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1 **Supplementary Material**

2

3 **Perfluoroalkyl Substances in the Yangtze River: Changing Exposure and Its** 4 **Implications after Operation of the Three Gorges Dam**

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58

59 Text S1 Water and sediment extraction procedures

60 Before analysis, the sediments were freeze-dried, sieved through a 0.5 mm pore size sieve and then kept
61 at -20 °C in the dark until extraction. For water sample pretreatment, 5 L water samples were filtered
62 through glass fiber filters (Whatman GF/F, 0.7 µm, UK) to remove suspended particles. Then 2 L filtrate
63 was spiked with 100 µL internal standard to reach a final concentration of 50 ng/L each. The spiked
64 filtrate was concentrated using solid-phase extraction (SPE) method through HLB SPE cartridges
65 (Waters, 6 mL, 500 mg, USA). The SPE cartridges were preconditioned with 10 mL methanol and 10
66 mL Milli-Q water, and then water samples were introduced to the cartridges at a flow rate of 5-10
67 mL/min. After loading of the water samples, the cartridges were rinsed with 10 mL of Milli-Q water to
68 remove weakly bound impurities. PFASs retained on the cartridges were eluted with 10 mL methanol
69 and then the eluate was evaporated to near dryness under a gentle stream of nitrogen and re-dissolved
70 in 1 mL methanol. The final extract was centrifuged, and then the supernatant transferred to a 2 mL
71 amber sample vial and stored at -20 °C until analysis. The extraction method for sediment is as follows
72 (Higgins et al., 2005): 2 g of each freeze-dried sediment sample were placed into a 50 mL polypropylene
73 (PP) tube, and 100 µL internal standard added until a final concentration of 25 µg/kg was reached. Then
74 the samples were mixed and placed in a refrigerator at 4 °C overnight. Ten milliliter of 1% acetic acid
75 solution was added to each tube, and the contents then vortexed, placed in a preheated sonication bath,
76 and sonicated for 15 min. Next, the tube was centrifuged at 3000 rpm for 2 min, and the acetic acid
77 solution decanted into a second 50 mL PP tube. An aliquot of the methanol/acetic acid (9:1) extraction
78 solvent mixture (5 mL) was then added to the original tube, and the contents again vortexed and
79 sonicated for 15 min at 60 °C before centrifuging and decanting the extract. This process of acetic acid
80 washing followed by methanol/acetic acid extraction was repeated one more time, after which a final
81 washing was performed with 10 mL acetic acid. For each sample, all washes and extracts were combined
82 together in a conical flask and the resulting mixture diluted to 300 mL using Milli-Q water to ensure
83 that the concentration of organic solvent in the solution was less than 5%. The procedure that then

84 followed was the same as for the water sample pretreatment described above.

85

86 Text S2 Ecological risk assessment

87 Ecological risk of residual PFASs in the aquatic environment was assessed by means of a risk quotient
88 (RQ) (Lin et al., 2010) at three different trophic levels (algae, daphnids, and fish). The RQ was
89 calculated as the ratio between the measured environmental concentration (MEC) and the predicted
90 no-effect concentration (PNEC, Equation 1) whose value is obtained from chronic toxicity data
91 (median effective concentration, EC50 or median lethal concentration, LC50) divided by an
92 assessment factor (AF) of 1000 (Garrido et al., 2016). When calculating RQ, the lowest reported
93 values of EC50 or LC50 of each trophic level were used. A second approach was to calculate the
94 mixture risk quotient ($MRQ_{MEC/PNEC}$, Equation 2) formed as the sum of all individual RQs at each
95 trophic level (Zhao et al., 2010). Toxicity data (EC50 or LC50) were collected from the ECOTOX
96 database provided by the USEPA ECOTOX (USEPA). If not already available in ECOTOX, the
97 lowest values of EC50 or LC50 were obtained using EPI Suite (EPIWEB v.4.1) (USEPA EPI Suite).
98 Table S7 presents the chronic toxicity data of each compound for selected algae, daphnids, and fish.
99 The levels of risk are divided into four categories, i.e., no risk ($RQ < 0.01$), low risk ($0.01 \leq RQ <$
100 0.1), medium risk ($0.1 \leq RQ < 1$), and high risk ($RQ \geq 1$) (Wang et al., 2018).

$$101 \quad RQ_i = \frac{MEC_i}{PNEC_i} \quad (1)$$

$$102 \quad MRQ_{\frac{MEC}{PNEC}} = \sum_{i=1}^{n=11} \frac{MEC_i}{(PNEC_{algae} \text{ or } PNEC_{daphnids} \text{ or } PNEC_{fish})} \quad (2)$$

103

104 Text S3 Models for calculation of K_d for different scenarios

105 The partition coefficient (K_d) was defined as the ratio between PFASs concentration bound to
106 sediment C_s and that in surface water C_w (shown in Equation 3). K_d was further normalized to the
107 organic carbon content of the sediment (f_{oc}) at each site to obtain the value of K_{oc} (shown in Equation

108 4) (Ogbeide et al., 2018):

109
$$K_d = C_s/C_w \quad (3)$$

110
$$K_{oc} = K_d/f_{oc} \quad (4)$$

111 In general, K_{oc} is proportional to K_{ow} ((Table S9) (Equation 5) (Karickhoff et al., 1979). Also, the
112 organic carbon content of the sediment (f_{oc}) is inversely proportional to grain size (α) as sediment
113 adsorption capacity decreases with strong channel erosion after TGD thickens the grain size (Equation
114 6) (Yang et al., 2017).

115
$$\text{Log } K_{oc} = \beta \times \text{Log } K_{ow} + b$$

116
$$(5)$$

117
$$f_{oc} \times \alpha = K \quad (6)$$

118 in which, β , b and K are constants. A proportional relationship between K_{ow} and $\alpha \times K_d$ can be described
119 by Equation 7 through calculating and simplifying Equation 3-6.

120
$$\text{Log}(\alpha K_d) = \beta \times \text{Log} K_{ow} + b \quad (7)$$

121 in which, c is a constant and $c=b+\text{Log}K$. Model parameters were estimated using measured data
122 collected from the Yangtze River in 2014. The resulting expression is

123
$$\text{Log}(\alpha K_d) = 0.374 \times \text{Log} K_{ow} - 0.039 \quad (R^2=0.857, p=0.024) \quad (8)$$

124 We also collected information from the literature on the mean grain size of sediment at different
125 hydrological sites in the Yangtze River (Table S10). Values of K_d were calculated for different
126 scenarios (Table S11). Assuming that the total loads of PFASs were constant in all the scenarios
127 considered, then C_w for scenario 1 and scenario 3 were calculated by Equation 9:

128
$$C_w = \frac{C_{w,2014} \times H_1 + C_{s,2014} \times H_2 \times \rho \times 1000}{H_1 + K_d \times H_2 \times \rho / 1000} \quad (9)$$

129 in which, $C_{w,2014}$ is the aqueous PFASs concentration in 2014 (ng/L); $C_{s,2014}$, concentration of PFASs
130 in sediment in 2014 ($\mu\text{g/kg}$); H_1 , mean water depth (Table S10); H_2 , mean depth of the bed sediment
131 layer (about 4.5 m) (Yuan, 2014); S refers to unit plane area of 1 m^2 , and ρ is sediment density of 1500
132 kg/m^3 (Chen et al., 2006). For these scenarios, the calculated value of C_w was used in evaluating risk.

133 Table S1 Information on sampling sites along the Yangtze River

No.	Sampling sites	Abbreviation	Latitude (°)	Longitude (°)	River system	Water level (m) in spring	Water flow (m ³ /s) in spring	Water level (m) in autumn	Water flow (m ³ /s) in autumn
01	Shigu	SG	26.88	99.98	Upper reach	1817.97	380.00	1820.87	2390.00
02	Panzhihua	PZH	26.57	101.7	Upper reach	986.07	468.00	991.96	3170.00
03	Xiluodu	XLD	28.25	103.66	Upper reach	561.9	900	599.23	4500.00
04	Pingshan	PS	28.65	104.17	Upper reach	381.29	1200	381.27	5200.00
05	Xiangjiaba	XJB	28.65	104.38	Upper reach	266.10	1480.00	270.74	5870.00
06	Yibin	YB	28.77	104.65	Upper reach	259.90	2900.00	264.20	10200.00
07	Luzhou	LZ	28.9	105.55	Upper reach	225.39	3300.00	229.32	10900.00
08	Zhutuo	ZT	29.02	105.85	Upper reach	197.81	3640.00	202.02	11400.00
09	Cuntan	CT	29.62	106.60	Upper reach	163.49	4510.00	174.31	13000.00
10	Badong	BD	31.04	110.40	Upper reach	162.43	6150.00	172.21	17000.00
11	Miaohe	MH	30.88	110.90	Upper reach	162.41	6000.00	172.20	18700.00
12	Huanglingmiao	HLM	30.85	111.12	Upper reach	65.24	6060.00	64.65	18800.00
13	Yichang	YC	30.69	111.28	Middle reach	39.37	5960.00	44.21	17200.00
14	Shashi	SS	30.29	112.26	Middle reach	31.69	7090.00	36.54	15600.00
15	Chenglingjilian	CLJL	29.45	113.15	Middle reach	22.53	11800	26.95	22800.00
16	Luoshan	LS	29.67	113.32	Middle reach	21.26	12200.00	26.14	23500.00
17	Wuhan	WH	30.62	114.32	Middle reach	15.73	13500.00	20.15	23200.00
18	Jiujiang	JJ	29.74	116.00	Lower reach	10.31	13400.00	15.25	26200.00
19	Datong	DT	30.78	117.64	Lower reach	6.31	16800.00	10.02	34900.00
20	Wuhu	WHU	31.46	118.34	Lower reach	6.26	17200.00	9.00	35700.00
21	Maanshan	MAS	31.77	118.47	Lower reach	5	19800.00	8.50	36700.00
22	Nanjing	NJ	32.17	118.94	Lower reach	4	20300.00	7.00	37600.00
23	Zhenjiang	ZJ	32.18	119.66	Lower reach	2	20600.00	6.00	38800.00
24	Xuliujing	XLJ	31.77	120.96	Lower reach	1	21000.00	1.00	40000.00
25	Gaochang	GC	28.80	104.42	Minjiang	275.51	1320.00	278.10	4460.00
26	Wusheng	WS	30.35	106.28	Jialingjiang	208.95	95.60	211.07	730.00
27	Beibei	BB	29.81	106.46	Jialingjiang	173.43	457.00	175.34	1530.00
28	Xiaoheba	XHB	30.18	105.84	Jialingjiang	229.72	117.00	229.99	177.00
29	Luoduxi	LDX	30.35	106.58	Jialingjiang	204.61	132.00	204.85	802.00
30	Wulong	WL	29.33	107.76	Wujiang	168.80	284.00	174.06	961.00
31	Xiaoxita	XXT	30.77	111.31	Huangbaihe	79.21	400.00	79.13	600.00
32	Yemingzhu	YMZ	30.74	111.29	Huangbaihe	68.00	500.00	68.00	700.00
33	Chenglingji	CLJ	29.42	113.13	Dongting Lake	22.56	5870.00	26.96	7200.00
34	Nanzui	NZ	29.07	112.29	Dongting Lake	28.63	353.00	28.91	1170.00
35	Zhouwenmiao	ZWM	28.91	112.06	Dongting Lake				
36	Xiangyin	XYIN	28.67	112.87	Dongting Lake				
37	Baihe	BH	32.83	110.11	Hanjiang	171.86	286.00	174.48	1240.00
38	Danjiangkou	DJK	32.51	111.51	Hanjiang	88.61	487.00	88.67	384.00
39	Xiangyang	XY	32.03	112.15	Hanjiang	64.79	477	64.89	656.00
40	Xiantao	XT	30.38	113.45	Hanjiang	23.24	434.00	24.25	672.00
41	Jijiazui	JJZ	30.57	114.23	Hanjiang	88.61	417	88.72	495.00
42	Taocha	TC	32.66	111.66	Hanjiang	138.12	0.00	159.62	0.00
43	Hukou	HK	29.74	116.21	Poyang Lake	9.76	4470.00	14.73	5820.00

134

135

Table S2 Recovery (%), LOD (ng/L or µg/kg) and LOQ (ng/L or µg/kg) for individual PFAS constituents in water and sediment samples

Compound	Water (n=2)						Sediment (n=2)					
	25 ng/L	RSD ^a	50 ng/L	RSD ^a	LOD ^b (ng/L)	LOQ ^c (ng/L)	25 µg/kg	RSD ^a	50 µg/kg	RSD ^a	LOD ^b (µg/kg)	LOQ ^c (µg/kg)
PFHxA	90	1.18	113	9.29	0.08	0.25	76	4.02	88	7.73	0.08	0.30
PFHpA	99	4.30	104	4.34	0.05	0.16	83	2.32	92	7.88	0.02	0.70
PFOA	98	3.84	109	2.93	0.01	0.02	85	4.31	95	7.33	0.03	0.10
PFNA	92	6.81	104	5.97	0.01	0.03	88	5.53	94	5.65	0.01	0.03
PFDA	70	1.27	108	1.57	0.03	0.06	76	15.39	71	6.59	0.02	0.05
PFUnDA	110	10.40	118	18.60	0.01	0.02	42	8.34	32	3.74	0.01	0.02
PFDoA	83	1.47	83	6.94	0.01	0.02	47	13.02	34	3.72	0.01	0.02
PFTeDA	91	3.84	73	2.21	0.01	0.03	63	12.65	31	5.64	0.01	0.03
PFBS	103	2.88	77	3.74	0.02	0.08	94	6.80	106	3.22	0.03	0.10
PFHxS	122	3.83	105	4.28	0.06	0.18	94	7.81	103	4.46	0.05	0.15
PFOS	107	3.62	84	2.36	0.001	0.01	76	15.65	59	18.24	0.02	0.06

^a relative standard deviation (%) (n=2); ^b LOD, limit of detection; ^c LOQ, limit of quantitation.

Table S3 Optimization of UHPLC-MS/MS parameters for multiple reaction monitoring (MRM) acquisition conditions of individual PFAS

Compound	Formula	MS/MS mass transition	Fragmentor (V)	Collision energy (V)	Quantitation referenced mass- labelled internal
PFHxA	CF ₃ (CF ₂) ₄ COO	313-269 ^a /119 ^b	70	3/10	[¹³ C ₆]-PFHxA
PFHpA	CF ₃ (CF ₂) ₅ COO	363-318.9 ^a /169 ^b	75	0/10	[¹³ C ₈]-PFOA
PFOA	CF ₃ (CF ₂) ₆ COO	413-368.9 ^a /168.9 ^b	75	4/16	[¹³ C ₈]-PFOA
PFNA	CF ₃ (CF ₂) ₇ COO	463-418.9 ^a /218.9 ^b	85	3/10	[¹³ C ₉]-PFNA
PFDA	CF ₃ (CF ₂) ₈ COO	513-469 ^a /269 ^b	90	5/15	[¹³ C ₉]-PFNA
PFUnDA	CF ₃ (CF ₂) ₉ COO	563-519 ^a /268.9 ^b	95	5/15/15	[¹³ C ₉]-PFNA
PFDoA	CF ₃ (CF ₂) ₁₀ CO	613-569.1 ^a /369 ^b	95	5/15	[¹³ C ₉]-PFNA
PFTeDA	CF ₃ (CF ₂) ₁₂ CO	713-669.1 ^a /468.9 ^b	110	10/15	[¹³ C ₉]-PFNA
PFBS	CF ₃ (CF ₂) ₃ SO ₃	299-80.1 ^a /99 ^b	130	40/30	[¹³ C ₈]-PFOS
PFOS	CF ₃ (CF ₂) ₇ SO ₃	498.9-80 ^a /99 ^b	167	60/56	[¹³ C ₈]-PFOS
PFHxS	CF ₃ (CF ₂) ₅ SO ₃	399-79.9 ^a /98.7 ^b	160	40/65	[¹³ C ₈]-PFOS
[¹³ C ₆]-PFHxA ^c		319-274 ^a /121 ^b	60	10/20	
[¹³ C ₈]-PFOA ^c		421-375.9	85	4	
[¹³ C ₈]-PFOS ^c		506.9-80	192	60	
[¹³ C ₉]-PFNA ^c		472-427 ^a /223 ^b	82	10/16	

^a The product ion used for quantification; ^b The product ion used for identification; ^c Internal standard.

48 Table S4 Summary of median and range (ng/L) of PFASs in water reported in the published literature

Type	Location	PFOA	PFHxA	PFBS	PFHpA	PFHxS	PFNA	PFDA	PFOS	PFUnDA	PFDoA	PFTeDA	ΣPFASs	Reference
Min-Max (Mean, Median)														
Youngsan and Nakdong River	Korea	1.30-28.00	0.81-17.00	1.00-15.00	1.60-110.00	0.83-17.00	0.45-7.70	0.17-9.60	0.38-68.00	0.32-5.20	0.10-2.80	NA	12.00-190.00 (13 PFASs)	Hong et al., 2013
River and canal	Singapore	5.40-38.20	NA	NA	ND-14.40	ND-67.80	0.90-78.30	0.70-28.20	1.30-156.20	0.20-3.60	ND-1.00	NA	17.60-301.50 (13 PFASs)	Nguyen et al., 2011
Rain	Singapore	0.60-1.30 (-, 1.00)	NA	NA	ND-0.80 (-, 0.40)	ND-1.70 (-, 1.10)	ND-0.90 (-, 0.40)	ND-0.80 (-, 0.40)	1.10-7.90 (-, 1.60)	0.40-0.90 (-, 0.40)	0.10-0.40 (-, 0.10)	NA	5.20-11.50 (-, 6.40) (13 PFASs)	Nguyen et al., 2011
Surface water, Ganges River	India	0.08-1.18	0.37-4.73	ND-10.20	0.34-3.27	ND-0.30	ND-0.19	ND-0.19	ND-1.73	NA	ND	NA	1.30-15.90 (9 PFASs)	Sharma et al., 2016
Water near waste recycling and disposal sites	Vietnam	ND-100.00	ND-77.00	ND-16.00	ND-88.00	ND-30.00	ND-100.00	ND-5.10	ND-11.00	ND-20.00	NA	NA	ND-360.00 (13 PFASs)	Kim et al., 2013
Taihu lake		20.00-23.00	9.40-10.00				1.10-1.20	0.42-0.57	2.00-2.80				7.7-7.6 (5 PFASs)	Lu et al., 2015
Resevior		0.47-7.90	0.12-0.49				0.11-0.41	0.05-0.10	ND-0.69				0.99-11 (5 PFASs)	Lu et al., 2015
River		0.64-57.00	0.15-31.00				0.09-1.50	ND-0.73	ND-5.00				1.3-97 (5 PFASs)	Lu et al., 2015
Surface water, Pearl river	China	0.14-26.48							ND-320.50				0.14-346.72 (18 PFASs)	Pan et al., 2014
Yangtze River	China Chongqing	4.10-35.00	ND-2.20	ND-3.40	ND-4.10	ND-<0.67	ND-3.10	ND-0.57	<0.01-14	ND-0.45	NA	NA		So et al., 2007
Yangtze River	Wuhan, China	2.80-5.60	ND	ND	ND	ND	ND	ND	2.30-25.50	ND	ND	ND		Jin et al., 2006
Yangtze River	China	ND	ND	ND	ND	ND	ND	ND	36.30-703.30	ND	ND	ND		Pan and You, 2010

49 ND: not detected; NA: not available.

50

51 Table S5 Summary of median and range (µg/kg) of PFASs in sediments reported in the published literature

Location	Country	PFOA	PFHxA	PFBS	PFHpA	PFHxS	PFNA	PFDA	PFOS	PFUnDA	PFDoA	PFTeDA	ΣPFASs	Reference
Orge River	France	ND	0.06 ± 0.01	ND	0.03 ± 0.01	0.10 ± 0.02	0.05 ± 0.01	0.30 ± 0.02	4.30 ± 0.30	0.29 ± 0.01	1.7 ± 0.0	0.86 ± 0.03		Labadie and Chevreuil, 2011
Homebush Bay	Australia	ND-0.16 (0.03, -)	ND	ND	ND	ND-0.10 (0.04, -)	ND-0.11 (0.04, -)	ND-0.81 (0.26, -)	0.80-6.20 (2.10, -)	0.10-0.61 (0.22, -)	ND-1.10 (0.34, -)	ND		Thompson et al., 2011
Sludge	Kenya	0.03-0.35	ND-0.37	NA	NA	ND-0.83	ND-0.07	ND-0.34	ND-0.67	ND-0.40	ND-0.24			Chirikona et al., 2015
Rivers and lakes	Korea	ND-0.28 (0.05, -)	ND-0.05 (0.01, -)	NA	ND-0.06 (0.00, -)	ND-0.01 (0.00, -)	ND-0.15 (0.04, -)	ND-0.08 (0.03, -)	0.01-0.48 (0.12, -)	ND-0.09 (0.04, -)	0.01-0.13 (0.05, -)	NA	0.03-1.09 (0.35, -) (10 PFASs)	Lam et al., 2014
Bohai Sea	China	0.11-0.81 (0.29, 0.24)	ND	0.04-0.19 (0.06, 0.05)	ND	ND-0.09 (0.03, 0.03)	ND-0.16 (0.09, ND)	0.02-0.03 (0.02, 0.02)	0.02-0.74 (0.11, 0.04)	0.02-0.03 (0.03, 0.02)	0.03-0.05 (0.03, 0.03)	ND-0.04 (0.03, ND)	0.50-2.78 (0.98, 0.73) (18 PFASs)	Chen et al., 2016
Bohai Sea	China	0.08-0.64 (0.30, 0.28)	ND	0.05-0.09 (0.06, 0.05)	ND-0.18 (0.14, 0.14)	ND-0.03 (ND, ND)	ND-0.15 (0.08, ND)	0.02-0.03 (0.02, 0.02)	ND-0.14 (0.06, 0.06)	0.02-0.04 (0.03, 0.02)	ND-0.04 (0.03, 0.03)	ND-0.04 (ND, ND)	0.33-1.75 (0.72, 0.61) (18 PFASs)	Chen et al., 2016
Kyoto River	Japan	1.30-3.90	NA	ND-2.20	NA	ND-3.30	NA	NA	0.80-6.40	NA	0.20-3.40	NA		Senthilkumar et al., 2007
Sediment Pearl River	China	0.05-0.99											0.10-8.66 (18 PFASs)	Pan et al., 2014
Tangxun Lake, Wuhan	China	0.48-6.35	ND	21.10-78.40	ND-1.29	0.89-4.16	ND	ND-0.41	23.90-623.00	ND-3.27	ND-18.40	NA		Zhou et al., 2013
Yangtze River	China	ND	ND	ND	ND	ND	ND	ND	72.90-536.70	ND	ND	ND		Zhou et al., 2013
Yangtze River	China	0.19-62.45	ND-1.62	ND-0.06	ND-0.19	ND-0.03	ND-3.19	ND-3.85	ND-1.56	ND-2.29	ND-0.98	ND-0.07		Zhou et al., 2013
East China Sea	China	ND-1.40	ND-3.04	ND	ND-4.82	ND-	ND-1.24	ND-0.82	ND-32.40	ND	ND	NA		Yan et al., 2015
Liao River	China	0.02-0.18	NA	ND-0.13	ND-0.13	ND-0.1	0.01-0.07	0.01-0.50	0.04-0.48	0.01-0.03	ND-0.04	NA		Yang et al., 2011
Taihu	China	ND-0.85	ND-0.34	NA	ND-0.73	ND-0.34	ND-0.37	0.13-0.35	0.13-6.95	0.18-1.52	ND-0.23			Yang et al., 2011
Dianchi Lake	China	ND-0.71					ND-0.30	ND-0.18	0.07-0.83	ND-0.17	ND-0.14			Zhang et al., 2012
Baltic Sea	China	5.79-50.60	ND-13.70	NA	ND-5.44	28.00-326.00	ND-40.60	ND-45.50	30.90-1630.00	ND-81.90	ND-8.13	NA		Falandysz et al., 2012

ND: not detected; NA: not available.

154 Table S6 Ratios between PFASs along the Yangtze River, China

	PFOA/PFNA		PFHpA/PFOA		PFOS/PFOA	
	Spring	Autumn	Spring	Autumn	Spring	Autumn
SG	-	-	0.00	0.00	0.00	13.47
PZH	513.39	-	0.00	-	0.00	-
XLD	-	-	0.00	0.00	0.00	50.04
PS	-	-	0.00	0.00	0.00	26.03
XJB	3.03	-	0.00	0.00	1.07	71.07
YB	-	-	0.00	0.00	0.00	2.97
GC(MJ)	268.18	-	0.00	0.00	0.00	2.18
LZ	-	-	0.00	0.00	0.00	0.03
ZT	-	-	0.00	0.00	0.00	0.07
CT	809.01	-	0.00	0.00	0.00	0.05
XHB(JLJ)	-	11.70	0.00	0.41	0.00	4.10
WS(JLJ)	-	-	0.00	0.00	0.00	0.85
LDX(JLJ)	1.16	-	2.23	0.00	1.89	6.33
BB(JLJ)	-	-	0.00	0.00	0.00	1.70
WL(WJ)	-	-	-	0.00	-	4.64
BD	-	-	0.00	0.00	0.00	0.10
MH	-	-	0.00	0.00	0.00	0.00
HLM	-	-	0.00	0.00	0.00	0.00
XXT(HBH)	-	-	0.00	0.00	0.00	0.00
YMZ(HBH)	461.04	-	0.00	0.00	0.00	0.11
YC	148.99	-	0.00	0.00	0.03	0.00
SS	-	-	0.00	0.00	0.00	0.00
CLJL	9.47	-	0.00	0.00	0.00	0.00
NZ(DTH)	-	-	0.00	0.00	0.00	0.00
ZWM(DTH)	14.71	-	0.00	0.00	0.00	0.00
XYIN(DTH)	-	-	0.00	0.00	0.00	0.00
CLJ(DTH)	5.30	10689.00	0.00	0.00	0.25	0.06
LS	-	-	0.00	0.00	0.00	0.08
WH	141.94	642.86	0.00	0.00	0.00	0.00
BH (HJ)	-	-	0.00	-	0.00	-
DJK (HJ)	-	-	0.00	-	0.00	-
TC (HJ)	3.77	1.97	0.00	0.00	0.00	0.00
XY (HJ)	-	-	0.00	0.00	0.00	11.12
XT (HJ)	-	-	0.00	0.00	0.00	0.00
JJZ (HJ)	-	36.09	0.00	0.00	0.01	0.66
JJ	-	220.93	0.00	0.00	0.00	0.00
HK (PYH)	181.38	-	0.00	0.00	0.00	0.00
DT	-	56.31	0.00	0.00	0.00	0.00
WHU	-	118.32	-	0.00	-	0.01
MAS	-	37.98	0.00	0.00	0.00	0.11
NJ	61.95	-	0.00	0.00	0.02	0.03
ZJ	-	211.33	0.00	0.00	0.00	0.08
XLJ	-	119.65	0.00	0.00	0.00	0.02
Mean	187.38	1104.19			0.54	7.84

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157 Table S7 Selected chronic toxicity data of target compounds for algae, daphnids, and
 158 fish from the EPI Suite (EPIWEB v.4.1)

	Chronic EC50 or LC50 (mg/L)		
	fish	daphnids	algae
PFHxA	13.996	11.306	36.829
PFHpA	4.347	4.15	16.864
PFOA	1.341	1.495	7.576
PFNA	0.405	0.53	3.354
PFDA	0.121	0.186	1.468
PFUnDA	0.036	0.065	0.636
PFDoA	0.011	0.022	0.274
PFTeDA	0.000888	0.003	0.05
PFBS	344.669	186.865	351.893
PFHxS	33.403	24.981	73.203
PFOS	3.035	3.131	14.277

159 EC50: median effective concentration;

160 LC50: median lethal concentration.

161 Table S8 Industrial output value (100 million yuan) of manufacturing industries in major cities along the Yangtze River

Category	Sub-category	Yunnan Province	Sichuan Province	Chongqing	Enshi,* Hubei Province	Yichang, Hubei Province	Jingzhou, Hubei Province	Yueysng, Hunan Province	Wuhan, Hubei Province	Jiujiang, Jiangxi Province	Tongling, Anhui Province	Maanshan,** Anhui Province	Nanjing, Jiangsu Province	Zhenjiang, Jiangsu Province	Shanghai
		SG, XLD, XJB	PZH, PS, YB, LZ	ZT, CT	BD	MH, HLM, YC	SS	CLJL	WH	JJ	DT	WHU, MAS	NJ	ZJ	XLJ
Food processing and production Textile, fiber, rubber, and leather	Food Processing	508.7	2529.3	663.8	87.1	299.6	541.6	na	346.9	na	3.0	166.9	17.0	179.5	341.3
	Food Production	167.1	786.6	168.4	7.8	182.0	26.9	999.0	133.0	na	na	79.0	11.6	20.2	638.0
	Textile Industry	19.7	838.1	183.7	1.4	85.6	130.4	224.0	88.9	na	10.2	10.3	9.6	105.8	226.2
	Textile, Garments, and Fashion Industry	7.5	188.9	101.8	2.1	40.9	36.4	na	82.9	735.0	4.7	41.2	36.9	147.3	508.0
	Chemical Fibers	15.0	166.9	4.7	na	2.7	na	na	5.0	na	na	0.5	4.7	29.1	40.5
	Rubber and plastic products	127.9	766.3	401.2	8.3	114.8	84.0	na	206.4	na	1.6	30.8	16.9	98.6	876.0
	Leather, Furs, Down and Related Products	5.6	280.4	144.7	9.9	2.4	2.8	na	11.0	na	0.8	7.1	6.7	53.4	178.8
Paper and printing	Papermaking and Paper Products	64.8	470.3	212.7	2.2	72.0	82.4	170.0	77.9	na	1.4	41.2	3.3	217.7	284.0
	Printing and Record Processing	65.2	255.9	105.5	1.1	34.7	5.0	na	75.8	na	1.0	5.7	4.4	26.5	188.5
Chemical and pharmaceutical products	Raw Chemical Material and Chemical Products	829.0	2322.7	760.3	12.4	1173.8	217.0	na	240.9	na	121.1	108.0	179.5	1323.2	2619.6
	Medical and pharmaceutical Products	259.4	1009.2	323.0	15.5	117.4	34.0	106.0	196.1	na	3.8	9.9	21.4	36.7	596.1
Metal products	Smelting and Pressing of Ferrous Metals	1126.0	2461.1	739.1	1.0	41.9	34.1	na	927.6	na	41.7	717.0	77.8	307.4	1555.5
	Smelting and Pressing of Nonferrous Metals	1470.4	763.6	554.6	1.2	102.0	17.0	na	42.0	na	1088.7	30.4	32.2	146.0	471.3
	Metal Products	75.9	912.5	378.8	5.6	120.8	98.5	na	308.4	na	5.5	107.5	38.5	331.0	942.1
	Instruments and Meters, Manufacturing	15.3	62.9	139.7	0.8	9.5	3.8	na	48.8	na	na	6.8	28.7	316.6	59.1
	Metal products, Machinery and Equipment Repairing	na	58.8	11.7	26.6	7587.0	0.6	na	15.1	na	na	3.8	0.6	0.5	77.8
Motor manufactory	Motor manufacturing	169.9	1926.9	3011.3	1.3	13.7	126.4	na	2063.5	39.6	10.3	86.7	159.9	302.7	4884.1
Aviation and equipment	Railway, Watercraft, Aviation and other Transportation Equipment manufacturing	38.4	508.7	1336.0	na	59.7	2.3	na	325.2	7.5	8.2	90.6	34.1	316.2	706.9
Electric machinery and equipment	Electric Machinery and Equipment	104.8	1030.1	863.0	7.3	183.9	79.4	548.0	750.0		155.2	79.9	76.9	1287.8	2126.2
	Telecommunication Computer, Equipment and Other Electronic Equipment Manufacturing	24.1	3643.1	2128.3	2.0	48.7	5.0	158.0	1261.9	6.7	38.8	34.2	213.9	341.1	5444.3

162 All the data above were obtained from the Yearbook of 2014 for different cities. Sampling sites in the same city were merged. Data in the province yearbook were used for sampling sites in Sichuan and Yunnan
163 provinces. JJ, CLJL and BD were not included in cluster analysis due to inadequate data.* Enshi Tujia and Miao Autonomous Prefecture; ** Data in Maanshan were from the Yearbook of 2015. na: not available.

164 Table S9 log K_{ow} of target PFASs from EPI Suite (EPIWEB v.4.1)

Target PFASs	Log K_{ow}	
PFH _x A	4.37	
PFHpA	5.33	
PFOA	6.30	
PFNA	7.27	
PFDA	6.15	
PFUnDA	6.82	
PFDoDA	7.49	
PFTeDA	8.83	
PFBS	1.82	
PFOS	4.49	
PFH _x S	3.16	

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184 Table S10 Estimated median particle size of sediment (mm) for different scenarios downstream of the
185 TGD obtained in other studies*

site	Prior to TGD (2003)	Post-TGD (2014, measured)	Post-TGD (20 years after operation of TGD)	Post-TGD (50 years after operation of TGD)	Water depth
YC	0.322	54.514	58.514	64.365	14.41
SS	0.198	0.233	10.000	11.000	14.57
CLJ	0.176	0.226	0.260934	10.000	15.96
JJ	0.170	0.223	0.259643	0.260903	16.84
DT	0.127	0.300	0.257121	0.258369	17.79
NJ	0.109	0.187	0.255859	0.257101	17.96
XLJ	0.094	0.146	0.254548	0.255783	18.70

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* Data on estimated median particle size (D_{50}) (Prior-TGD and Post-TGD (2014)) were obtained from Luo (2012); data on estimated median size (Post-TGD (20 years after operation of TGD) and Post TGD (50 years after operation of TGD)) were obtained from Yuan (2014), in which estimates were given of sediment incipient motion velocity and maximum sediment size. According to graphs of estimated riverbed elevation for Yangtze River in Yuan's study, the maximum sediment size is attained at Shashi (SS) in 2023 and Chenglingji (CLJ) in 2053. In the present study, we assumed the correlation between downstream distance and D_{50} post-TGD was similar to that prior to TGD described by Luo (2012). Also, we assumed that when the maximum D_{50} was reached, there would be a slight increase of 10% in sediment median size for post-TGD (20 years after operation of TGD) and post-TGD (50 years after operation of TGD). The data for water depth were taken from Andreadis et al. (2013).

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Table S11 Estimated $\log K_d$ for different scenarios downstream of the TGD

Scenario	Site	PFOA	PFHxA	PFBS	PFHpA	PFHxS	PFNA	PFDA	PFOS	PFUnDA	PFDoA	PFTeDA
Prior to TGD (2003)	YC	2.46	1.74	1.12	2.47	1.73	2.82	3.18	2.45	3.54	3.90	3.40
	SS	2.67	1.95	1.33	2.68	1.94	3.03	3.39	2.66	3.75	4.11	3.62
	CLJ	2.72	2.00	1.39	2.73	1.99	3.08	3.44	2.71	3.81	4.16	3.67
	JJ	2.74	2.01	1.40	2.75	2.00	3.09	3.46	2.73	3.82	4.18	3.68
	DT	2.86	2.14	1.53	2.87	2.13	3.22	3.58	2.86	3.95	4.31	3.81
	NJ	2.93	2.21	1.59	2.94	2.19	3.29	3.65	2.92	4.01	4.37	3.87
	XLJ	2.99	2.27	1.66	3.01	2.26	3.35	3.72	2.99	4.08	4.44	3.94
Post-TGD (2014)	YC	0.23	-0.49	-1.11	0.24	-0.50	0.59	0.95	0.22	1.31	1.67	1.18
	SS	2.60	1.88	1.26	2.61	1.87	2.96	3.32	2.59	3.68	4.04	3.55
	CLJ	2.61	1.89	1.28	2.62	1.88	2.97	3.33	2.60	3.70	4.06	3.56
	JJ	2.62	1.90	1.28	2.63	1.88	2.98	3.34	2.61	3.70	4.06	3.56
	DT	2.49	1.77	1.15	2.50	1.76	2.85	3.21	2.48	3.57	3.93	3.44
	NJ	2.69	1.97	1.36	2.71	1.96	3.05	3.42	2.69	3.78	4.14	3.64
	XLJ	2.80	2.08	1.47	2.81	2.07	3.16	3.52	2.79	3.89	4.25	3.75
Post-TGD (20 years after operation of TGD)	YC	0.20	-0.52	-1.14	0.21	-0.53	0.56	0.92	0.19	1.28	1.64	1.15
	SS	0.97	0.24	-0.37	0.98	0.23	1.33	1.69	0.96	2.05	2.41	1.91
	CLJ	2.55	1.83	1.21	2.56	1.82	2.91	3.27	2.54	3.63	3.99	3.50
	JJ	2.55	1.83	1.22	2.56	1.82	2.91	3.27	2.54	3.64	4.00	3.50
	DT	2.56	1.83	1.22	2.57	1.82	2.92	3.28	2.55	3.64	4.00	3.50
	NJ	2.56	1.84	1.22	2.57	1.83	2.92	3.28	2.55	3.64	4.00	3.50
	XLJ	2.56	1.84	1.23	2.57	1.83	2.92	3.28	2.55	3.65	4.00	3.51
Post-TGD (50 years after operation of TGD)	YC	0.16	-0.56	-1.18	0.17	-0.58	0.52	0.88	0.15	1.24	1.60	1.10
	SS	0.92	0.20	-0.41	0.94	0.19	1.28	1.65	0.92	2.01	2.37	1.87
	CLJ	0.97	0.24	-0.37	0.98	0.23	1.33	1.69	0.96	2.05	2.41	1.91
	JJ	2.55	1.83	1.21	2.56	1.82	2.91	3.27	2.54	3.63	3.99	3.50
	DT	2.55	1.83	1.22	2.57	1.82	2.91	3.28	2.55	3.64	4.00	3.50
	NJ	2.56	1.83	1.22	2.57	1.82	2.92	3.28	2.55	3.64	4.00	3.50
	XLJ	2.56	1.84	1.22	2.57	1.83	2.92	3.28	2.55	3.64	4.00	3.50

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Fig. S1 Box-data plot for seasonal concentrations of PFASs in water and sediment (A, water concentrations in spring; B, water concentrations in autumn; C, sediment concentrations in spring; D, sediment concentrations in autumn). The box denotes 25% and 75% percentiles and the solid horizontal line in a box represents the median value. Scatter plots by the side of the boxes represent concentrations of individual PFAS.

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208 Fig. S2 NMDS (left) and ANOSIM (right) analyses of PFASs concentrations in water samples in spring
209 and autumn (W is water, S is spring, and A is autumn).

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216 Fig. S3 Correlation between spring and autumn PFOA concentrations for (A) main stream and (B)
217 tributaries.

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221 Fig. S4 Relative contributions of individual PFAS (%) to the total PFASs in water samples from the
222 Yangtze River in (A) spring and (B) autumn seasons.

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Fig. S5 NMDS (left) and ANOSIM (right) analyses of PFASs concentrations in sediment samples in spring and autumn (first S is sediment, second S is spring, A is autumn)

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 230 Fig. S6 Relative contribution of individual PFAS (%) to the total PFASs in sediment samples from the
 231 Yangtze River in (A) spring and (B) autumn.
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Fig. S7 Ecological risks related to individual PFASs for each trophic level under different scenarios in the Yangtze River

Fig. S8 Mixture risk quotients (MRQs) for fish experiencing different reductions in PFASs for scenarios of 20, 30, 40 and 50 years after operation of TGD: (a) 10%; (b) 20%; and (c) 50% reduction per decade).

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