH$	Fluka (97%)	312.9/ 268.8
Perfluoroheptanoic acid	PFHpA	$C_6F_{13}COOH$	Lanc. Syn.[b] (98%)	362.9/ 318.9
Perfluorooctanoic acid	PFOA	$C_7F_{15}COOH$	Lanc. Syn.[b] (95%)	412.9/ 368.9
Perfluorononanoic acid	PFNA	$C_8F_{17}COOH$	Lanc. Syn.[b] (97%)	462.9/ 418.9
Perfluorodecanoic acid	PFDA	$C_9F_{19}COOH$	Lanc. Syn.[b] (97%)	512.9/ 469.0
Perfluoroundecanoic acid	PFUnA	$C_{10}F_{21}COOH$	ABCR (96%)	562.9/ 519.0
Perfluorododecanoic acid	PFDoA	$C_{11}F_{23}COOH$	Alfa Aesar (96%)	612.9/ 568.9
Perfluorotridecanoic acid	PFTrDA	$C_{12}F_{25}COOH$	Well. Lab.[a] (>98%)	663.1/ 618.9

a: Well. Lab. Wellington Laboratories. b: Lanc. Syn. Lancaster Synthesis.

Supplementary Table S11 Concentrations of PFAS in the procedural blanks and the LOD in this study.

Analytes	Procedural blanks (n=5)	LOD ($pg\ g^{-1}$ dw)
PFBA	11.8±1.7	16.9
PFPeA	3.8±0.7	6.0
PFHxA	2.0±0.6	3.9
PFHpA	0.8±0.4	2.0
PFOA	1.4±0.5	2.9
PFNA	2.6±1.4	6.7
PFDA	1.4±0.8	3.8
PFUdA	2.8±1.6	7.6
PFDoA	2.4±1.0	5.5
PFTrDA	1.6±0.8	4.0
PFBS	8.0±1.9	13.7
PFHxS	2.6±1.4	6.7
PFOS	9.4±1.9	15.0

Supplementary Table S12 Statistics of sample recoveries.

Mass labelled Compounds	Max	Min	Mean	SD
[$^{13}C_4$]-PFBA	108%	44%	70%	15%
[$^{13}C_2$]-PFHxA	86%	31%	57%	14%
[$^{13}C_4$]-PFOA	85%	25%	51%	15%
[$^{13}C_4$]-PFOS	79%	34%	55%	9%
[$^{13}C_5$]-PFNA	80%	28%	50%	13%
[$^{13}C_2$]-PFDA	73%	20%	43%	13%
[$^{13}C_2$]-PFUnA	66%	15%	42%	13%

Supplementary Table S13 The results of ^{210}Pb calculations for the PM Lake, using the CRS dating model.

Depth	Drymass	Chronology			Sedimentation Rate	
		Date	Age			
cm	g cm^{-2}	AD	year	±	g cm^{-2} y^{-1}	cm y^{-1}
0.5	0.23	2013	2	0	0.09	0.14
1.0	0.32	2011	5	0	0.07	0.14
1.5	0.28	2008	7	1	0.07	0.13
2.0	0.3	2004	11	1	0.08	0.13
2.5	0.24	2000	15	1	0.09	0.17
3.0	0.26	1997	18	2	0.07	0.17
3.5	0.25	1994	21	2	0.08	0.13
4.0	0.3	1990	25	2	0.1	0.13
4.5	0.23	1986	29	3	0.1	0.21
5.0	0.23	1983	32	3	0.1	0.21
5.5	0.32	1980	35	3	0.09	0.16
6.0	0.27	1978	37	4	0.22	0.16
6.5	0.35	1977	38	3	0.16	0.32
7.0	0.25	1973	42	4	0.09	0.32
7.5	0.34	1969	46	4	0.08	0.13
8.0	0.32	1967	48	4	0.19	0.13
8.5	0.3	1966	49	4	0.14	0.32
9.0	0.22	1963	52	5	0.14	0.32
9.5	0.33	1961	54	4	0.12	0.21
10.0	0.29	1958	57	6	0.07	0.21

Supplementary Table S14 The results of ^{210}Pb calculations for the QY Lake, using the CRS dating model.

Depth	Drymass	Chronology			Sedimentation Rate	
		Date	Age			
cm	g cm^{-2}	AD	year	±	g cm^{-2} y^{-1}	cm y^{-1}
0.25	0.29	2014	0	0	0.12	0.2
0.75	0.23	2011	3	0	0.09	0.19
1.25	0.15	2009	5	0	0.09	0.29
1.75	0.18	2007	7	1	0.11	0.3
2.25	0.21	2005	9	1	0.09	0.21
2.75	0.15	2003	11	1	0.12	0.41
3.25	0.19	2002	12	2	0.12	0.33
3.75	0.1	2000	14	2	0.07	0.37
4.25	0.14	1999	15	2	0.09	0.31
4.75	0.22	1997	17	3	0.08	0.19
5.25	0.13	1994	20	3	0.07	0.26
5.75	0.15	1992	22	3	0.08	0.28
6.25	0.19	1990	24	4	0.08	0.2
6.75	0.1	1988	26	3	0.06	0.29
7.25	0.17	1986	28	4	0.06	0.18
7.75	0.16	1983	31	4	0.08	0.25
8.25	0.14	1981	33	4	0.08	0.27
8.75	0.19	1979	35	4	0.07	0.17
9.25	0.18	1976	38	5	0.06	0.18
9.75	0.17	1973	41	4	0.05	0.15
10.25	0.15	1970	44	6	0.06	0.19
10.75	0.26	1967	47	5	0.07	0.14
11.25	0.15	1963	51	6	0.08	0.28
11.75	0.18	1961	53	6	0.06	0.16
12.25	0.18	1958	56	7	0.07	0.18

Supplementary Table S15 Equations for calculating transfer rate coefficient (D) (mol $h^{-1}\cdot Pa^{-1}$)

Process	Equations
Air-water	
Diffusion	$D_{\text{air to water}} = D_{\text{water to air}} = 1/[1/(K_{12}\times A_2\times Z_{11})+1/(K_{21}\times A_2\times Z_{22})]$
Wet deposition	$D_{\text{wet deposition}} = A_2\times K_w\times S_c\times X_{13}\times Z_{13}$
Dry deposition	$D_{\text{dry deposition}} = A_2\times K_d\times X_{13}\times Z_{13}$
Water-sediment	
Diffusion	$D_{\text{water to sediment}} = D_{\text{sediment to water}} = 1/[1/(K_{23}\times A_2\times Z_{22})+L_3/(B_3\times A_2\times Z_{22})]$
Deposition	$D_{\text{particulate deposition}} = A_2\times K_s\times Z_{23}$
Sediment	
Resuspension	$D_{\text{resuspension}} = A_3\times K_r\times Z_{33}$
Burial	$D_{\text{sediment burial}} = A_3\times K_b\times Z_{33}$
Reaction	
Degradation in air	$D_{\text{air degredation}} = U_1\times A_1\times h_1\times Z_1$
Degradation in water	$D_{\text{water degredation}} = U_2\times A_2\times h_2\times (Z_{22}+Z_{23})$
Degradation in sediment	$D_{\text{sediment degredation}} = U_3\times A_3\times h_3\times Z_3$
Advection	
Advective air flows	$D_{\text{air out}} = Q_{\text{air out}}\times Z_1$
Advective water flows	$D_{\text{water out}} = Q_{\text{water out}}\times Z_2$

Note: 1, 2, 3 represents air, water (liquid), and sediment (solid), respectively. 0 means the outside of this system. The meanings of other parameters are given in Supplementary Supplementary Table S17.

Supplementary Table S16 Equations for calculating fugacity capacity (Z) ($mol\ m^{-3}\cdot Pa^{-1}$)

Phase	Sub-phase	Equations
Air		$Z_1=(1-X_{13})\times Z_{11}+X_{13}\times Z_{13}$
	Air	$Z_{11}=1/RT$
	Particle	$Z_{13}=Z_{11}\times O_{13}\times K_{oa}\times \rho_{13}$
Water		$Z_2=(1-X_{23})\times Z_{22}+X_{23}\times Z_{23}$
	Water	$Z_{22}=1/H$
	Suspended solids	$Z_{23}=O_{23}\times K_{oc}\times \rho_{23}\times Z_{22}$
Sediment		$Z_3=(1-X_{33})\times Z_{32}+X_{33}\times Z_{33}$
	Pore water	$Z_{32}=1/H$
	Solids	$Z_{33}=O_{33}\times K_{oc}\times \rho_{33}\times Z_{32}$

Note: Z_1, Z_2, and Z_3 are fugacity capacity for air, water and sediment, respectively. Z_{11}, Z_{13}, Z_{22}, Z_{23}, Z_{32}, and Z_{33} are fugacity capacity for sub-phases in air, water, and sediment, respectively. The meanings of other parameters are given in Supplementary Supplementary Table S17.

Supplementary Table S17 Parameters for the QWASI fugacity model design for the glacial rivers

Parameters	Meaning	Value	Unit	Reference
$Q_{air\ in}$	Air advection flow into the lake	1.13×10^{10}	$m^3\ h^{-1}$	Xu et al. (2013) [4]
$Q_{air\ out}$	Air advection flow out of the lake	1.13×10^{10}	$m^3\ h^{-1}$	
$Q_{water\ in}$	Flow rate of glacial meltwater into the lake	7.42×10^{4}	$m^3\ h^{-1}$	Ju et al. (2008) [5]
$Q_{water\ out}$	Water advection flow out of the lake	6.68×10^{4}	$m^3\ h^{-1}$	This study
$C_{air\ in}$	PFAS concentration in air advection flow	[a]	$mol\ m^{-3}$	This study
$C_{water\ in}$	PFAS concentration in the estuary of the pre-lake river	[a]	$mol\ m^{-3}$	This study
K_{12}	Air-side molecular transfer coefficient over water	3	$m\ h^{-1}$	
K_{21}	Water-side molecular transfer coefficient over air	0.03	$m\ h^{-1}$	Thibodeaux (1996) [6] and Herrera (1977) [7]
K_{23}	Water-side molecular transfer coefficient over sediment	0.01	$m\ h^{-1}$	
K_w	Wet deposition velocity	4.05×10^{-5}	$m\ h^{-1}$	Ju et al. (2008) [5]
K_d	Dry deposition velocity	5.69×10^{-1}	$m\ h^{-1}$	Mackay and Paterson (1991) [8]
S_c	Rain scavenging rate	2×10^{5}	$m\ h^{-1}$	Mackay et al. (1986) [9]
A_1, A_2, A_3	Interface areas of air, water, and sediment	6×10^{6}	m^2	This study (based on measure)
X_{13}	Volume fractions of solids in air	9.84×10^{-11}	$v\ v^{-1}$	Mackay and Paterson (1991) [8]

Parameters	Meaning	Value	Unit	Reference
X_{23}	Volume fractions of solids in water	4.29×10^{-6}	v v^{-1}	
X_{33}	Volume fractions of solids in sediment	0.5	v v^{-1}	Mackay and Paterson (1991) [8]
L_3	Diffusion path lengths in sediment	0.025	m	This study (based on measure)
B_3	Molecular diffusivities in sediment	2.49×10^{-6}	m^2 h^{-1}	Xu et al. (2013) [4]
K_s	Water sedimentation rate	4.60×10^{-6}	m h^{-1}	Chen et al. (2006) [10] and Hu et al. (1998) [11]
K_r	Sediment resuspension rate	1.10×10^{-8}	m h^{-1}	Parnis and Mackay (2020) [12]
K_b	Sediment burial rate	6.00×10^{-7}	m h^{-1}	
U_1, U_2, U_3	Degradation rate of PFAS in air, water, and sediment	1.14×10^{-7}	h^{-1}	Russel et al. (2008) [13] and Washington et al. (2009) [14]
h_1	Thickness of air	7.00×10^{2}	m	Xu et al. (2013) [4]
h_2	Thickness of water	1.5	m	This study (based on measure)
h_3	Thickness of sediment	0.2	m	This study (based on measure)
ρ_{13}	Densities of solids in air	1.5	t m^{-3}	Mackay and Paterson (1991) [8]
ρ_{23}	Densities of solids in water	1.8	t m^{-3}	This study

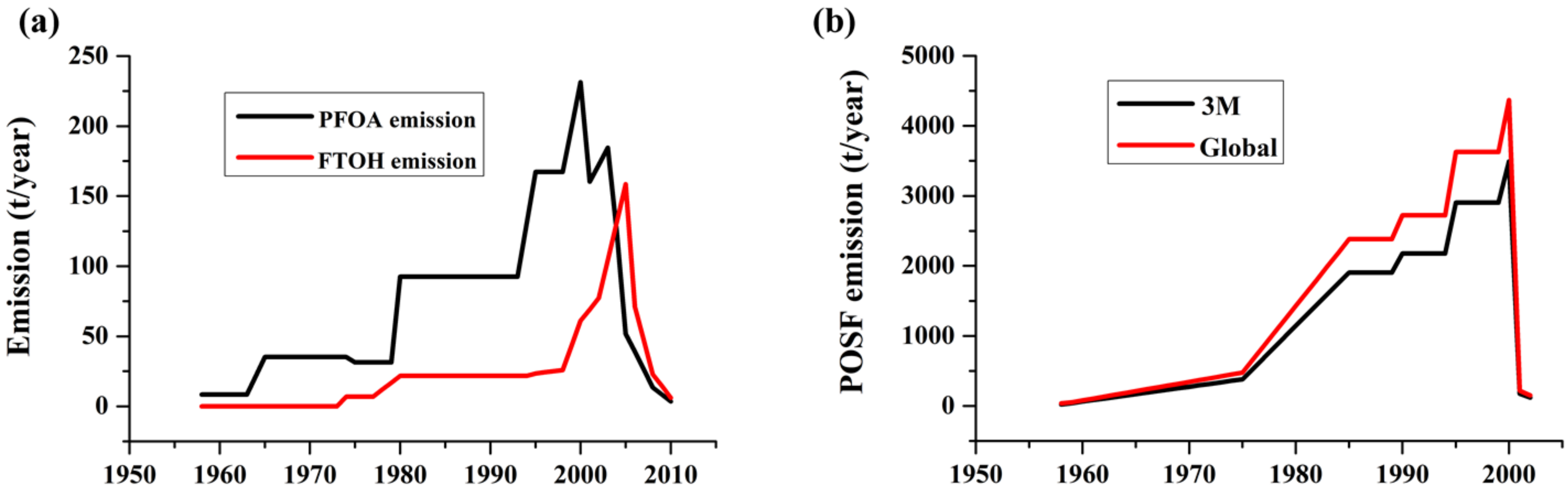
Parameters	Meaning	Value	Unit	Reference
				(based on measurement)
ρ_{33}	Densities of solids in sediment	2.49	t m^{-3}	Zhou et al. (2022) [15]
R	Gas constant	8.314	Pa m^3 mol^{-1} K^{-1}	Mackay et al. (1997) [16]
T	Local average temperature	275.15	K	This study (based on measure)
H	Henry's constant	[a]	Pa m^3 mol^{-1}	EPA Chemistry Dashboard
K_{oa}	Adsorption coefficient between water and air	[a]	—	EPA Chemistry Dashboard
K_{oc}	Adsorption coefficient between water and sediment	[a]	—	This study (based on measure)
O_{13}	Content of organic carbon in solids in air	0.2	%	Xu et al. (2013) [4]
O_{23}	Content of organic carbon in solids in water	0.441	%	
O_{33}	Content of organic carbon in solids in sediment	1.00	%	This study (based on measure)

Note: [a] data presented in Supplementary Table S18. EPA Chemistry Dashboard is available at https://comptox.epa.gov/dashboard/.

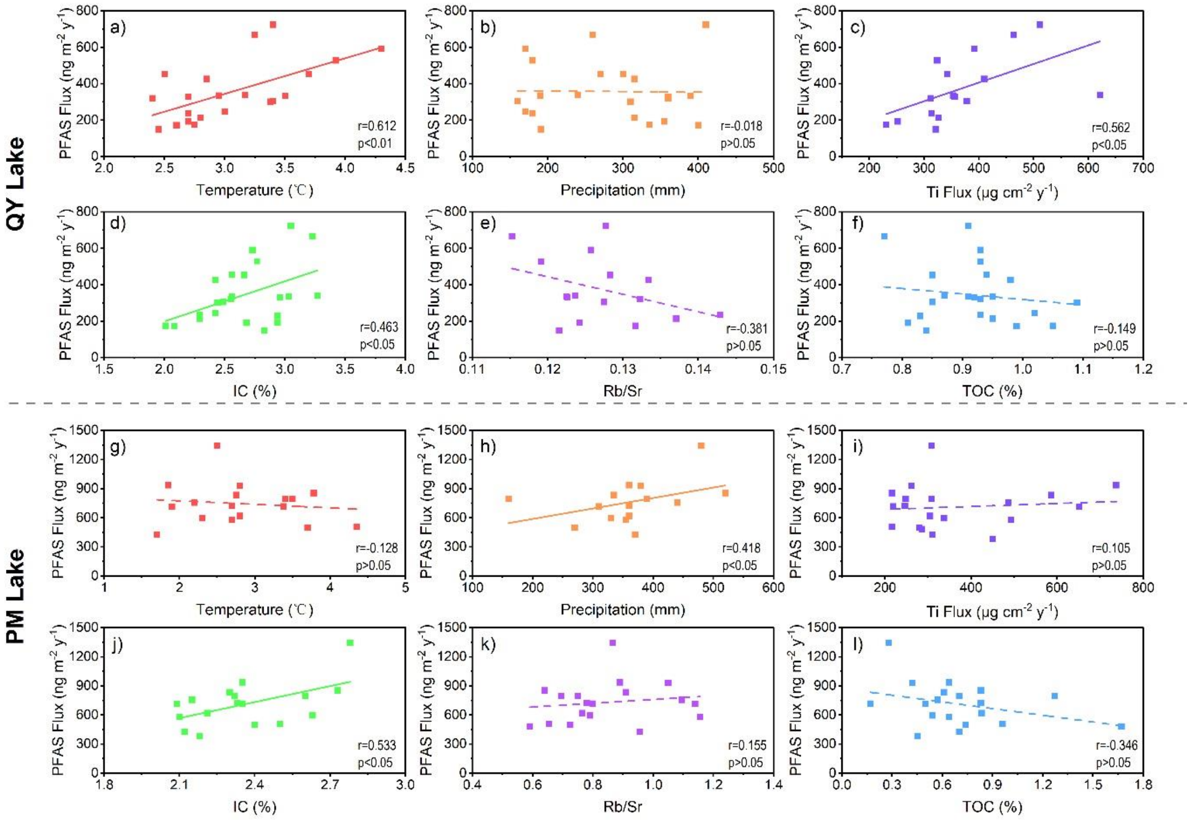
Supplementary Table S18 Parameter values for individual PFAS in the QWASI fugacity model

Parameter	PFBA	PFPeA	PFHxA	PFHpA	PFOA	PFNA	PFDA	PFUdA	PFDoA	PFTrDA	PFBS	PFHxS	PFOS
Molecular weight ($g\ mol^{-1}$)	214.0	264.1	314.1	364.1	414.1	464.1	514.1	564.1	614.1	664.1	300.1	400.1	500.1
$C_{air\ in}$ ($pg\ m^{-3}$)	1.48	0.80	1.14	0.84	2.62	0.69	0.59	0.15	0.04	0.21	0.43	0.38	5.00
$C_{water\ diversion}$ ($pg\ L^{-1}$)	344.1	74.6	46.8	65.4	49.6	24.7	5.6	3.3	4.0	0.5	36.8	7.3	16.4
H	5.0×10^{-5}	3.0×10^{-10}	2.4×10^{-10}	2.1×10^{-10}	1.9×10^{-10}	1.2×10^{-9}	1.5×10^{-10}	3.3×10^{-10}	3.4×10^{-10}	3.5×10^{-10}	3.0×10^{-10}	1.9×10^{-10}	1.8×10^{-11}
K_{oa}	2.9×10^{3}	5.8×10^{3}	6.8×10^{3}	1.2×10^{4}	1.4×10^{4}	1.6×10^{4}	1.9×10^{4}	5.0×10^{4}	5.6×10^{4}	9.1×10^{4}	1.4×10^{4}	1.9×10^{4}	5.6×10^{4}
K_{oc}	5.7×10^{2}	6.2×10^{2}	3.0×10^{2}	4.4×10^{2}	2.7×10^{2}	1.1×10^{3}	1.1×10^{4}	1.4×10^{4}	1.1×10^{4}	1.1×10^{4}	2.7×10^{2}	2.8×10^{2}	4.8×10^{3}

Supplementary Figure

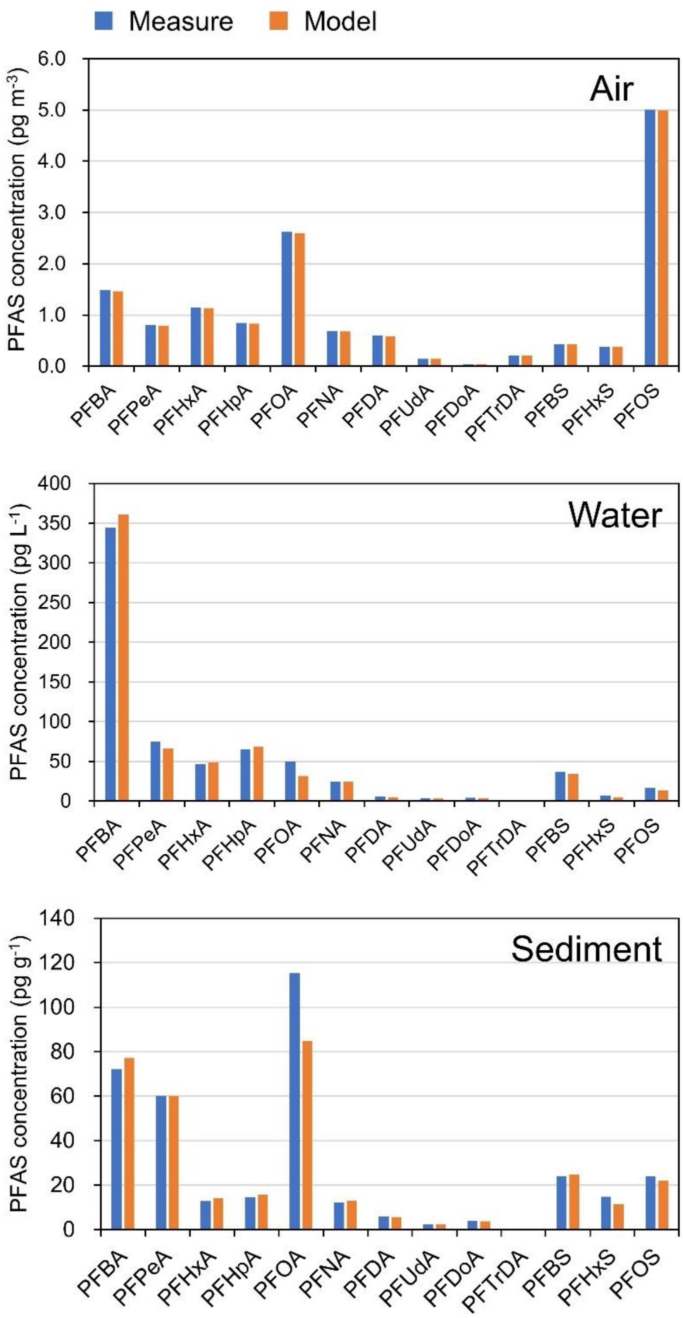


Supplementary Fig. S1 Emissions for PFOA, FTOH, and PFOSF-based substances: (a) global emission of PFOA (from direct manufacture and use and unintentional residuals) and residual FTOH; (b) global emission of PFOSF and that released by the 3M company.

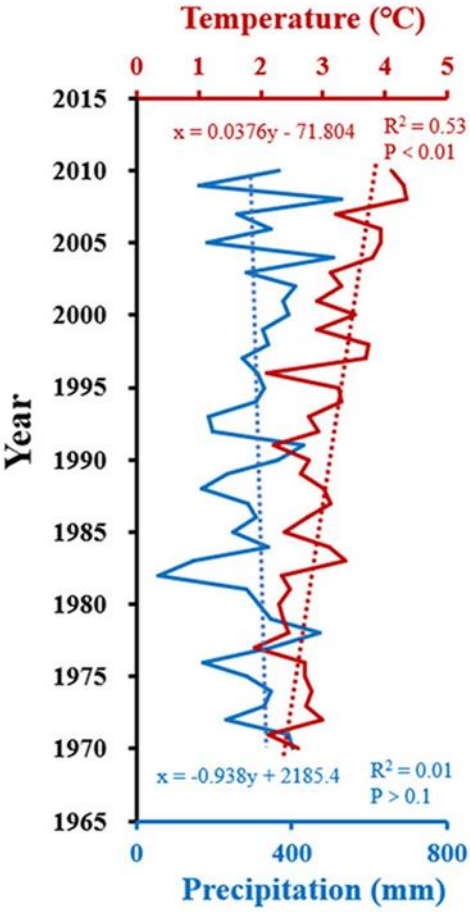


Supplementary Fig. S2 Correlation between PFAS flux with climate indicators (temperature, and precipitation) and geochemical indicators (Ti flux, IC, TOC, and Rb/Sr): a-f) for the QY lake; g-l) for the PM lake.

Note: Solid lines represent significant correlations ($p<0.05$), while dashed lines represent insignificant correlations ($p>0.05$).

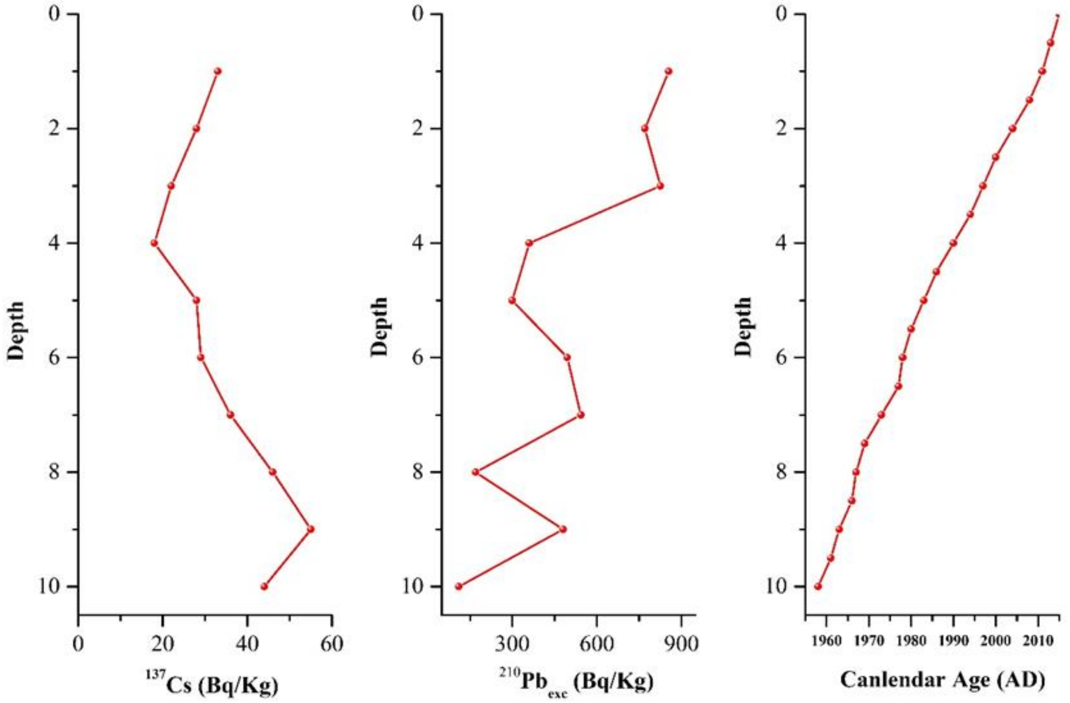


Supplementary Fig. S3 Comparison of measured and modeled concentration for individual PFAS in multimedia of the glacial rivers: a) air; b) water; c) sediment.

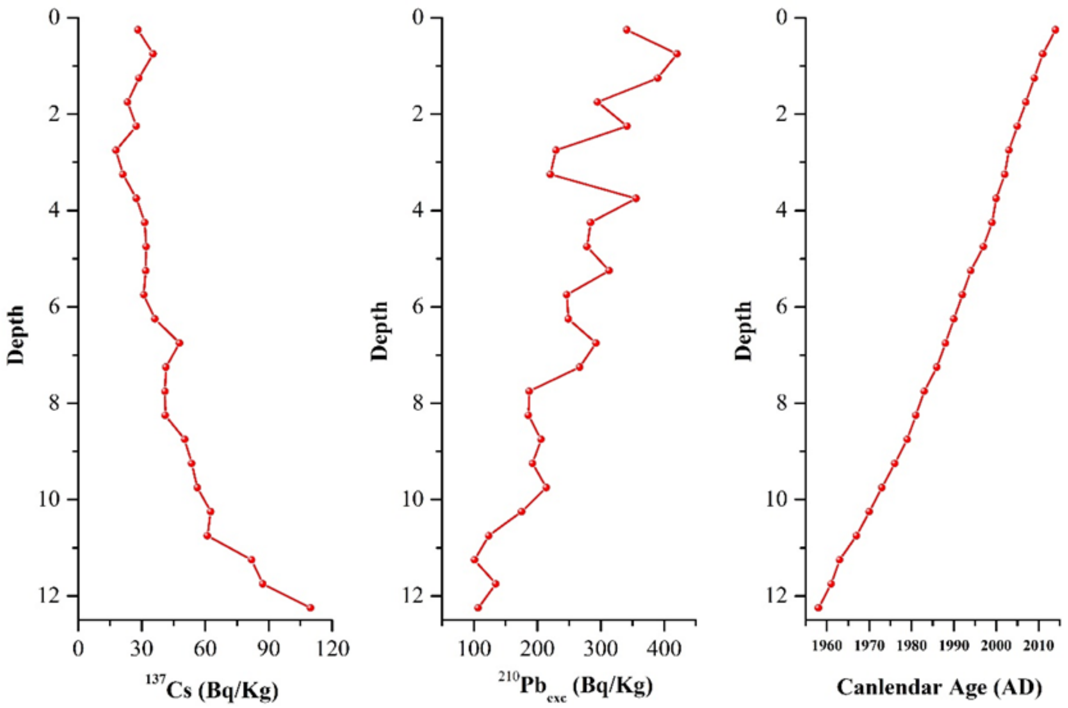


Supplementary Fig. S4 Temperature and precipitation variations during 1970−2010, from Langkazi meteorological station.

Note: the red dotted line is the linear trend of temperature over time, and the blue dotted line represents the linear trend between precipitation and time.



Supplementary Fig. S5 ^{210}Pb and ^{137}Cs activities and depth-age model of the core from the PM lake



Supplementary Fig. S6 ^{210}Pb and ^{137}Cs activities and depth-age model of the core from the QY lake.

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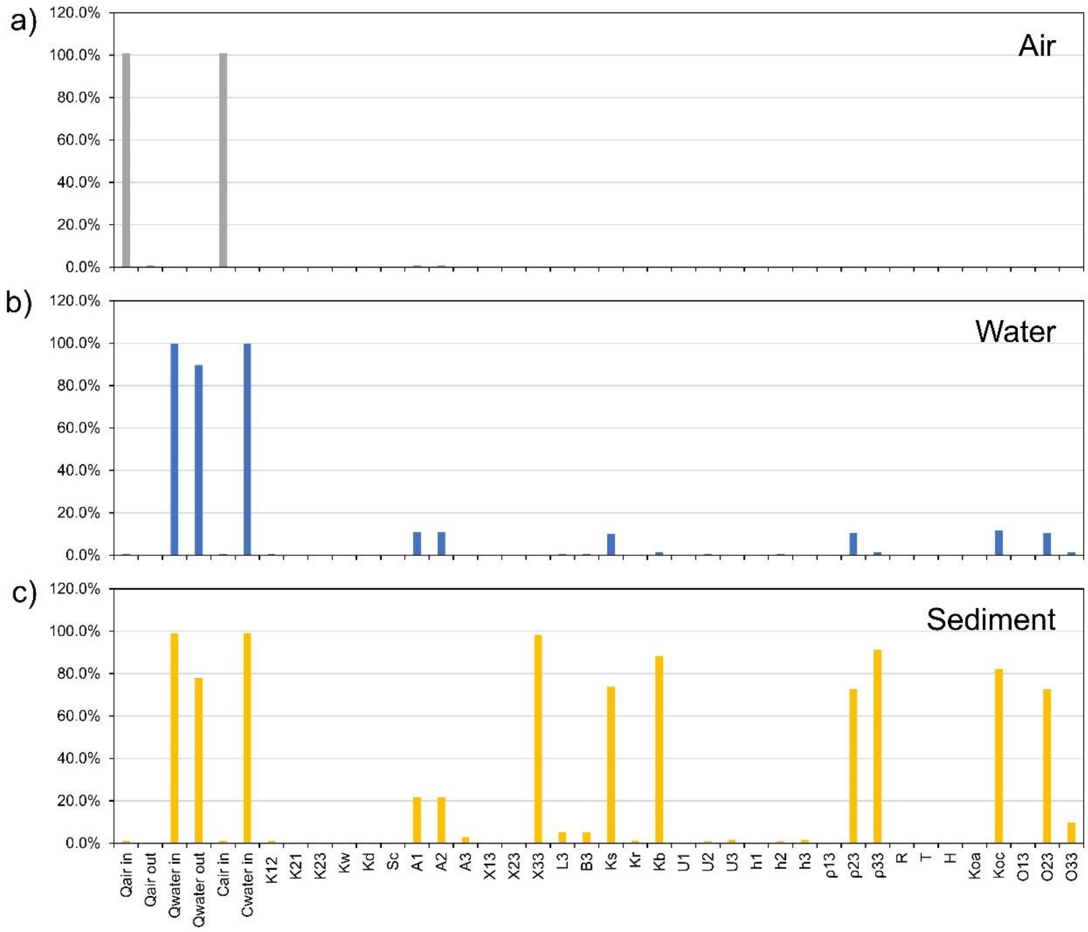

Supplementary Fig. S7 Sensitivity analysis of 40 parameters used in the QWASI fugacity model in the pre-lake rivers of the PM lake for different media: a) air; b) water; and c) sediment.

Supplementary Reference

1. Gao K, Gao Y, Li Y, Fu J, Zhang A. A rapid and fully automatic method for the accurate determination of a wide carbon-chain range of per-and polyfluoroalkyl substances (C4–C18) in human serum. *Journal of Chromatography A* **1471**, 1-10 (2016).

2. Gao K*, et al.* Prenatal exposure to per-and polyfluoroalkyl substances (PFASs) and association between the placental transfer efficiencies and dissociation constant of serum proteins–PFAS complexes. *Environmental Science & Technology* **53**, 6529-6538 (2019).

3. Wang X*, et al.* Accumulation of perfluoroalkyl compounds in tibetan mountain snow: temporal patterns from 1980 to 2010. *Environmental Science & Technology* **48**, 173-181 (2014).

4. Xu F-L*, et al.* Multimedia fate modeling of polycyclic aromatic hydrocarbons (PAHs) in Lake Small Baiyangdian, Northern China. *Ecological Modelling* **252**, 246-257 (2013).

5. Ju J*, et al.* Composition, spatial distribution and environmental significance of water ions in lake Pumayun Co and its catchment, Southern Tibet. *Journal of Lake Science* **20**, 591-599 (2008).

6. Thibodeaux LJ. *Environmental chemodynamics: Movement of chemicals in air, water, and soil*. John Wiley & Sons (1996).

7. Banks RB, Herrera FF. Effect of wind and rain on surface reaeration. *Journal of the Environmental Engineering Division* **103**, 489-504 (1977).

8. Mackay D, Paterson S. Evaluating the multimedia fate of organic chemicals: a level III fugacity model. *Environmental Science & Technology* **25**, 427-436 (1991).

9. Mackay D, Paterson S, Schroeder WH. Model describing the rates of transfer processes of organic chemicals between atmosphere and water. *Environmental Science & Technology* **20**, 810-816 (1986).

10. Chen J, Zhou W, Deng A, Sun G. Formation and evolution of the longitudinal profile of the Lower Yellow River in modern times. *Sediment Study* **2**, 1-8 (2006).

11. Hu C, Ji Z, Huang Y, Chen D. Analysis on dredging practice in rivers, lakes and reservoirs. *J Sed Res* **12**, 47-55 (1998).

12. Parnis JM, Mackay D. *Multimedia environmental models: the fugacity approach*. CRC press (2020).

13. Russell MH, Berti WR, Szostek B, Buck RC. Investigation of the biodegradation potential of a fluoroacrylate polymer product in aerobic soils. *Environmental Science & Technology* **42**, 800-807 (2008).

14. Washington JW, Ellington JJ, Jenkins TM, Evans JJ, Yoo H, Hafner SC. Degradability of an acrylate-linked, fluorotelomer polymer in soil. *Environmental Science & Technology* **43**, 6617-6623 (2009).

15. Zhou Y*, et al.* Optimizing the fugacity model to select appropriate remediation pathways for perfluoroalkyl substances (PFASs) in a lake. *Journal of Hazardous Materials* **438**, 129558 (2022).

16. Mackay D, Shiu WY, Ma K-C. *Illustrated handbook of physical-chemical properties of environmental fate for organic chemicals*. CRC press (1997).