Supporting Information:

Occurrence of per- and polyfluorinated alkyl substances in wastewater treatment plants in Northern Italy.

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Table S1. Basic information on the four wastewater treatment plants investigated in this study and located in the province of Milan, Italy.

WWTP	Municipalities	Treatment processes	Treatment capacity (m^3/h)	Avarage Daily flow rate (m^3/d)	HRT (h) aerobic oxidation	Equivalent person (p.e.)	Population load % p.e.	Industrial load % p.e.
A	Assago, Buccinasco, Corsico, Cesano Boscone	ME, WD, DE, AOAC, MBR, UF, DI	5,000	81,498 in July; 67,660 in October; 62,428 in February; 64,027 in May	8 in July; 9 h in October; 9 in February; 10h in May	160,000	81	19
B	San Donato Milanese – San Giulianio – Mediglia	ME, DE, AOAC, SF, FI, DI	139	31,132 in July; 26,519 in October; 30,477 in February; 28,196 in May	12h in July; 14h in October; 12 in May; 13 in February	80,000	90	10
C	Bresso, Paderno D., Cinisello B., Cusano, Cormano	ME, WD, PS, AOAC, DE, SF, FI, DI	5,000	56,110 in July; 55,837 in October; 54,199 in February; 53,699 in May	10h in July; 10h in October; 11h in February; 11h in May	300,000	87	13
D	Mariano Comense, Meda, Lentate sul Seveso, Cambiate, Cesano Maderno, Barlassina, Bovisio Masciago, Vanzago, Varedo, Seveso, Rho, Senago, Solaro, Pero, Pogliano, Pregnana, Parabiago fraz Villastanza,	ME, WD, PS, AOAC, DE, NI, SS, FI, DI	29,520	132,632 in July; 153,707 in October; 141,820 in February; 127,072 in May	6h in July; 5h in October; 6h in February; 6 in May	720,000	85	15

	Nerviano, Novate, Lazzate, Limbiate, Misinto, Arese, Baranzate, Bollate, Ceriano Laghetto, Cesate, Cogliate, Garbagnate, Lainate							

ME= mechanical treatment including coarse and fine screening, grit removal

WD= water deoling

DE= denitrification

NI= nitrification

MBR= membrane bioreactor

DI= water disinfection with oxidant agents or UV

AOAC= aerobic oxidation with activeted sludge

PS= primary sedimentation

SS= secondary sedimentation

SF= final sedimentation

UF= ultrafiltration,

FI= filtration

Table S2. Quality control parameters for the adopted analytical methods. Linearity range, recoveries, and relative standard deviation (RDS %) obtained by triplicate analyses, and limits of quantification (LOQs) for water and sludge samples.

PFAS	MRM Transitions (quantifier and qualifier) (*m/z*)	Internal standard for quantification (IS)	Linearity R^2 (range 0.01-25 µg L^{-1})	Recoveries and RDS (%) in water	MLOQ water (ng L^{-1})	Recoveries and RDS	MLOQ sludge (ng kg^{-1} dw)

						(%) in sludge	
PFBA	213 -> 169 not detected	$^{13}C_4$-PFBA	0.9923	92±6	9.87	104±18	0.11
PFPeA	263 -> 219 not detected	^{13}C-PFPeA	0.9901	91±5	9.94	94±18	0.15
PFHxA	313 -> 269 313 -> 119	$^{13}C_5$-PFHxA	0.9857	89±4	7.69	93±19	0.12
PFHpA	363 -> 319 363 -> 169	$^{13}C_4$-PFHpA	0.9852	87±4	5.88	98±19	0.08
PFOA	413 -> 369 413 -> 169	$^{13}C_8$-PFOA	0.9872	82±5	1.61	87±18	0.09
PFNA	463 -> 419 463 -> 219	$^{13}C_9$-PFNA	0.9891	86±4	4.16	97±19	0.09
PFDA	513 -> 469 513 -> 219	$^{13}C_6$-PFDA	0.9902	75±3	8.33	93±18	0.13
PFUnDA	563 -> 519 563 -> 269	$^{13}C_7$-PFUdA	0.9771	76±9	9.87	89±18	0.08
PFDoDA	613 -> 569 613 -> 169	$^{13}C_2$-PFDoA	0.9834	78±3	2.94	92±18	0.10
PFTrDA	663 -> 619 663 -> 169	$^{13}C_2$-PFTrDA	0.9863	85±4	1.14	66±18	0.17
PFTeDA	713 -> 319 713 -> 669	$^{13}C_2$-PFTreDA	0.9863	72±9	1.38	93±19	0.17
PFBS	299 -> 80 299-> 99	$^{13}C_3$-PFBS	0.9879	94±3	9.88	96±18	0.07
PFPeS	349 -> 80 349 -> 99	$^{13}C_4$-PFHpA	0.9852	90±2	8.97	-	-
PFHxS	399 -> 80 not reported	^{13}C-PFHxS	0.9825	88±4	9.95	93±19	0.07
PFHpS	449 -> 80 449 -> 99	^{13}C-PFNA	0.9891	75±3	10.01	-	-
PFOS	499 -> 80 499 -> 99	$^{13}C_8$-PFOS	0.9933	73±5	2.85	91±19	0.09
PFDS	599 -> 80 599 -> 99	^{13}C-PFDoA	0.9834	81±5	9.94	-	-
PFOSA	498 -> 78 498 -> 169	$^{13}C_8$-PFOSA	0.9881	75±5	9.99	-	-
NEtFOSA	584 -> 419 584 -> 526	d_5-NEtFOSA	0.9958	76±4	10.02	-	-

NMeFOSAA	570 -> 419 570 -> 512	d_3-N-MeFOSAA	0.9739	73±4	4.21	-	-
4: 2 FTS	327 -> 307 327 -> 81	$^{13}C_2$ - 4:2 FTS	0.986	87±4	4.54	-	-
6: 2 FTS	427 -> 407 427 -> 81	$^{13}C_2$ - 6:2 FTS	0.9874	74±3	9.48	-	-
8: 2 FTS	527 -> 507 527 -> 81	$^{13}C_2$ - 8:2 FTS	0.9815	77±3	4.76	-	-
FBSA	298 -> 78 298 -> 119	^{13}C-PFBS	0.9840	90±3	1.62	-	-
FHxSA	398 -> 78 398 -> 169	^{13}C-PFxS	0.9825	87±4	10	-	-

Table S3. PFAS concentrations (µg L^{-1}) measured in water samples collected from the influents of four WWTPs (A, B, C, D) in July and October 2021, May and February 2022.

PFAS	July				October				May				February			
	A	B	C	D	A	B	C	D	A	B	C	D	A	B	C	D
PFBA	1.74	3.34	1.66	1.68	1.85	5.56	1.87	2.02	0.76	3.11	3.14	2.28	0.82	2.38	3.78	3.12
PFPeA	5.32	6.14	21.37	5.61	6.28	5.56	18.38	16.72	1.91	18.34	11.81	13.90	10.99	30.04	13.65	9.17
FBSA	0.07	0.17	<LOQ	0.68	<LOQ	0.67	<LOQ	0.91	0.08	0.28	0.13	0.15	<LOQ	<LOQ	<LOQ	0.21
PFBS	3.21	1.58	2.71	<LOQ	5.88	3.97	2.89	6.21	1.73	1.21	6.49	1.15	2.10	1.25	2.36	11.7
PFHxA	2.29	5.65	3.54	4.93	4.38	4.13	1.70	4.05	7.87	1.10	2.11	2.62	2.12	0.85	3.47	4.48
4:2 FTS	0.20	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ
PFPeS	<LOQ	<LOQ	0.76	<LOQ	<LOQ	1.49	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ
PFHpA	1.57	2.67	1.93	1.83	4.06	2.84	1.23	1.75	0.73	0.59	1.97	0.95	1.19	0.71	1.95	1.31
FHxSA	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ
PFHxS	5.06	3.80	<LOQ	<LOQ	<LOQ	13.54	<LOQ	10.46	7.05	393	<LOQ	10.5	5.31	<LOQ	<LOQ	11.6
PFOA	1.82	2.72	2.65	2.48	3.81	7.55	2.29	2.92	1.04	1.04	3.79	2.00	1.43	1.67	2.50	3.24
6:2 FTS	<LOQ	<LOQ	<LOQ	0.83	0.28	<LOQ	<LOQ	0.36	<LOQ	0.67	0.43	0.09	<LOQ	<LOQ	0.07	0.56
PFHpS	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	0.72	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ
PFNA	0.02	0.35	0.82	1.09	0.18	0.71	0.33	0.69	<LOQ	0.54	1.74	0.90	0.22	0.28	1.11	1.19
PFOSA	<LOQ	<LOQ	<LOQ	0.21	<LOQ	<LOQ	<LOQ	0.20	<LOQ	0.05	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ
PFOS	3.77	<LOQ	1.32	2.27	6.44	13.10	1.26	<LOQ	1.83	2.61	1.18	2.33	1.87	3.22	2.47	3.42
PFDA	<LOQ	0.02	0.03	0.04	<LOQ	<LOQ	<LOQ	0.90	<LOQ	1.83	0.72	0.46	<LOQ	0.02	0.26	0.32
8:2 FTS	0.77	0.76	1.06	0.80	0.79	0.83	0.84	0.83	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ
PFUnDA	0.22	0.16	<LOQ	<LOQ	<LOQ	0.24	<LOQ	0.27	0.12	1.83	3.23	0.51	1.21	0.34	<LOQ	<LOQ

NMeFOSAA	2.58	<LOQ	15.7	<LOQ	<LOQ	6.18	10.9	9.40	5.44	5.96	7.21	7.69	7.80	3.96	20.3	13.0
NEtFOSA	0.26	0.29	<LOQ	0.35	<LOQ	0.31	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ
PFDS	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	0.08	<LOQ	0.16	<LOQ	<LOQ	0.03	<LOQ	<LOQ	<LOQ
PFDoDA	0.18	0.21	<LOQ	0.19	0.15	0.16	0.11	0.28	0.23	0.79	0.76	0.81	0.50	0.34	3.24	0.28
PFTrDA	3.93	1.93	0.36	0.47	<LOQ	<LOQ	<LOQ	0.20	1.35	5.05	5.58	15.16	3.32	2.68	1.90	0.28
PFTeDA	0.78	1.13	0.98	0.61	0.26	<LOQ	<LOQ	0.58	<LOQ	0.13	2.25	<LOQ	0.93	2.06	3.27	0.11
Total	33.8	30.9	54.9	24.1	34.4	66.9	41.8	58.8	30.2	438	53.3	61.5	39.9	49.8	60.4	64.5

Table S4. PFAS concentrations (μg L^{-1}) measured in water samples collected from the effluents of four WWTPs (A, B, C, D) in July and October 2021, May and February 2022.

PFAS	July				October				May				February			
	A	B	C	D	A	B	C	D	A	B	C	D	A	B	C	D
PFBA	1.55	2.11	5.78	2.26	2.43	3.82	0.70	3.40	1.24	1.64	5.68	3.25	1.58	1.83	2.31	6.46
PFPeA	8.16	2.10	7.94	6.27	12.57	4.09	1.15	4.48	5.01	1.21	6.84	3.09	4.27	1.71	6.08	3.88
FBSA	0.15	0.10	0.12	0.15	0.26	0.25	<LOQ	0.28	0.19	0.21	0.18	0.17	0.15	0.10	0.19	0.28
PFBS	3.74	1.37	3.76	4.28	6.09	2.17	0.80	66.1	4.19	0.83	4.22	5.40	3.02	0.63	5.76	14.9
PFHxA	3.60	2.69	7.32	7.24	7.72	2.68	0.46	5.43	2.71	1.99	4.11	4.47	2.78	1.34	5.84	5.70
4:2 FTS	<LOQ	0.24	0.36	0.26	<LOQ	<LOQ	0.12	0.34	0.09	0.11	0.26	0.07	0.15	<LOQ	0.25	<LOQ
PFPeS	<LOQ	<LOQ	<LOQ	<LOQ	0.65	<LOQ	0.62	0.58	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ
PFHpA	1.89	1.74	1.74	2.96	3.33	1.18	0.57	1.79	0.88	1.07	2.39	0.91	0.64	0.53	2.33	2.36
FHxSA	<LOQ	<LOQ	<LOQ	<LOQ	0.70	0.62	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ
PFHxS	0.54	0.12	<LOQ	<LOQ	2.56	1.36	0.84	1.46	20.7	0.06	0.04	0.42	0.76	<LOQ	0.31	0.43
PFOA	3.64	1.47	2.44	3.13	4.49	1.87	0.51	3.60	1.70	1.03	4.53	1.87	1.51	0.85	3.39	3.39
6:2 FTS	<LOQ	0.47	<LOQ	0.62	0.02	<LOQ	<LOQ	0.22	<LOQ	0.26	26.1	0.46	<LOQ	<LOQ	<LOQ	0.90
PFHpS	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ
PFNA	0.21	0.20	0.57	0.83	0.13	<LOQ	<LOQ	0.47	0.05	0.01	2.30	0.55	<LOQ	0.01	1.16	0.80
PFOSA	<LOQ	<LOQ	<LOQ	0.26	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	0.35	0.07	<LOQ	<LOQ	<LOQ	<LOQ
PFOS	3.97	2.48	3.65	3.30	5.01	2.04	3.11	7.77	1.91	1.50	3.61	2.29	3.06	1.82	5.74	12.0
PFDA	<LOQ	0.25	0.33	<LOQ	<LOQ	<LOQ	<LOQ	6.41	<LOQ	<LOQ	2.00	0.93	0.63	<LOQ	0.03	0.18
8:2 FTS	0.76	0.76	0.77	0.81	0.75	0.76	0.76	0.78	<LOQ	<LOQ	0.15	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ
PFUnDA	0.24	0.17	<LOQ	<LOQ	0.15	0.10	0.11	0.11	0.67	0.16	1.37	0.74	0.12	0.19	<LOQ	0.13
NMeFOSAA	1.10	<LOQ	1.21	0.39	1.73	1.82	2.76	3.67	0.17	0.05	1.93	0.71	0.38	1.96	1.47	0.86
NEtFOSA	0.23	0.23	0.24	0.33	0.26	0.25	0.31	0.30	<LOQ	<LOQ	0.06	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ
PFDS	<LOQ	<LOQ	<LOQ	<LOQ	0.01	<LOQ	<LOQ	<LOQ	0.03	<LOQ	0.07	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ
PFDoDA	0.17	0.95	0.21	0.16	0.10	0.09	0.09	0.09	0.55	0.26	0.45	0.48	0.33	0.43	0.35	0.20

PFTrDA	0.82	1.42	4.40	5.06	0.10	<LOQ	0.21	<LOQ	1.50	2.96	1.56	7.75	2.41	4.95	6.91	4.46
PFTeDA	1.01	3.07	0.71	0.80	0.34	<LOQ	0.40	0.08	0.39	<LOQ	<LOQ	<LOQ	2.09	7.73	2.41	1.03
Total	31.8	22.0	41.6	39.1	49.4	23.1	13.5	107	42.0	13.4	68.2	33.6	23.9	24.1	44.6	57.9

Table S5. PFAS mass load (g/day) in influent wastewaters from four WWTPs (A, B, C, D).

PFAS	July				October				May				February			
	A	B	C	D	A	B	C	D	A	B	C	D	A	B	C	D
PFBA	142	104	92.9	222	125	148	104	311	48.8	87.6	168	289	51.0	72.6	205	443
PFPeA	434	191	1198	744	425	147	1026	2571	122	517	634	1766	686	916	740	1300
FBSA	5.39	5.21	0.04	90.0	0.05	17.7	0.04	139	4.87	7.87	7.00	18.4	0.05	0.02	0.04	30.1
PFBS	261	49.2	152	0.65	398	105	162	953	111	34.2	349	145	131	38.0	128	1665
PFHxA	186	176	198	653	296	109	95.1	623	504	31.1	113	333	132	25.8	188	635
4:2 FTS	16.2	0.07	0.13	0.30	0.15	0.06	0.13	0.35	0.14	0.06	0.12	0.29	0.14	0.07	0.12	0.32
PFPeS	0.36	0.14	42.4	0.59	0.30	39.6	0.25	0.69	0.29	0.13	0.24	0.57	0.28	0.14	0.24	0.64
PFHpA	128	83.2	109	243	275	75.4	68.4	268	46.4	16.7	106	120	74.5	21.7	106	186
FHxSA	0.41	0.15	0.28	0.66	0.34	0.13	0.28	0.76	0.32	0.14	0.27	0.63	0.31	0.15	0.27	0.71
PFHxS	412	118	0.28	0.66	0.34	359	0.28	1607	451	11089	0.27	1334	331	0.15	0.27	1652
PFOA	148	85	149	329	257	200	128	449	66.8	29.2	203	254	89.5	50.1	135	459
6:2 FTS	0.39	0.15	0.26	110	18.7	0.12	0.26	55.7	0.30	18.9	23.1	10.9	0.29	0.14	3.53	79.3
PFHpS	0.41	0.15	0.28	0.66	0.34	0.13	0.28	0.77	0.32	0.14	38.7	0.63	0.31	0.15	0.27	0.71
PFNA	1.95	10.8	46.0	144	12.5	18.8	18.6	106	0.13	15.3	93.7	114	13.5	8.55	60.2	168
PFOSA	0.41	0.15	0.28	28.2	0.34	0.13	0.28	30.7	0.32	1.45	0.26	0.63	0.31	0.15	0.27	0.70
PFOS	307	0.04	74.2	301	436	347	70.6	0.22	117	73.5	63.6	296	117	98.1	134	484
PFDA	0.34	0.48	1.50	4.68	0.28	0.11	0.23	138	0.27	51.5	38.9	58.1	0.26	0.51	14.1	44.9
8:2 FTS	62.5	23.7	59.6	106	53.5	22.0	47.0	127	0.15	0.07	0.13	0.30	0.15	0.07	0.13	0.33
PFUnDA	17.7	5.0	0.28	0.65	0.33	6.30	0.28	42.0	7.41	51.6	173	65.0	75.7	10.5	0.27	0.70
NMeFOSAA	210	0.06	881	0.28	0.14	164	607	1445	348	168	387	977	487	121	1102	1849
NEtFOSA	21.2	8.92	0.28	46.8	0.34	8.13	0.28	0.77	0.32	0.14	0.27	0.63	0.31	0.15	0.27	0.71
PFDS	0.40	0.15	0.28	0.66	0.33	0.13	0.28	12.7	0.32	4.58	0.27	0.63	1.87	0.15	0.27	0.70
PFDoDA	14.7	6.49	0.08	25.7	9.94	4.20	6.18	42.5	14.8	22.3	40.7	102	31.0	10.2	175	40.1
PFTrDA	320	60.1	20.1	62.2	0.04	0.01	0.03	30.2	86.4	142	300	1926	207	81.8	103	16.2
PFTeDA	63.7	35.1	55.2	80.5	17.9	0.02	0.04	88.4	0.04	3.68	121	0.09	130	62.7	177	92.1
Total	2756	963	3082	3196	2328	1773	2336	9044	1933	12366	2862	7817	2562	1519	3273	9151

Table S6. PFAS mass load (g/day) in effluent wastewaters from four WWTPs (A, B, C, D).

PFAS	July				October				May				February			
	A	B	C	D	A	B	C	D	A	B	C	D	A	B	C	D
PFBA	126	65.7	324	300	164	101	38.9	523	79.3	46.2	305	413	99.0	55.8	125	917
PFPeA	665	62.3	446	831	851	108	64.2	688	321	34.1	367	393	266	52.0	330	550
FBSA	12.5	3.18	6.58	19.8	17.3	6.71	0.04	43.2	12.0	6.02	9.89	22.1	9.21	3.12	10.3	39.5
PFBS	305	42.6	211	568	412	57.6	44.5	10166	268	23.4	226	686	189	19.2	312	2108
PFHxA	293	83.8	411	960	522	71.0	25.8	835	173	56.0	221	568	174	40.7	316	809
4:2 FTS	0.18	7.49	20.2	34.3	0.15	0.06	6.60	52.9	5.71	3.07	13.9	8.65	9.45	0.07	13.5	0.32
PFPeS	0.36	0.14	0.25	0.59	44.3	0.12	34.4	89.5	0.29	0.13	0.24	0.57	0.28	0.14	0.24	0.64
PFHpA	154	54.0	97.4	393	225	31.2	31.6	275	56.2	30.0	128	115	39.8	16.2	126	335
FHxSA	0.41	0.15	0.28	0.66	47.3	16.5	0.28	0.77	0.32	0.14	0.27	0.63	0.31	0.15	0.27	0.71
PFHxS	43.7	3.61	0.28	0.66	173	36.0	41.2	225	1323	1.75	2.41	53.1	47.7	0.15	16.8	60.8
PFOA	296	45.7	137	414	304	49.5	28.5	554	109	29.1	243	237	94.2	26.0	184	481
6:2 FTS	0.38	14.7	0.26	82.7	1.56	0.13	0.26	34.5	0.30	7.30	1400	58.7	0.29	0.14	0.26	128
PFHpS	0.41	0.15	0.28	0.66	0.34	0.13	0.28	0.77	0.32	0.14	0.27	0.63	0.31	0.15	0.27	0.71
PFNA	16.8	6.18	31.9	109	8.67	0.05	0.12	72.6	3.01	0.41	124	69.8	0.13	0.25	62.7	113
PFOSA	0.41	0.15	0.28	35.0	0.34	0.13	0.28	0.77	0.32	0.14	18.8	9.00	0.31	0.15	0.27	0.71
PFOS	324	77.1	205	438	339	54.1	174	1194	122	42.2	194	290	191	55.5	311	1696
PFDA	0.34	7.76	18.7	0.55	0.28	0.11	0.23	985	0.27	0.12	107	118	39.5	0.13	1.58	25.6
8:2 FTS	61.6	23.8	43.1	107	50.9	20.2	42.2	120	0.15	0.07	7.81	0.30	0.15	0.07	0.13	0.34
PFUnDA	19.6	5.40	0.28	0.66	9.85	2.56	6.23	17.6	43.0	4.62	73.4	94.1	7.53	5.71	0.27	18.5
NMeFOSAA	90.0	0.06	67.6	52.3	117	48.2	154	563	10.8	1.52	103	90.6	23.7	59.7	79.8	122
NEtFOSA	18.7	7.18	13.6	44.4	17.2	6.70	17.4	45.3	0.32	0.14	3.11	0.63	0.31	0.15	0.27	0.71
PFDS	0.40	0.15	0.28	0.66	0.73	0.13	0.28	0.76	2.03	0.14	3.64	0.63	0.31	0.15	0.27	0.70
PFDoDA	14.0	29.5	11.7	21.0	7.05	2.43	5.11	13.7	35.4	7.26	24.1	60.8	20.4	13.1	19.0	28.8
PFTrDA	66.9	44.1	247	671	6.52	0.01	12.0	0.09	96.3	83.5	83.7	984	150	151	374	633
PFTeDA	82.2	95.6	39.9	106	23.2	0.02	22.1	13.0	24.7	0.02	0.04	0.09	130	236	131	146
Total	2593	684	2333	5192	3343	614	756	16514	2687	378	3661	4276	1493	735	2416	8217

Table S7. Percent difference between influent and effluent concentrations of single PFAS for WWTP A. Negative values indicate an increase in PFAS concentration from influent to effluent. The value 0% was obtained when the concentration of PFAS was lower than MLOQ in both influent and effluent.

PFAS	July	October	May	February
PFBA	11	-31	-62	-94
PFPeA	-53	-100	-162	61
FBSA	-132	-31455	-146	-18122
PFBS	-16	-3	-141	-43
PFHxA	-57	-76	-66	-31
4:2 FTS	99	0	-3830	-6569
PFPeS	0	-14503	0	0
PFHpA	-20	18	21	46
FHxSA	0	-13885	0	0
PFHxS	89	-51372	-193	85
PFOA	-100	-18	-63	-5
6:2 FTS	0	92	0	0
PFHpS	0	0	0	0
PFNA	-761	30	-2160	99
PFOSA	0	0	0	0
PFOS	-5	22	-5	-64
PFDA	0	0	0	-15095
8:2 FTS	1	5	0	0
PFUnDA	-10	-2850	-480	90
NMeFOSAA	57	-81979	97	95
NEtFOSA	12	-4990	0	0
PFDS	0	-118	-539	83
PFDoDA	5	29	-139	34
PFTrDA	79	-168199	-11	27
PFTeDA	-29	-30	-55943	-123

Table S8. Percent difference between influent and effluent concentrations of single PFAS for WWTP B. Negative values indicate an increase in PFAS concentration from influent to effluent. The value 0% was obtained when the concentration of PFAS was lower than MLOQ in both influent and effluent.

PFAS	July	October	May	February
PFBA	36	31	47	23
PFPeA	66	26	93	94
FBSA	39	62	23	-12552
PFBS	13	45	31	49
PFHxA	52	35	-80	-58
4:2 FTS	-10495	0	-4691	0
PFPeS	0	100	0	0
PFHpA	35	58	-79	25
FHxSA	0	-12374	0	0
PFHxS	97	90	100	0
PFOA	46	75	0	49
6:2 FTS	-9861	0	61	0
PFHpS	0	0	0	0
PFNA	43	100	97	97
PFOSA	0	0	90	0
PFOS	-173778	84	42	43
PFDA	-1516	0	100	75
8:2 FTS	0	8.	0	0
PFUnDA	-8	59	91	46
NMeFOSAA	0	70	99	50
NEtFOSA	19	18	0	0
PFDS	0	0	97	0
PFDoDA	-355	42	67	-28
PFTrDA	27	0	41	-84
PFTeDA	-172	0	99	-276

Table S9. Percent difference between influent and effluent concentrations of single PFAS for WWTP C. Negative values indicate an increase in PFAS concentration from influent to effluent. The value 0% was obtained when the concentration of PFAS was lower than MLOQ in both influent and effluent.

PFAS	July	October	May	February
PFBA	-249	63	-81	39
PFPeA	63	94	42	55
FBSA	-14373	0	-41	-23432
PFBS	-39	72	35	-144
PFHxA	-107	73	-94	-68
4:2 FTS	-15778	-5091	-11346	-10860

PFPeS	99	-13649	0	0
PFHpA	10	54	-21	-19
FHxSA	0	0	0	0
PFHxS	0	-16881	-804	-6142
PFOA	8	78	-19	-36
6:2 FTS	0	0	-5948	93
PFHpS	0	0	99	0
PFNA	31	99	-32	-4
PFOSA	0	0	-6909	0
PFOS	-176	146	-205	-132
PFDA	-1152	0	-177	89
8:2 FTS	28	10	-6009	0
PFUnDA	0	-2163	58	0
NMeFOSAA	92	75	73	93
NEtFOSA	-4748	-6105	-1055	0
PFDS	0	0	-1265	0
PFDoDA	-14135	17	41	89
PFTrDA	-1126	-37484	72	-264
PFTeDA	28	-57296	100	26

Table S10. Percent difference between influent and effluent concentrations of single PFAS for WWTP D. Negative values indicate an increase in PFAS concentration from influent to effluent. The value 0% was obtained when the concentration of PFAS was lower than MLOQ in both influent and effluent.

PFAS	July	October	May	February
PFBA	-35	-68	-43	-107
PFPeA	-12	73	78	58
FBSA	78	69	-20	-31
PFBS	-86546	-966	-370	-27
PFHxA	-47	-34	-70	-27
4:2 FTS	-11320	-15061	-2899	0
PFPeS	0	-12885	0	0
PFHpA	-62	-2	4	-80
FHxSA	0	0	0	0
PFHxS	0	86	96	96
PFOA	-26	23	7	-5
6:2 FTS	24	38	-439	-61
PFHpS	0	0	0	0

PFNA	24	31	39	32
PFOSA	-24	97	-1317	0
PFOS	-45	-545236	2	-249
PFDA	88	-615	-103	43
8:2 FTS	0	6	0	0
PFUnDA	0	58	-44	-2547
NMeFOSAA	-18624	61	91	93
NEtFOSA	5	-5790	0	0
PFDS	0	94	0	0
PFDoDA	18	68	41	28
PFTrDA	-980	100	49	-3806
PFTeDA	-31	85	0	-59

Table S11. PFAS concentrations (ng kg^{-1} dry weight) in sludge after primary treatment and secondary treatment by activated sludge.

PFAS	Primary										Secondary										Super biologico			
	July		October		May		February				July	JuLy	October		May		February				July	October	May	February
	A	D	A	D	A	D	A	B	C	D	A	D	A	D	A	D	A	B	C	D	D	D	D	D
PFBA	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ
PFBS	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	96.6	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ
PFDeA	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ
PFDoDA	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ
PFHpA	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	108	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ
PFHxA	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	239	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ
PFHxS	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ
PFNA	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ
PFOA	102	<MLOQ	165	<MLOQ	92.6	<MLOQ	96.6	<MLOQ	<MLOQ	<MLOQ	440	<MLOQ	185	<MLOQ	350	98.6	224	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ
PFOS	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	142	<MLOQ	<MLOQ	<MLOQ	250	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ
PFPeA	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ
PFTeDA	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ

PFTrDA	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ
PFUnDA	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ	<MLOQ

Table S12. Comparison of PFAS levels found in wastewater treatment plants in previous studies.

PFAS	Sampling site	Influent concentration (ng/L)	Effluent concentration (ng/L)	Sampling time	References
PFBA, PFPeA, FBSA, PFBS, PFHxA, 4:2 FTS, PFPeS, PFHpA, FHxSA, PFHxS, PFOA, 6:2 FTS, PFHpS, PFNA, PFOSA, PFOS, PFDA, 8:2 FTS, PFUnDA, *N-MeFOSAA, NEtFOSA*, *PFDS*, PFDoDA, PFTrDA, PFTeDA	Milan, Italy	<LOQ-393000 (concentration range) 24100-438000 (total PFAS concentration)	<LOQ-66100 (concentration range) 13500-107000 (total PFAS concentration)	2021-2022	In this study
PFBA, PFPeA, PFHxA, PFHpA, PFOA, PFNA, PFDA, PFUnDA, PFDoDA, PFBS, PFHxS, PFOS	North Milan, Italy	-	15-1128 (mean values, total PFAS concentration)	2010-2013	Castiglioni et al., 2015
PFOA, PFHpA, PFOS, PFNA, PFDA, PFHxA, PFHxS	Europe	-	23.9-304 (concentration range, average values)	2010	Loos et al., 2013
PFBS, PFHxS, PFOS, PFBA, PFPeA, PFHxA, PFHpA, PFOA, PFNA, PFDA, 8:2 FTS, 10:2 FTS	Belgium	<0.2-2726 (concentration range) 26-3406 (total PFAS concentration)	-	2013-2020	Jeong et al., 2022
PFBA, PFPeA, PFHxA, PFHpA, PFOA, PFNA,	Ebro, Guadalquivir,	nd-1870 (concentration	nd-91 (concentration range, mean values)	2010-2011	Campo et al., 2014

PFDA, PFUnDA, PFDoDA, PFTrDA, PFTeDA, PFHxDA, PFODA, PFBS, PFHxS, PFHpS, PFOS, PFNS, PFDS, PFOSA	Jucar and Llobregat Rivers, Spain	range, mean values)			
PFOA, PFBA, PFPA, PFHxA, PFHpA, PFOS	Baltic sea catchment	-	0.1-5 (concentration range)	2001-2019	Undeman et al., 2019
PFOA, PFOS	Bayreuth, Germany	-	20.00-3900 PFOA; 106-336 PFOS (concentration range)	2007	Becker et al., 2010
PFBS, PFHxS, PFHpS, PFOS, PFDS, FOSA, MeFOSA, N-EtFOSAA, FOSAA, MeFOSAA, PFBA, PFPeA, PFHxA, PFHpA, PFOA, PFNA, PFDA, PFUnDA, PFDoDA, PFTrDA, PFTeDA, 6:2 FTCA, 8:2 FTCA: 10:2 FTCA, 6:2 FTUCA, 8:2 FTUCA, 10:2 FTUCA, 6:2 FTSaAM, 6:2 FTAB, 4:2 FTSA; 6:2 FTSA; 8:2 FTSA	France	5.3×10^6 - 1.2×10^8 (total PFAS concentration range)	3.6×10^6 -1.3×10^7 (total PFAS concentration range)	2015-2016	Dauchy et al., 2019
PFBA, PFPeA, PFHxA, PFHpA, PFOA, PFNA, PFDA, PFUnDA, PFDoDA, HFPO-DA, ADONA, PF4OPeA, PF5OHxA, PFO2HpA, PFBS, PFPeS, PFHxS, PFHpS, PFOS, FTS, 6:2 and 8:2 Cl-PFESA, PFEESA	China	0.001-85.7 (concentration range, mean value)	0.004-848 (concentration range, mean value)	2015-2016	Jiang et al., 2023
PFBA, PFPeA, PFHxA, PFHpA, PFOA, PFNA, PFDA, PFBS, PFHxS, PFHpS, PFOS, 6:2 FTS, 6:2 Cl-PFESA; PFOSA	China	4.2 -1133.9 (concentration range)	2.4-1088.5 (concentration range)	2020-2021	Mu et al., 2022

PFBA, PFPeA, PFHxA, PFHpA, PFOA, PFNA, PFDA, PFUnDA, PFDoDA, PFTrDA PFTeDA, PFBS, PFPeS, PFHXS, PFHpS, PFOS, PFNS, PFDS, 4:2 FTS; 6:2 FTS, 8:2 FTS, GenX, ADONA	Korea	<LOQ-479000 (concentration range, industrial wastewater) nd-5560 (concentration range, municipal wastewater)	<LOQ-229000 (concentration range, industrial wastewater) nd-4870 (concentration range, municipal wastewater)	2018	Kim et al., 2021
PFBA, PFPeA, PFHxA, PFHpA, PFOA, PFNA, PFDA, PFUnDA, PFDoDA, HFPO-DA, ADONA, PF4OPeA, PF5OHxA, PFO2HpA, PFBS, PFPeS, PFHxS, PFHpS, PFOS, FTS, 6:2 and 8:2 Cl-PFESA, PFEESA	Sudan, Tanzania, Kenya, Africa	nd-15.6 (concentration range, mean value)	nd-22.5 (concentration range, mean value)	2015-2016	Jiang et al., 2023
PFPrA, PFBA, PFPeA, PFHxA, PFHpA, PFOA, PFNA, PFDA, PFUnDA, PFDoDA, PFTrDA, PFTeDA, PFPrS, PFBS, PFPeS, PFHxS, PFHpS, PFOS, PFNS, PFDS, 4:2 FTS, 6:2 FTS, 8:2 FTS; 6:2 FTCA, 5:3 FTCA; 6:2 FTUCA, FBSA, FHxSA, PFOSA, N-MeFOSAA, N-Et-FOSAA, HFPO-DA, ADONA, 19Cl-PF3OUdS, PFEESA, PF4OPeA, PF5OHxA, 3,6-OPFHpA, 6:2 diPAP; 6:2/8:2 diPAP, PFHxPA	Southeast United States	31.1-186.0 (total PFAS concentration)	171.4-266.6 (total PFAS concentration)	2020-2021	Kim et al., 2022

PFBA, PFPeA, PFHxA, PFHpA, PFOA, PFNA, PFDA, PFUnDA, PFDoDA, PFTrA, PFTeA, PFBS, PFPeS, PFHxS, PFHpS, PFOS, PFDS, 6:2 FTS, 8:2 FTS, 6:2 Cl-PFESA; 8:2 Cl-PFESA	Australia	9.27-412 (total PFAS concentration)	34.1-520 (total PFAS concentration)	2017	Cogann et al., 2022
PFODA, PFOA, 6:2 FTS; PFHxDA, PFOA, PFPeA, PFTrDA, FOSA, PFBA, PFDoDA, NEtFOFAA, PFBS, PFUdA, NMeFOSAA, 4:2 FTS; PFDA, PFHpA; PFPrOPrA; PFDS, PFHpS, NaDONA; PFNA; PFHxA; PFNS; PFHxS	Michgan, USA	50-1114 (total PFAS concentration, mean value)	42-859 (total PFAS concentration, mean value)	2018-2020	Helmer et al., 2022

perfluoropropanoic acid (PFPrA); perfluorobutanoic acid (PFBA); perfluoropentanoic acid (PFPeA); perfluoro-butane sulfonamide (FBSA), perfluorohexanoic acid (PFHxA); perfluoroheptanoic acid (PFHpA); perfluorooctanoic acid (PFOA); perfluoro-n-octadecanoic acid (PFODA); perfluorononanoic acid (PFNA); perfluorodecanoic acid (PFDA); perfluorohexadecanoic acid (PFHxDA) perfluoroundecanoic acid (PFUnDA); perfluorododecanoic acid (PFDoDA); perfluorortridecanoic acid (PFTrDA); perfluorotetradecanoic acid (PFTeDA); perfluoropropane sulfonic acid (PFPrS); perfluorobutane sulfonic acid (PFBS); perfluorononanesulfonic acid (PFNS); Perfluorodecanesulfonic acid (*PFDS*); perfluoropentane sulfonic acid (PFPeS); perfluorohexane sulfonic acid (PFHxS); fluorohexane sulphonamide (FHxSA); perfluorheptane sulfonic acid (PFHpS), perfluorooctanesulfonic acid (PFOS); perfluorononane sulfonic acid (PFNS); perfluorodecane sulfonic acid (PFDS), 4:2 fluorotelomer sulfonic acid (4:2 FTS); 6:2 fluorotelomer sulfonic acid (6:2 FTS); 8:2 fluorotelomer sulfonic acid (8:2 FTS); 6:2 fluorotelomer carboxylic acid (6:2 FTCA); 8:2 fluorotelomer carboxylic acid (8:2 FTCA); 10:2 fluorotelomer carboxylic acid (10:2 FTCA); 5:3 fluorotelomer carboxylic acid (5:3 FTCA); 6:2 fluorotelomer unsaturated carboxylic acid (6:2 FTUCA); 8:2 fluorotelomer unsaturated carboxylic acid (8:2 FTUCA); 10:2 fluorotelomer unsaturated carboxylic acid (10:2 FTUCA); perfluorobutane sulfonamide (FBSA); perfluorohexane sulfonamide (FHxSA); perfluorooctane sulfonamide (PFOSA); N-methyl-perfluoro-1-octanesulfonamidoacetic acid (N-MeFOSAA); N-ethyl-perfluoro-1-octanesulfonamidoacetic acid (N-EtFOSAA); hexafluoropropylene oxide-dimer acid (HFPO-DA); 4,8-dioxa-3H-perfluorononanoic acid (ADONA); sodium salt of ADONA (NaDONA); 19-chloroeicosafluoro-3-oxaundecane-1-sulfonic acid (19Cl-PF3OUdS) perfluoro(2-ethoxyethane)sulfonic acid (PFEESA); perfluoro-3-methoxypropanoic acid (PF4OPeA); perfluoro-4-methoxybutanoic acid (PF5OHxA); nonafluoro-3,6-dioxaheptanoic acid (3,6-OPFHpA); 6:2 fluorotelomer phosphate diester (6:2 diPAP); 6:2/8:2 fluorotelomer phosphate diester (6:2/8:2 diPAP); perfluorohexylphosphonic acid (PFHxPA); perfluoro-3,6-dioxaheptanoic acid (PFO2HpA); fluorotelomer sulfonamide propyl N,N-dimethylamine (6:2 FTSaAM); 4:2 fluorotelomer sulfonamide betaines (4:2 FTAB) 6:2 fluorotelomer sulfonamide betaines (6:2 FTAB), 8:2 fluorotelomer sulfonamide betaines (8:2 FTAB); fluorotelometer sulphonate anion (4:2 FTSA; 6:2 FTSA; 8:2 FTSA); chlorinated polyfluoroalkyl ether sulfonic acids (6:2 and 8:2 Cl-PFESA), perfluoro (2-ethoxyethane) sulfonate (PFEESA), , 9-chlorohexadecafluoro-3-oxanonane-1-sulfonate (6:2 Cl-PFESA), perfluorohexadenoic acid (PFHxDA), perfluoro-2-propoxypropionic acid (PFPrOPrA); hexafluoropropylene oxide-dimer acid (HFPO-DA); perfluorophosphonic acid (PFPA); perfluorononanesulfonic acid (PFNS), 2,3,3,3-tetrafluoro-2-(eptafluoropropossi)-propanoato di ammonio (GenX).

nd=not detected

Figure S1. Removal of PFAS (expressed in percentage) from the four WWTPs (A, B, C, D) during July and October 2021, May and February 2022.

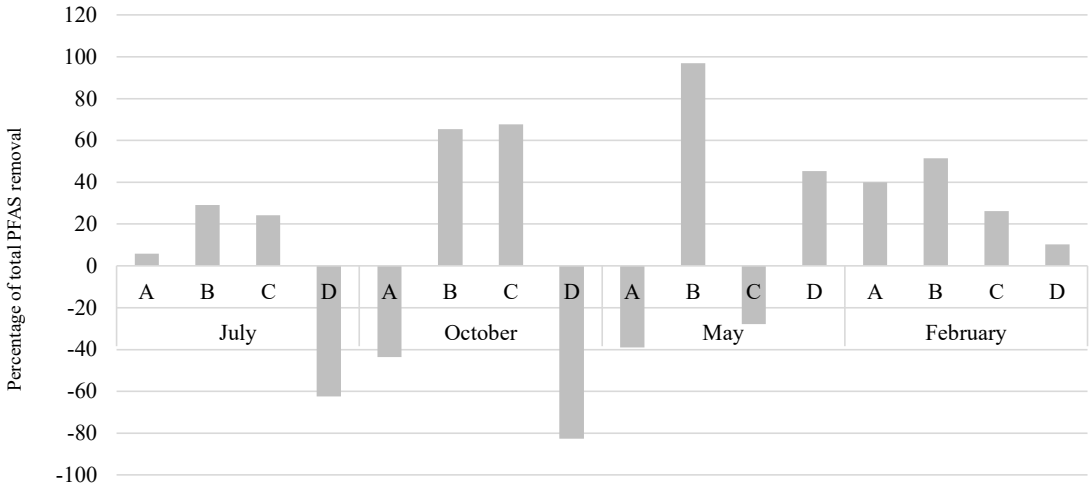
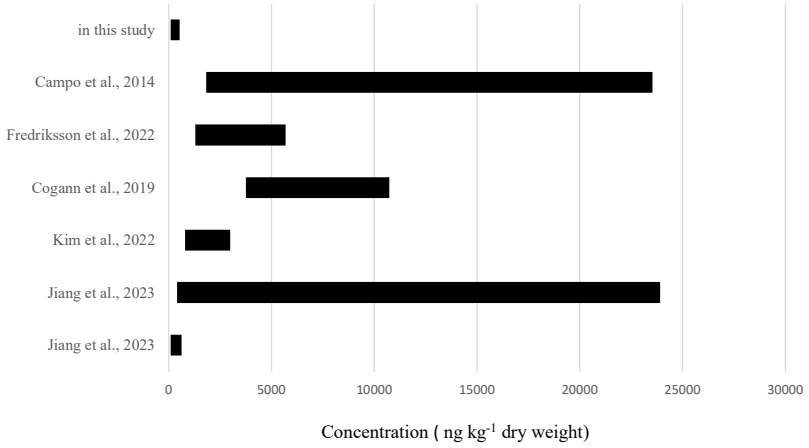


Figure S2. Comparison of PFOA levels found in sludge wastewater samples collected from Plant A with previous studies.


in this study
Campo et al., 2014
Fredriksson et al., 2022
Cogann et al., 2019
Kim et al., 2022
Jiang et al., 2023
Jiang et al., 2023
0
5000
10000
15000
20000
25000
30000
Concentration (ng kg-1 dry weight)