Supplemental Material for:

Characteristics of Legacy and Emerging Per- and Polyfluoroalkyl Substances in Atmospheric Total Suspended Particulate from The Coastal Areas in China

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Text S1. Sample Preparation

Cut the quartz membrane with particles attached into thin strips about 0.5 cm wide, put into a 50 mL polypropylene centrifuge tube(PP tube), add 2 ng of mixed internal standards (PFAC-MXA), vortex for 30 s, and let stand overnight. The samples were extracted with 25 mL 0.1% NH_4OH/methanol in a sonication water bath for 30 min, centrifuged at 4000 r/min for 10 min and collecting the supernatant into new PP tubes. 10 mL 0.1% NH_4OH/methanol was added to the remaining part and the extraction procedure repeated. The two extracts were combined and evaporated to 5 mL under a gentle stream of dry nitrogen gas. The concentrated extracts was purified by Cleanert PestiCarb SPE cartridges (made of graphitized carbon, 500 mg/6 mL, Bonna-Angla Technologies, China). The PestiCarb cartridges were activated with 5 mL methanol, 5 mL of ultrapure water, and 5 mL of methanol at a rate of 1-2 drop(s) per second, respectively. The sample extracts were cleaned up with activated PestiCarb cartridges, the effluent was collected and eluted with 5 ml 0.1% NH_4OH/methanol. The combined eluates (~10 mL) were evaporated to 0.5 ml under a gentle stream of dry nitrogen gas, filtered through a 0.22μm nylonfilter and transferred into a injection vial, and finally stored at 4 °C for analysis.

Text S2. Instruments Analysis for target PFASs analysis

PFASs were determined by ultra-high performance liquid chromatography tandem triple quadrupole mass spectrometer instrument (Waters, ACQUITY-UPLC/XEVO-TQS) with an electrospray ionization (ESI) source. A 5 µL volume of extracted sample was injected on a ZORBAX Eclipse Plus C18 column (100 × 2.1 mm, 1.8-Micron), which was maintained at 55 °C. 5 mM ammonium acetate solution(A) and methanol(B) were used as the mobile phase, the flow rate was 0.4 mL/min. The gradient elution procedure was as follows: initial composition was 85% A and 15% B, which was held for 1 min, then A decreased to 45% between 1-1.5 min, then decreased to 30% in four minutes, then decreased to 20% in 1.5 minutes, then decreased to 0% between 7-12 min and was held for 2.4 min, then increased to 85% in 0.5 minutes finally. The mass spectrometer was operated in the negative electrospray ionization multiple reaction monitoring (MRM) mode with the following MS parameters: electrospray voltage 3000 V; nebulizer gas flow at 15psi; gas flow of 300 L/h; sheath gas flow of 660 L/h. sheath gas temperature was 350 °C. Other parameters such as precursor/product ions and collsion energy (CE) were shown in Table S3.

Table S1 The date, time, volume and meteorological parameters during the sampling campaign in Laoshan.

Date	Number	Time	Volume[a]	Weather	AQI
		36.15°N，120.68°E			
20210416	LS-01	7:53~7:58	field blank	cloudy	194
20210417	LS-02	8:03~7:34 (next day)	440.97	sunny	72
20210418	LS-03	8:00~7:30 (next day)	434.46	sunny	37
20210419	LS-04	8:00~7:30 (next day)	434.46	sunny	52
20210420	LS-05	8:05~7:40 (next day)	448.73	overcast	58
20210421	LS-06	8:00~7:35 (next day)	461.78	rainy	41
20210422	LS-07	8:00~11:15	60.04	rainy	36
20210423	LS-08	8:10~7:38 (next day)	420.55	sunny	37
20210425	LS-09	8:17~7:40 (next day)	431.99	sunny	59
20210426	LS-10	8:05~17:42	196.14	rainy	69
20210427	LS-11	8:25~13:50	110.99	cloudy	68
20210428	LS-12	8:12~21:12	254.55	cloudy	108
20210429	LS-13	8:16~7:30 (next day)	134.15	rainy	69
20210430	LS-14	8:06~7:55 (next day)	453.18	rainy	42
20210501	LS-15	8:09~8:15 (next day)	458.57	sunny	45
20210502	LS-16	8:30~7:40 (next day)	440.82	sunny	34
20210503	LS-17	8:15~21:00	235.55	rainy	41
20210505	LS-18	8:11~8:00 (next day)	446.77	sunny	74
20210506	LS-19	8:22~8:00 (next day)	449.68	cloudy	107
20210508	LS-20	20:16~8:55 (next day)	233.70	cloudy	59
20210509	LS-21	9:06~22:12	242.02	overcast	44
20210510	LS-22	13:36~7:39 (next day)	333.46	overcast	45
20210512	LS-23	15:37~8:10 (next day)	318.09	sunny	33
20210513	LS-24	8:22~7:14 (next day)	447.76	sunny	36
20210514	LS-25	7:25~13:18	108.69	rainy	34
20210515	LS-26	9:30~19:13	195.65	rainy	26
20210516	LS-27	20:18~7:58 (next day)	215.54	overcast	26

a: the total volume of each sample at normal atmospheric pressure, m^3

Table S2 The date, time, volume, position and type of Xisha Islands samples.

Date	Number	Time	Total time	Volume[a]	Position (start-end)		Type
	field blank2	13:45~13:50			Tanmen Port		Blank
20210305	XS-01	14:00~18:30	4h30min	89.06	Tanmen Port	**N** 18°46.461‘ **E** 110°57.244’	Day
	XS-02	18:50~6:30 (next day)	11h40min	230.90	**N** 18°46.461‘ **E** 110°57.244’	**N** 17°21.686‘ **E** 111°53.490’	Night
20210306	XS-03	6:50~11:50 12:00~18:30	11h30min	227.60	**N** 17°21.686‘ **E** 111°53.490’	**N** 16°50.306‘ **E** 112°19.643’	Day
20210307	XS-04	6:52~18:30	11h38min	230.24	**N** 16°50.306‘ **E** 112°19.643’	**N** 16°58.596‘ **E** 112°16.065’	Day
	XS-05	18:40~6:33 (next day)	11h53min	238.18	**N** 16°58.596‘ **E** 112°16.065’		Night
20210308	XS-06	6:47~12:54	6h07min	122.60	**N** 16°58.294‘ **E** 112°16.051’	**N** 16°28.483‘ **E** 111°44.193’	Day
	XS-07	18:40~6:40 (next day)	12h	237.49	**N** 16°28.483‘ **E** 111°44.193’		Night
20210309	XS-08	19:00~22:15	3h15min	64.32	**N** 16°28.133‘ **E** 111°43.974‘		Night
20210310	XS-09	7:00~9:40 16:30~18:30	4h40min	92.36	**N** 16°28.133‘ **E** 111°43.974’	**N** 16°28.531‘ **E** 111°43.581’	Day
	XS-10	19:00~6:30 (next day)	11h30min	227.60	**N** 16°28.531‘ **E** 111°43.581’		Night
	field blank3	18:40~18:45			**N** 16°28.456‘ **E** 111°44.181’		Blank
20210311	XS-11	7:00~18:30	11h30min	227.60	**N** 16°28.531 **E** 111°43.581’	**N** 16°28.456‘ **E** 111°44.181’	Day
	XS-12	19:00~6:30 (next day)	11h30min	227.60	**N** 16°28.456‘ **E** 111°44.181’		Night
20210312	XS-13	7:00~11:15 11:35~16:36	9h16min	183.40	**N** 16°28.456‘ **E** 111°44.181’	**N** 16°30.358‘ **E** 111°36.150’	Day
	XS-14	19:00~6:30 (next day)	11h30min	227.60	**N** 16°30.358‘ **E** 111°36.150’		Night
20210313	XS-15	7:00~16:57	9h57min	196.92	**N** 16°30.358‘ **E** 111°36.150’	**N** 16°28.029‘ **E** 111°43.894’	Day
	XS-16	19:00~6:30 (next day)	11h30min	230.50	**N** 16°28.029‘ **E** 111°43.894’		Night
202103014	XS-17	7:00~13:28	6h28min	127.98	**N** 16°28.029‘ **E** 111°43.894’	**N** 16°30.496‘ **E** 111°36.274’	Day
	XS-18	19:00~6:30 (next day)	11h30min	230.50	**N** 16°30.496‘ **E** 111°36.274’		Night
20210315	XS-19	7:00~16:12; 16:25~18:30	11h17min	223.31	**N** 16°30.496‘ **E** 111°36.274’	**N** 16°28.126‘ **E** 111°43.623’	Day

	XS-20	19:00~6:30(next day)	11h30min	227.60	**N** 16°28.126‘ **E** 111°43.623’		night
20210316	XS-21	7:00~7:52; 16:31~18:30	2h51min	56.40	**N** 16°28.126‘ **E** 111°43.623’	**N** 17°15.138‘ **E** 111°22.538’	Day
	XS-22	19:00~6:30 (next day)	11h30min	230.50	**N** 17°18.861‘ **E** 111°21.033’	**N** 18°46.438‘ **E** 110°51.676’	Night
20210317	XS-23	7:00~13:00	4h	88.29	**N** 18°50.321‘ **E** 110°49.943’	**N** 19°14.248‘ **E** 110°37.151’	Day

a: the total volume of each sample at normal atmospheric pressure, m^3

Table S3 List of PFASs monitored in current study and the optimized UPLC-MS/MS parameters

Compound Group	Compound name	Parent Ions	Product Ions Monitored[m/z]	RT [min]	Fragmentor [V]	Collision energy [V]
Perfluoroalkyl carboxylic acid (PFCAs)	Perfluorobutanoic acid (PFBA)	213	169	3.05	60	4
	Perfluoropetanoic acid (PFPeA)	263	219	3.5	60	4
	Perfluorohexanoic acid (PFHxA)	313	269,119	4.02	80	4,24
	Perfluoroheptanoic acid (PFHpA)	363	319,169	4.71	80	4,16
	Perfluorooctanoic acid(PFOA)	413	369,219,169	5.56	80	8,16,20
	Perfluorononanoic acid (PFNA)	463	419,219,169	6.48	80	8,16,20
	Perfluorodecanoic acid (PFDA)	513	469,269,219	7.38	80	8,16,16
	Perfluoroundecanoic acid (PFUnDA)	563	519319,269	8.06	80	8,16,16
	Perfluorododecanoic acid (PFDoDA)	613	569,319,269,169	8.57	110	8,20,20,28
	Perfluorotridecanoic acid(PFTrDA)	663	619,319,269,169	9	110	8,20,24,32
	Perfluorotetradecanoic acid(PFTeDA)	712.9	669,369,219,169	9.4	110	12,20,24,32
Perfluoroalkyl sulfonic acids (PFSAs)	Potassium perfluoro-1-butanesulfonate (PFBS)	298.9	99,80	3.57	110	32,44
	Sodium perfluoro-1-pentanesulfonate(PFPeS)	348.9	99,80	4.07	120	44,44
	Potassium perfluorohexane sulfonate(PFHxS)	398.9	99,80	4.75	120	40,52
	Sodium perfluoro-1-heptanesulfonate(PFHpS)	448.9	99,80	5.59	140	49,59
	Potassium perfluorooctane sulfonate(PFOS)	498.9	99,80	6.5	130	48,53
	Sodium perfluoro-1-nonanesulfonate(PFNS)	548.9	99,80	7.38	150	58,63
	Sodium perfluoro-1-decanesulfonate(PFDS)	598.9	99,80	8.04	150	64,68
FASA	Perfluoro-1-octanesulfonamide(FOSA)	497.9	169,78,48	8.1	121	110
Fluorinated telomer sulfonates (n:2 FTSs)	Sodium 1H,1H,2H,2H-perfluorohexanesulfonate(4-2 FTS)	327	307,81,80	3.97	140	20,32,45
	Sodium 1H,1H,2H,2H-perfluorooctanesulfonate(6-2 FTS)	427	407,81,80	5.52	140	28,44,49
	Sodium 1H,1H,2H,2H-perfluorodecanesulfonate(8-2 FTS)	527	507,81,80	7.36	170	32,48,59

FASAA	N-ethylperfluorooctanesulfonamidoacetic acid(N-EtFOSAA)	584	526,482.9,419	8.08	130	20,16,20
	N-methylperfluorooctanesulfonamidoacetic acid(N-MeFOSAA)	570	512,482.9,419	7.74	140	24,16,20
Polyfluoroalkyl ether carboxylic acids (PFECAs)	2,3,3,3-Tetrafluoro-2-(1,1,2,2,3,3,3-heptafluoropropoxy)propanoic acid(HFPO-DA)	285	185,169,119	4.21	60	16,4,32
	Sodium dodecafluoro-3H-4,8-dioxanonanoate(ADONA)	377	251,85	4.82	80	8,40
Perfluoroalkyl ether sulfonic acid（Cl-PFESA）	Potassium 9-chlorohexadecafluoro-3-oxanonane-1-sulfonate(6:2 Cl-PFESA)	530.9	350.9,83	7.04	130	28,32
	Potassium 11-chloroeicosafluoro-3-oxaundecane-1-sulfonate(8:2 Cl-PFESA)	630.9	450.9,83	8.36	142	32,36
FASA	Perfluoro-1-butanesulfonamide(FBSA)	298	78,64,48	4.29	110	32,88,85
	Perfluoro-1-hexanesulfonamide(FHxSA)	397.9	78.64.48	6.19	130	35.92.90
Internal Standards	Perfluoro-n-[$^{13}C_4$]butanoic acid($^{13}C_4$-PFBA)	217	172	3.05	60	8
	Perfluoro-n-[$^{13}C_5$]pentanoic acid($^{13}C_5$-PFPeA)	268	223	3.5	60	4
	Perfluoro-n-[1,2,3,4,6-$^{13}C_5$]hexanoic acid($^{13}C_5$-PFHxA)	318	273,120	4.02	80	4,24
	Perfluoro-n-[$^{13}C_4$]heptanoic acid($^{13}C_4$-PFHpA)	367	322,169	4.71	80	8,16
	Perfluoro-n-[$^{13}C_8$]octanoic acid($^{13}C_8$-PFOA)	421	376,172	5.56	90	8,16
	Perfluoro-n-[$^{13}C_9$]nonanoic acid($^{13}C_9$-PFNA)	472	427,223	6.48	80	8,16
	Perfluoro-n-[1,2,3,4,5,6-$^{13}C_6$]decanoic acid($^{13}C_6$-PFDA)	519	474,219	7.38	80	8,16
	Perfluoro-n-[1,2,3,4,5,6,7-$^{13}C_7$]undecanoic acid($^{13}C_7$-PFUnDA)	570	525,270	8.06	80	8,20
	Perfluoro-n-[1,2-$^{13}C_2$]dodecanoic acid($^{13}C_2$-PFDoDA)	615	570,269	8.57	110	8,20
	Perfluoro-n-[1,2-$^{13}C_2$]tetradecanoic acid($^{13}C_2$-PFTeDA)	715	670,169	9.4	110	12,32
	N-methyl-d_3-perfluoro-1-octanesulfonamidoacetic acid($^{2}H_3$-N-MeFOSAA)	573	482.9,419	7.73	140	16,20
	N-ethyl-d_5-perfluoro-1-octanesulfonamidoacetic acid($^{2}H_5$-N-EtFOSAA)	589	531,419	8.07	130	20,20
	Sodium perfluoro-1-(2,3,4-$^{13}C_3$)butanesulfonate($^{13}C_3$-PFBS)	302	99,80	3.57	140	36,44
	Sodium perfluoro-1-(1,2,3-$^{13}C_3$)hexanesulfonate($^{13}C_3$-PFHxS)	402	99,80	4.75	170	48,60
	Sodium perfluoro-1-($^{13}C_8$)octanesulfonate($^{13}C_4$-PFOS)	507	99,80	6.49	130	48,53
	Sodium 1H,1H,2H,2H-perfluoro-1-(1,2-$^{13}C_2$)hexanesulfonate($^{13}C_2$-4-2 FTS)	329	309,81	3.97	140	20,40
	Sodium 1H,1H,2H,2H-perfluoro-1-(1,2-$^{13}C_2$)octanesulfonate($^{13}C_2$-6-2 FTS)	429	409,81	5.52	170	24,48

Sodium 1H,1H,2H,2H-perfluoro-1-(1,2-$^{13}C_2$)decanesulfonate($^{13}C_2$-8-2 FTS)	529	509,81	7.37	170	32,56
Perfluoro-1-($^{13}C_8$)octanesulfonamide($^{13}C_8$-FOSA)	506	78	8.1	121	44

Table S4 The method detection limits (MDLs) and recoveries of target compounds

Target compounds	Internal Standard	MDLs ($pg\cdot m^{-3}$)	Recovery (%, Avg±SD)
PFBA	MPFBA	0.009	93.1±0.9
PFPeA	MPFPeA	0.023	94.2±0.9
PFHxA	MPFHxA	0.060	93.0±0.4
PFHpA	MPFHpA	0.044	94.7±0.3
PFOA	MPFOA	0.046	92.5±0.6
PFNA	MPFNA	0.144	90.5±0.2
PFDA	MPFDA	0.062	90.9±0.4
PFUnDA	MPFUnDA	0.069	92.9±0.5
PFDoDA	MPFDoDA	0.071	93.2±0.2
PFTrDA	MPFDoDA	0.061	118.4±0.7
PFTeDA	MPFTeDA	0.313	94.0±0.4
PFBS	MPFBS	0.093	94.9±0.4
PFPeS	MPFBS	0.115	91.7±0.6
PFHxS	MPFHxS	0.197	93.1±0.5
PFHpS	MPFHxS	0.092	96.6±0.4
PFOS	MPFOS	0.381	93.6±0.7
PFNS	MPFOS	0.222	101±5.6
PFDS	MPFOS	0.350	90.2±0.2
N-MeFOSAA	d3-N-MeFOSAA	0.841	95.1±0.5
N-EtFOSAA	d3-N-EtFOSAA	0.449	94.2±1.7
4:2 FTS	M4:2 FTS	0.515	128.4±0.3
6:2 FTS	M6:2 FTS	0.012	124.3±0.5
8:2 FTS	M8:2 FTS	0.108	126.8±0.4
FBSA	MPFBS	0.089	93.6±0.3
FHxSA	MPFOS	0.056	98.3±1.5
PFOSA	MPFOSA	0.069	90.8±0.1
6:2 Cl-PFESA	/	0.018	72.1±1.5
8:2 Cl-PFESA	/	0.102	69.3±1.5
HFPO-DA	/	0.128	73.0±1.7
ADONA	/	0.052	88.4±3.0

Table S5 Concentrations of 30 legacy and emerging PFASs (19 PFAS were detected) in Laoshan atmosphere (pg/m^3)

	PFBA	PFHxA	PFHpA	PFOA	PFNA	PFDA	PFUnDA	PFDoDA	PFTrDA	PFTeDA	PFBS	PFPeS	PFHxS	PFHpS	PFOS	HFPO-DA	6:2 Cl-PFESA	6-2 FTSA	PFOSA	ΣPFASs
20210417	0.92	0.93	0.15	24.5	0.33	0.08	0.17	<0.071	n.d.	n.d.	0.64	0.43	<0.197	n.d.	0.81	<0.128	n.d.	<0.012	<0.069	29.3
20210418	2.02	2.02	2.01	29.5	0.60	0.39	0.24	0.13	0.41	n.d.	n.d.	0.66	0.24	0.092	2.63	0.24	n.d.	<0.012	<0.069	40.9
20210419	4.52	3.85	6.62	48.0	1.57	1.53	1.14	0.87	1.71	1.77	0.92	0.30	0.41	0.166	5.34	0.15	0.84	0.33	<0.069	80.1
20210420	n.d.	0.63	0.69	10.4	0.54	0.14	0.20	0.09	0.37	1.05	n.d.	0.43	0.27	n.d.	0.59	<0.128	n.d.	<0.012	<0.069	15.6
20210421	0.65	1.09	1.44	7.72	0.78	0.41	0.45	0.26	0.65	1.11	0.60	0.30	0.21	n.d.	0.91	n.d.	n.d.	<0.012	<0.069	16.6
20210422	2.71	1.83	n.d.	12.3	2.32	0.22	1.31	0.46	n.d.	n.d.	4.58	n.d.	1.15	n.d.	1.69	n.d.	n.d.	0.04	n.d.	28.5
20210423	0.72	0.43	<0.044	9.10	1.12	0.14	0.39	0.09	0.43	n.d.	0.71	n.d.	<0.197	n.d.	0.31	n.d.	n.d.	<0.012	<0.069	13.7
20210425	1.60	1.29	0.98	20.4	1.10	0.33	0.36	0.19	0.51	1.14	0.84	0.69	0.37	n.d.	2.19	n.d.	0.86	<0.012	<0.069	32.9
20210426	n.d.	0.87	0.49	15.3	1.11	0.20	0.45	0.19	0.85	n.d.	1.61	1.89	0.46	n.d.	1.12	n.d.	1.12	<0.012	<0.069	25.6
20210427	2.47	1.67	0.31	11.6	3.33	0.45	1.26	0.38	1.63	n.d.	n.d.	n.d.	0.61	n.d.	1.67	n.d.	n.d.	0.02	0.10	25.5
20210428	5.56	1.82	2.88	28.2	1.08	0.35	0.50	0.23	0.75	1.90	n.d.	0.31	0.29	n.d.	1.56	0.62	0.94	3.07	<0.069	50.0
20210429	4.08	2.24	4.15	46.6	1.99	0.63	0.77	0.38	1.35	3.60	n.d.	3.60	0.71	n.d.	4.45	0.95	n.d.	0.02	0.09	75.6
20210430	1.24	0.37	0.11	8.82	0.46	<0.062	0.16	<0.071	n.d.	n.d.	n.d.	0.19	0.24	n.d.	0.51	n.d.	n.d.	<0.012	<0.069	12.3
20210501	0.86	0.49	0.34	11.0	0.45	0.11	0.18	0.08	0.34	n.d.	n.d.	1.29	0.23	n.d.	1.08	n.d.	n.d.	n.d.	<0.069	16.5
20210502	n.d.	0.59	0.07	5.60	0.38	0.10	0.16	<0.071	0.34	n.d.	n.d.	0.34	0.21	n.d.	0.49	n.d.	0.50	<0.012	<0.069	8.89
20210503	n.d.	n.d.	0.11	13.6	0.89	0.09	0.30	0.10	0.62	n.d.	n.d.	2.64	0.53	n.d.	0.68	n.d.	0.89	<0.012	<0.069	20.5
20210505	1.06	1.24	1.45	20.5	0.79	0.35	0.29	0.16	0.46	1.10	n.d.	0.79	0.29	n.d.	2.69	0.17	n.d.	<0.012	<0.069	31.3
20210506	2.58	0.66	1.41	37.2	1.65	0.92	0.62	0.51	0.93	1.47	0.83	n.d.	0.51	0.284	5.37	n.d.	0.81	1.13	<0.069	57.0
20210508	0.72	0.52	n.d.	6.12	0.49	n.d.	0.26	n.d.	n.d.	n.d.	n.d.	0.34	0.38	n.d.	0.63	<0.128	1.03	<0.012	n.d.	10.6
20210509	0.56	0.47	n.d.	3.18	0.42	<0.062	0.25	n.d.	n.d.	n.d.	n.d.	n.d.	0.29	n.d.	0.39	n.d.	n.d.	n.d.	<0.069	5.65
20210510	1.76	1.08	1.10	34.4	1.47	0.31	0.47	0.17	0.61	n.d.	0.86	<0.115	0.52	n.d.	0.79	0.72	n.d.	<0.012	<0.069	44.4
20210512	0.51	0.80	1.36	6.43	1.18	0.34	0.49	0.21	0.59	1.52	0.84	n.d.	0.33	n.d.	0.80	n.d.	n.d.	<0.012	<0.069	15.4
20210513	0.45	0.91	2.91	5.19	2.01	0.69	0.68	0.25	0.50	1.12	0.60	2.06	<0.197	n.d.	0.56	0.09	n.d.	<0.012	<0.069	18.2
20210514	1.73	3.00	2.54	5.98	2.11	0.49	1.02	0.37	n.d.	n.d.	2.56	0.39	0.74	n.d.	1.10	n.d.	n.d.	n.d.	n.d.	22.1

20210515	1.71	0.94	0.23	5.63	2.08	0.30	1.00	0.19	0.96	n.d.	n.d.	n.d.	0.38	n.d.	0.50	n.d.	n.d.	n.d.	<0.069	14.0
20210516	0.87	0.91	0.75	3.34	1.26	0.18	0.62	0.19	0.86	n.d.	n.d.	n.d.	0.34	n.d.	0.83	n.d.	n.d.	<0.012	<0.069	10.2

Table S6 Concentrations of 30 legacy and emerging PFASs (14 PFAS were detected) in Xisha Islands APM samples (pg/m^3)

	PFBA	PFHxA	PFHpA	PFOA	PFNA	PFDA	PFUnDA	PFDoDA	PFTrDA	PFPeS	PFHxS	PFOS	DONA	6-2 FTSA	ΣPFASs
20210305d[a]	n.d.	0.82	0.08	3.03	1.69	1.33	1.00	0.77	n.d.	n.d.	n.d.	0.43	n.d.	n.d.	9.15
20210305n[b]	0.50	0.37	<0.044	1.36	0.98	0.61	0.54	0.34	0.57	0.15	0.52	<0.381	n.d.	n.d.	6.36
20210306d	0.20	0.14	<0.044	0.92	0.64	0.47	0.33	0.22	n.d.	n.d.	0.26	<0.381	n.d.	n.d.	3.59
20210307d	n.d.	0.00	<0.044	1.24	0.78	0.57	0.37	0.25	0.47	<0.115	0.50	<0.381	n.d.	0.01	4.68
20210307n	0.46	0.37	<0.044	1.14	0.78	0.54	0.45	0.28	0.54	n.d.	0.55	<0.381	n.d.	0.01	5.53
20210308d	1.00	0.67	0.06	1.69	1.49	1.01	0.87	0.49	n.d.	n.d.	n.d.	0.54	n.d.	n.d.	7.82
20210308n	0.49	0.41	<0.044	1.39	1.16	0.62	0.60	0.31	0.48	0.12	0.52	0.60	n.d.	0.01	6.74
20210309n	1.84	1.26	0.12	4.42	2.54	1.60	1.22	0.79	1.62	n.d.	2.05	0.78	n.d.	n.d.	18.2
20210310d	1.09	0.98	0.08	2.40	1.99	1.14	0.96	0.55	0.00	n.d.	1.26	0.96	n.d.	n.d.	11.4
20210310n	0.50	0.48	<0.044	1.41	1.16	0.60	0.53	0.28	0.57	n.d.	0.58	0.41	n.d.	0.01	6.57
20210311d	0.55	0.53	<0.044	1.26	1.24	0.61	0.56	0.28	0.59	n.d.	0.54	<0.381	n.d.	n.d.	6.58
20210311n	0.48	0.40	<0.044	1.10	1.22	0.58	0.49	0.26	0.59	n.d.	0.52	<0.381	n.d.	n.d.	6.06
20210312d	0.60	0.27	0.04	1.18	1.10	0.59	0.45	0.39	0.35	<0.115	0.31	<0.381	n.d.	n.d.	5.74
20210312n	0.47	0.38	<0.044	1.17	1.64	0.65	0.66	0.30	0.63	<0.115	0.54	<0.381	n.d.	n.d.	6.93
20210313d	0.60	0.61	0.14	1.58	1.93	0.78	0.67	0.34	0.68	n.d.	0.62	0.87	n.d.	0.02	8.84
20210313n	0.57	0.68	0.15	1.99	1.70	0.88	0.85	0.42	0.86	n.d.	0.57	0.76	n.d.	0.01	9.45
20210314d	0.96	0.88	<0.044	3.64	2.56	1.56	1.63	0.82	1.95	0.15	0.97	0.60	n.d.	0.02	15.8
20210314n	0.47	0.45	<0.044	2.07	1.75	1.25	1.28	0.65	1.36	n.d.	0.52	0.61	n.d.	0.01	10.5
20210315d	0.47	0.40	<0.044	1.46	1.08	0.71	0.70	0.35	0.54	<0.115	0.54	<0.381	n.d.	<0.012	6.75
20210315n	0.46	0.46	<0.044	1.57	1.58	0.87	0.92	0.43	0.94	<0.115	0.52	<0.381	1.19	0.01	9.44
20210316d	1.86	0.78	0.13	2.53	1.88	1.62	1.10	0.43	n.d.	0.34	2.06	<0.381	4.26	0.03	17.4
20210316n	0.45	0.41	<0.044	0.96	0.92	0.58	0.51	0.29	0.57	<0.115	n.d.	<0.381	n.d.	0.01	5.21
20210317d	n.d.	0.31	0.08	1.57	1.06	1.04	1.27	0.26	n.d.	0.22	0.66	n.d.	1.57	0.04	8.07

[a] d is present sampling in daytime, [b] n is present sampling in night.

Table S7 Pearson rank correlations between the PFASs components in Laoshan APM samples

Laoshan(n=26)	PFHxA	PFHpA	PFOA	PFNA	PFDA	PFUnDA	PFDoDA	PFTrDA	PFTeDA	PFBS	PFPeS	PFHxS	PFHpS	PFOS	HFPO-DA	6:2 Cl-PFESA	6:2 FTSA	PFOSA
PFBA	.715**	.658**	.703**	.435*	.596**	.529**	.657**	.496**	.556**	.126	-.010	.374	.364	.657**	.588**	.174	.646**	.152
PFHxA		.800**	.540**	.471*	.716**	.647**	.770**	.419*	.387	.359	.006	.417*	.265	.591**	.329	.016	.166	-.048
PFHpA			.665**	.272	.866**	.390*	.730**	.547**	.709**	.023	.276	.084	.403*	.686**	.475*	.139	.283	.137
PFOA				.134	.648**	.160	.565**	.513**	.600**	-.015	.266	.200	.570**	.832**	.668**	.265	.331	.293
PFNA					.467*	.916**	.639**	.554**	.171	.397*	.033	.644**	.106	.282	.117	-.142	.022	.205
PFDA						.560**	.901**	.670**	.574**	.127	.078	.172	.691**	.806**	.201	.181	.205	.185
PFUnDA							.786**	.517**	.157	.537**	-.094	.698**	.175	.348	.034	-.093	.042	-.001
PFDoDA								.646**	.467*	.427*	-.010	.508**	.595**	.757**	.126	.160	.191	.062
PFTrDA									.490*	-.227	.207	.142	.356	.580**	.292	.201	.177	.671**
PFTeDA										-.131	.415*	.066	.259	.640**	.582**	.124	.371	.342
PFBS											-.144	.704**	.038	.065	-.165	-.021	-.086	-.579*
PFPeS												.108	-.159	.187	.391*	.167	-.127	.269
PFHxS													.063	.297	.165	.044	-.042	-.245
PFHpS														.773**	-.051	.325	.272	.033
PFOS															.328	.277	.248	.226
HFPO-DA																-.024	.354	.281
6:2 Cl-PFESA																	.424*	-.158
6:2 FTSA																		.028

*: Correlation is significant at the 0.05 level (2-tailed).

**: Correlation is significant at the 0.01 level (2-tailed).

Table S8 Pearson rank correlations between the PFASs components in Xisha Islands APM samples

Xisha(n=23)	PFHxA	PFHpA	PFOA	PFNA	PFDA	PFUnDA	PFDoDA	PFTrDA	PFPeS	PFHxS	PFOS	ADONA	6:2 FTSA
PFBA	.742**	.460	.613**	.660**	.633**	.421*	.445*	.254	.236	.816**	.468*	.418*	.034
PFHxA		.574*	.868**	.868**	.792**	.658**	.782**	.324	-.077	.606**	.629**	.114	-.044
PFHpA			.473*	.528**	.523*	.361	.284	-.075	.104	.493*	.464*	.386	.345
PFOA				.855**	.906**	.793**	.907**	.486*	.071	.635**	.464*	.138	.087
PFNA					.832**	.787**	.812**	.546**	.027	.599**	.635**	.139	.098
PFDA						.906**	.846**	.316	.302	.640**	.321	.441*	.321
PFUnDA							.784**	.444*	.298	.471*	.221	.318	.445*
PFDoDA								.492*	-.080	.342	.478*	-.038	-.084
PFTrDA									-.128	.274	.338	-.272	.001
PFPeS										.391	-.437*	.788**	.669**
PFHxS											.319	.541**	.292
PFOS												-.303	-.271
ADONA													.639**

*: Correlation is significant at the 0.05 level (2-tailed).

**: Correlation is significant at the 0.01 level (2-tailed).

Table S9 Source profiles of PFASs in Laoshan obtained from PCA-MLR models (*n*=26)

Species	KMO measure	Rotated Component Coefficients		
		F1	**F2**	**F3**
PFUnDA	.657	**0.976**	0.021	0.117
PFNA	.665	**0.93**	0.044	0.005
PFDoDA	.809	0.713	0.231	0.622
PFHxA	.760	0.629	0.454	0.316
PFTrDA	.892	0.533	0.35	0.381
HFPO-DA	.513	0.012	**0.938**	-0.136
PFTeDA	.657	0.108	0.754	0.319
PFOA	.695	0.064	0.73	0.566
PFBA	.860	0.466	0.674	0.286
PFHpA	.725	0.364	0.641	0.49
PFHpS	.608	0.041	0.004	**0.948**
PFOS	.774	0.233	0.458	**0.801**
PFDA	.757	0.503	0.331	0.745
Eigenvalue		7.36	1.98	1.39
% of Variance		56.7	15.2	10.7
Cumulative % of Variance		56.7	71.9	82.6
MLR results				
Possible sources		material intermediates preparation /fluoropolymer processing aids	fluoropolymer manufacturing	metal electroplating and electrochemical
Profile contributions		0.263	0.736	0.57
Source contributions (%)		16.8%	46.9%	36.3%

The total KMO test :.730;

Bartlett's test :.000;

The values with bold font represent the components with positive loading greater than 0.80.

Table S10 Source profiles of PFASs in Xisha Islands obtained from PCA-MLR models (n=23)

Species	KMO Measure	Rotated Component Coefficients		
		F1	**F2**	**F3**
PFDoDA	.642	**0.952**	0.149	-0.183
PFUnDA	.767	**0.895**	0.164	0.328
PFOA	.735	**0.867**	0.414	-0.012
PFDA	.773	**0.861**	0.398	0.252
PFNA	.903	0.786	0.521	-0.08
PFHxA	.848	0.706	0.608	-0.198
PFBA	.782	0.327	**0.822**	0.043
PFHxS	.633	0.291	0.798	0.3
PFHpA	.615	0.209	0.732	0.113
PFOS	.636	0.3	0.594	-0.589
PFPeS	.680	0.026	0.137	**0.904**
6:2 FTSA	.505	0.095	0.12	**0.864**
Eigenvalue		6.73	2.25	1.19
% of Variance		56.1	18.8	9.88
Cumulative % of Variance		56.1	74.9	84.8
MLR results				
Possible sources		textile treatment agents	precious metals	metal electroplating and electrochemical
Profile contributions		0.779	0.615	0.064
Source contributions (%)		53.4%	42.2%	4.39%

The total KMO test :.731;

Bartlett’s test :.000;

The values with bold font represent the components with positive loading greater than 0.80.

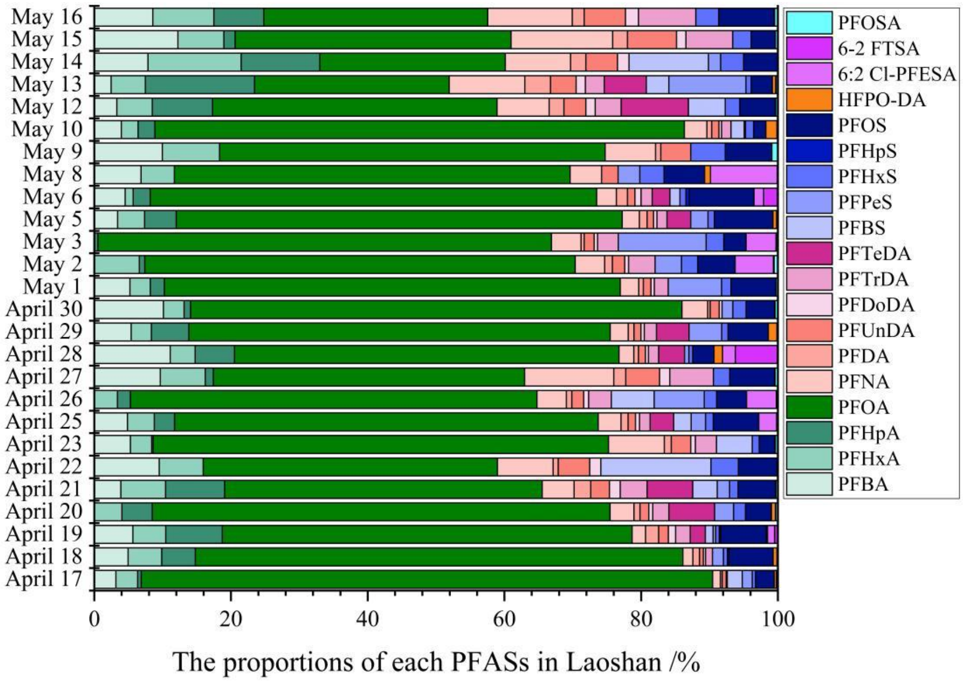

Fig S1. Profiles of PFASs in APM at Laoshan, China.

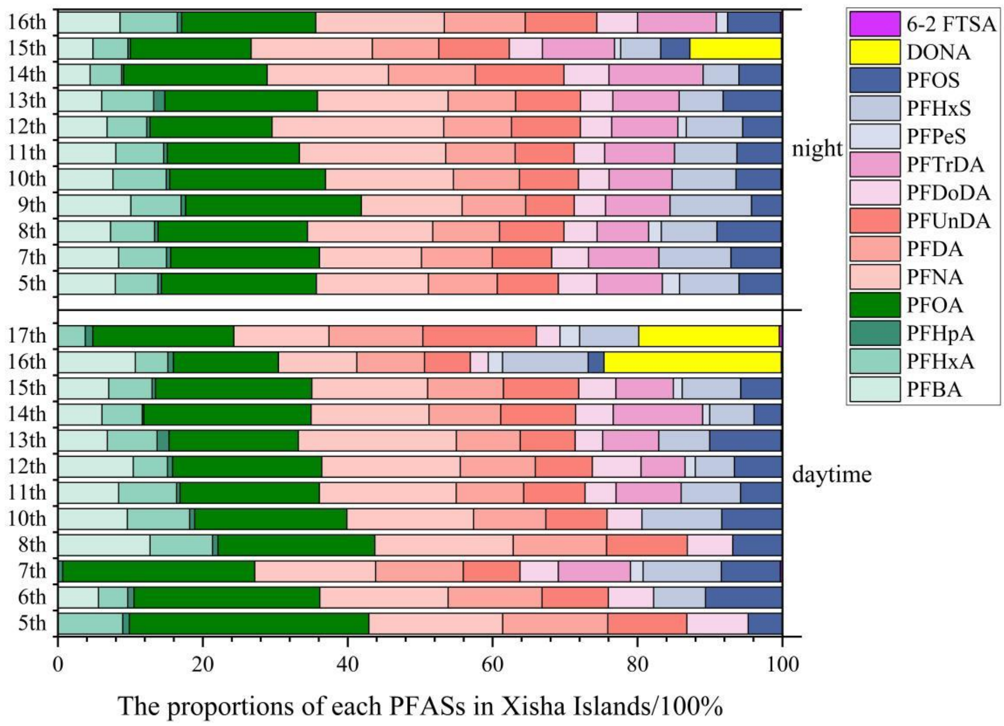

Fig S2. Profiles of PFASs in APM at Xisha Islands, China