Electronic Supplementary Material (ESI) for Environmental Science: Processes & Impacts.

Supporting Information of:

Legacy and Emerging Airborne Per- and Polyfluoroalkyl Substances (PFAS) Collected on $PM_{2.5}$ Filters in Close Proximity to a Fluoropolymer Manufacturing Facility

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Table S1. List of targeted per- and polyfluoroalkyl substances.

Compound	**Molecular weight**	**Formula**	**CAS#**	**Mass-labeled standard**
Perfluorocarboxylic acids (PFCAs)				
Perfluoro-n-butanoic acid (PFBA)	214.0	$C_4HF_7O_2$	375-22-4	$^{13}C_4$-PFBA
Perfluoro-n-pentanoic acid (PFPeA)	264.0	$C_5HF_9O_2$	2706-90-3	$^{13}C_5$-PFPeA
Perfluoro-n-hexanoic acid (PFHxA)	314.1	$C_6HF_{11}O_2$	307-24-4	$^{13}C_5$PFHxA
Perfluoro-n-heptanoic acid (PFHpA)	364.1	$C_7HF_{13}O_2$	375-85-9	$^{13}C_4$-PFHpA
Perfluoro-n-octanoic acid (PFOA)	414.1	$C_8HF_{15}O_2$	335-67-1	$^{13}C_8$-PFOA
Perfluoro-n-nonanoic acid (PFNA)	464.1	$C_9HF_{17}O_2$	375-95-1	$^{13}C_9$-PFNA
Perfluoro-n-decanoic acid (PFDA)	514.1	$C_{10}HF_{19}O_2$	335-76-2	$^{13}C_6$-PFDA
Perfluoro-n-undecanoic acid (PFUdA)	564.1	$C_{11}HF_{21}O_2$	2058-94-8	$^{13}C_7$-PFUdA
Perfluoro-n-dodecanoic acid (PFDoA)	614.1	$C_{12}HF_{23}O_2$	307-55-1	$^{13}C_2$-PFDoA
Perfluoro-n-tridecanoic acid (PFTrDA)	664.0	$C_{13}HF_{25}O_2$	72629-94-8	$^{13}C_2$-PFDoA
Perfluoro-n-tetradecanoic acid (PFTeDA)	714.0	$C_{14}HF_{27}O_2$	376-06-7	$^{13}C_2$-PFTeDA
Perfluoro-n-hexadecanoic acid (PFHxDA)	814.1	$C_{16}HF_{31}O_2$	67905-19-5	$^{13}C_2$-PFTeDA
Perfluoro-n-octadecanoic acid (PFODA)	914.1	$C_{18}HF_{35}O_2$	16517-11-6	$^{13}C_2$-PFTeDA
Perfluorosulfonic acids (PFSAs)				
Perfluorobutane sulfonic acid (PFBS)	300.1	$C_4HF_9O_3S$	375-73-5	$^{13}C_3$-PFBS
Perfluoropentane sulfonic acid (PFPeS)	350.1	$C_5HF_{11}O_3S$	2706-91-4	$^{13}C_3$-PFBS
Perfluorohexane sulfonic acid (PFHxS)	400.1	$C_6HF_{13}O_3S$	355-46-4	$^{13}C_3$-PFHxS
Perfluoroheptane sulfonic acid (PFHpS)	488.2	$C_7HF_{15}O_3S$	375-92-8	$^{13}C_3$-PFHxS
Perfluorooctane sulfonic acid (PFOS)	500.1	$C_8HF_{17}O_3S$	1763-23-1	$^{13}C_8$-PFOS
Perfluorononane sulfonic acid (PFNS)	567.2	$C_9HF_{19}O_3S$	68259-12-1	$^{13}C_8$-PFOS
Perfluorodecane sulfonic acid (PFDS)	617.2	$C_{10}HF_{21}O_3S$	335-77-3	$^{13}C_8$-PFOS
Perfluorododecane sulfonic acid (PFDoS)	700.1	$C_{12}HF_{25}O_3S$	79780-39-5	$^{13}C_8$-PFOS
Perfluoroalkyl ether carboxylic and sulfonic acids (PFECAs and PFESAs)				
Perfluoro-2-methoxyacetic acid (PFMOAA)	180.0	$C_3HF_5O_3$	674-13-5	$^{13}C_4$-PFBA
Perfluoro-2-methoxypropanoic acid (PMPA)	230.0	$C_4HF_7O_3$	13140-29-9	$^{13}C_4$-PFBA
Perfluoro (3,5-dioxahexanoic) acid (PFO2HxA)	246.0	$C_4HF_7O_4$	39492-88-1	$^{13}C_3$-HFPO-DA
1,1,2,2-tetrafluoro-2-(1,2,2,2-tetrafluoro-ethoxy)ethane sulfonate (NVHOS)	297.9	$C_4H_2F_8O_4S$	801209-99-4	$^{13}C_3$-PFBS
Perfluoro-2-ethoxypropanoic acid (PEPA)	280.0	$C_5HF_9O_3$	267239-61-2	^{13}C3-HFPO-DA
Perfluoro (3,5,7-trioxaoctanoic) acid (PFO3OA)	312.0	$C_5HF_9O_5$	39492-89-2	$^{13}C_3$-HFPO-DA
Hexafluoropropylene oxide-dimer acid (HFPO-DA), parent acid of “GenX” (GenX)	330.1	$C_6HF_{11}O_3$	13252-13-6	$^{13}C_3$-HFPO-DA
Perfluoro (3,5,7,9-tetraoxadecanoic) acid (PFO4DA)	378.0	$C_6HF_{11}O_6$	39492-90-5	$^{13}C_3$-HFPO-DA
Perfluoro 3,5,7,9,11-pentaoxadecanoic acid (PFO5DoA)	443.9	$C_7HF_{13}O_7$	39492-91-6	$^{13}C_9$-PFNA
2,2,3,3-tetrafluoro-3-((1,1,1,2,3,3-hexafluoro-3-(1,2,2,2-tetrafluoroethoxy)propan-2-yl)oxy)propanoic acid (HydroEve)	427.9	$C_8H_2F_{14}O_4$	773804-62-9	$^{13}C_3$-HFPO-DA
2-[1-[Difluoro[(1,2,2-trifluoroethenyl)oxy]methyl]-1,2,2,2-tetrafluoroethoxy]-1,1,2,2-tetrafluoroethanesulfonic acid (Nafion Byproduct 1)	443.9	$C_7HF_{13}O_5S$	29311-67-9	$^{13}C_3$-PFHxS
1,1,2,2-tetrafluoro-2-[1,1,1,2,3,3-hexafluoro-3-(1,2,2,2-tetrafluoroethoxy)propan-2-yl]oxyethanesulfonic acid (Nafion Byproduct 2)	463.9	$C_7H_2F_{14}O_5S$	749836-20-2	$^{13}C_3$-PFHxS
2,2,3,3,4,5,5,5-4-(1,1,2,2-tetrafluoro-2-sulfoethoxy) pentanoate (Nafion Byproduct 4)	440.9	$C_7H_2F_{12}O_6S$	2416366-18-0	$^{13}C_4$-PFBA

Table S2. Analytical detection limits for each targeted PFAS.

Analyte	Extract conc. (ng/mL)	Estimated air conc. (pg/m^3)
Perfluorocarboxylic acids (PFCAs)		
PFBA	0.13	0.0068
PFPeA	0.08	0.0042
PFHxA	0.05	0.0026
PFHpA	0.07	0.0037
PFOA	0.07	0.0037
PFNA	0.05	0.0026
PFDA	0.05	0.0026
PFUdA	0.08	0.0042
PFDoA	0.06	0.0032
PFTrDA	0.10	0.0053
PFTeDA	0.02	0.0011
PFHxDA	0.23	0.012
PFODA	0.20	0.011
Perfluorosulfonic acids (PFSAs)		
PFBS	0.25	0.013
PFPeS	0.20	0.011
PFHxS	0.26	0.014
PFHpS	0.10	0.0053
PFOS	0.05	0.0026
PFNS	0.12	0.0063
PFDS	0.09	0.0047
PFDoS	0.54	0.028
Perfluoroalkyl ether carboxylic and sulfonic acids (PFECAs and PFESAs)		
GenX	0.32	0.017
NVHOS	0.21	0.011
HydroEve	0.05	0.0026
PFMOAA	0.18	0.0095
PEPA	0.30	0.016
PMPA	0.04	0.0021
PFO2HxA	0.23	0.012
PFO3OA	0.17	0.0089
PFO4DA	0.13	0.0068
PFO5DoA	0.09	0.0047
Nafion Byproduct 1	0.05	0.0026
Nafion Byproduct 2	0.05	0.0026
Nafion Byproduct 4	0.06	0.0032

Note: The detection limits, expressed as air concentrations, were estimated using the analytical method detection limit, multiplying by a final extract volume of 100 µl, and dividing by a sample air volume of 1901 m^3 (sampling flow rate of 220 L min^{-1}, sampling duration of 6 days).

Table S3. Weekly-averaged PFAS concentrations in $PM_{2.5}$ (pg/m^3)[a]. Values in bold are detectable (i.e., greater than the field measurement detection limit). Values after "<" are the analytical detection limit based on injection of standards.

Legacy PFAS Weekly-Averaged $PM_{2.5}$ Concentrations (pg/m^3)														
Sample ID	**Sampling Duration**	**Air Volume (m^3)**	**PFBA**	**PFHxA**	**PFHpA**	**PFOA**	**PFNA**	**PFDoA**	**PFTeDA**	**PFHxDA**	**PFODA**	**PFHxS**	**PFOS**	**PFDoS**
SP1_1	091319-091919	1614	0.23	**1.08**	<0.0087	<0.0087	<0.0062	<0.0074	<0.0025	<0.029	<0.025	<0.032	**1.86**	<0.067
HV1_1	092019-092619	2180	0.20	**2.53**	<0.0064	0.035	<0.0046	<0.0055	<0.0018	<0.021	<0.018	<0.024	**0.63**	<0.050
SW1	092719-100319	1955	0.16	**6.10**	<0.0072	<0.0072	<0.0051	<0.0061	<0.0020	<0.024	<0.021	<0.027	<0.0051	<0.055
SW2	100419-101019	1922	**0.42**	**3.18**	<0.0073	<0.0073	<0.0052	0.040	<0.0021	**1.90**	<0.021	<0.027	**0.23**	<0.056
SW3	101119-101719	1922	**0.32**	**2.54**	<0.0073	<0.0073	<0.0052	<0.0062	<0.0021	**1.21**	<0.021	<0.027	**0.88**	<0.056
SW4	101819-102419	1901	**0.33**	**0.39**	<0.0074	<0.0074	<0.0053	<0.0063	<0.0021	<0.024	<0.021	<0.027	**0.85**	<0.057
SW5	102519-103119	1923	**0.30**	**0.55**	<0.0073	<0.0073	<0.0052	<0.0062	<0.0021	<0.024	<0.021	<0.027	**0.28**	<0.056
SW6	110119-110719	1861	**0.39**	**1.89**	<0.0075	<0.0075	<0.0054	<0.0064	<0.0021	<0.025	<0.022	<0.028	**1.16**	<0.058
SW7	110819-111419	1820	**0.45**	**0.52**	<0.0077	<0.0077	<0.0055	<0.0066	<0.0022	<0.025	<0.022	<0.029	**0.86**	<0.059
SW8	111519-112119	1850	**0.75**	**0.42**	<0.0076	<0.0076	<0.0054	<0.0065	<0.0022	<0.025	<0.022	<0.028	**0.36**	<0.058
SW9	112219-112819	1872	0.28	**1.73**	<0.0075	<0.0075	<0.0053	<0.0064	<0.0021	<0.025	<0.021	<0.028	**1.59**	<0.058
SW10	112919-120519	1864	**0.56**	**0.14**	<0.0075	<0.0075	<0.0054	<0.0064	<0.0021	<0.025	<0.022	<0.028	**2.71**	<0.058
SW11	120619-121219	1838	**0.65**	<0.0054	<0.0076	<0.0076	<0.0054	<0.0065	<0.0022	<0.025	<0.022	<0.028	**1.44**	<0.059
SW12	121319-121919	1856	**0.73**	<0.0054	<0.0075	<0.0075	<0.0054	<0.0065	<0.0022	<0.025	<0.022	<0.028	**1.29**	<0.058
SW13	122019-122619	1828	**0.62**	**0.29**	<0.0077	<0.0077	<0.0055	<0.0066	<0.0022	<0.025	<0.022	<0.028	**1.01**	<0.059
SW14	122719-010220	1818	**0.69**	**0.30**	<0.0077	<0.0077	<0.0055	0.031	<0.0022	**1.71**	<0.022	<0.029	**1.59**	<0.059
SW15	010320-010920	1725	**0.67**	<0.0058	<0.0081	<0.0081	<0.0058	<0.0070	<0.0023	<0.027	<0.023	<0.030	**3.19**	<0.063
SW16	011020-011620	1799	**0.92**	**1.41**	<0.0078	0.024	<0.0056	0.045	<0.0022	**1.07**	<0.022	**0.73**	**0.54**	**1.42**
SW17	011720-012320	1818	**0.70**	**0.24**	<0.0077	<0.0077	<0.0055	<0.0066	<0.0022	<0.025	<0.022	<0.029	**0.33**	<0.059
SW18	012420-013020	1810	**0.96**	**0.21**	<0.0077	0.022	<0.0055	<0.0066	<0.0022	<0.025	<0.022	<0.029	**0.68**	<0.060
SW21	021420-022020	1708	**0.75**	**3.21**	<0.0082	<0.0082	<0.0059	0.027	<0.0023	<0.027	<0.023	<0.031	**0.31**	**0.32**
SW22	022120-022720	1729	**0.62**	**1.12**	<0.0081	<0.0081	<0.0058	<0.0069	<0.0023	**1.41**	<0.023	<0.030	**1.10**	<0.063
SW23	022820-030520	1628	**0.68**	**3.52**	<0.0086	<0.0086	<0.0061	<0.0074	<0.0025	**1.02**	<0.025	<0.032	**1.51**	<0.066
SW24	030620-031220	1653	**0.54**	**1.15**	<0.0085	<0.0085	<0.0060	<0.0073	<0.0024	<0.028	<0.024	<0.032	**0.93**	<0.065
SW25	031320-031920	1672	**0.53**	**3.49**	<0.0084	<0.0084	<0.0060	**0.29**	<0.0024	<0.028	<0.024	<0.031	**1.69**	<0.065
SW26	032020-032620	1672	**0.49**	**1.72**	<0.0084	<0.0084	<0.0060	<0.0072	<0.0024	<0.028	<0.024	<0.031	**0.49**	<0.065
NE1	092719-100319	1902	0.08	**4.96**	<0.0074	0.042	<0.0053	<0.0063	<0.0021	<0.024	<0.021	<0.027	<0.0053	<0.057
NE2	100419-101019	1914	0.27	**0.73**	<0.0073	0.18	<0.0052	<0.0063	<0.0021	<0.024	<0.021	<0.027	**1.02**	<0.056
NE3	101119-101719	2089	0.19	**0.99**	<0.0067	<0.0067	<0.0048	<0.0057	<0.0019	<0.022	<0.019	<0.025	**2.05**	<0.052
NE4	101819-102419	2073	0.28	**0.86**	<0.0068	0.012	<0.0048	<0.0058	<0.0019	<0.022	<0.019	<0.025	**1.04**	<0.052
NE5	102519-103119	2114	0.25	**1.02**	<0.0066	<0.0066	**0.24**	<0.0057	<0.0019	<0.022	<0.019	<0.025	**0.70**	<0.051
NE6	110119-110719	1983	**0.37**	<0.0050	<0.0071	<0.0071	<0.0050	<0.0061	<0.0020	<0.023	<0.020	<0.026	**1.03**	<0.055

NE7	110819-111419	1949	**0.62**	<0.0051	<0.0072	0.059	<0.0051	<0.0062	<0.0021	<0.024	<0.021	<0.027	**1.71**	<0.055
NE8	111519-112119	2137	**0.59**	<0.0047	<0.0066	<0.0066	<0.0047	<0.0056	<0.0019	<0.022	<0.019	<0.024	**0.55**	<0.051
NE9	112219-112819	2050	**0.48**	<0.0049	<0.0068	<0.0068	<0.0049	<0.0059	<0.0020	<0.022	<0.020	**0.16**	**2.40**	<0.053
NE10	112919-120519	1863	**0.44**	<0.0054	**0.71**	<0.0075	<0.0054	0.0026	<0.0021	**5.33**	**2.75**	<0.028	**1.34**	<0.058
NE11	120619-121219	1839	**0.67**	<0.0054	<0.0076	0.011	<0.0054	<0.0065	<0.0022	<0.025	<0.022	<0.028	**0.86**	<0.059
NE12	121319-121919	1841	**0.56**	<0.0054	<0.0076	<0.0076	<0.0054	<0.0065	<0.0022	<0.025	<0.022	<0.028	**3.33**	<0.059
NE13	122019-122619	1828	**0.52**	<0.0055	<0.0077	<0.0077	<0.0055	0.047	0.0080	**0.92**	**1.32**	<0.028	**1.29**	<0.059
NE15	010320-010920	1788	**0.59**	<0.0056	<0.0078	<0.0078	<0.0056	<0.0067	<0.0022	<0.026	<0.022	<0.029	**2.05**	<0.060
NE18	021420-022020	1858	**0.59**	<0.0054	<0.0075	<0.0075	<0.0054	<0.0065	<0.0022	<0.025	<0.022	<0.028	**2.30**	<0.058
NE19	022120-022720	1720	**0.67**	<0.0058	<0.0081	<0.0081	<0.0058	<0.0070	<0.0023	<0.027	**2.55**	**0.22**	**1.75**	**3.65**
NE20	022820-030520	1652	**0.75**	<0.0061	<0.0085	0.0638	<0.0061	<0.0073	<0.0024	<0.028	<0.024	**0.12**	**3.36**	<0.065
NE21	030620-031220	1644	**0.61**	<0.0061	<0.0085	<0.0085	<0.0061	<0.0073	<0.0024	<0.028	<0.024	<0.032	**2.18**	<0.066
NE22	031320-031920	1662	**0.65**	<0.0060	<0.0084	<0.0084	<0.0060	<0.0072	<0.0024	<0.028	<0.024	<0.031	**4.70**	<0.065
NE23	032020-032620	1613	**0.60**	<0.0062	<0.0087	<0.0087	<0.0062	<0.0074	<0.0025	<0.029	<0.025	<0.032	**0.75**	<0.067
Field Measurement Detection Limit (pg/m³)[b]			0.29	0.026	0.025	0.63	0.042	0.058	0.034	0.073	0.59	0.014	0.0026	0.028
Average Field Blank (pg/m³)			0.18	0.0099	0.0034	0.15	0.016	0.0068	0.037	0.0098	0.056	0.00	0.00	0.00
Analytical Precision (CV, %)[c]			5.8	12.0	3.4	2.8	6.4	4.7	13.6	15.3	9.2	16.3	12.0	8.7

Emerging PFAS Weekly-Averaged $PM_{2.5}$ Concentrations (pg/m³)

Sample ID	Sampling Duration	Air Volume (m³)	PMPA	PFO2HxA	NVHOS	PEPA	PFO3OA	GenX	PFO4DA	PFO5DoA	Nafion BP1	Nafion BP2	Nafion BP4	Hydro Eve
SP1_1	091319-091919	1614	**1.19**	<0.029	**7.02**	<0.037	**0.042**	<0.040	<0.016	<0.011	**0.13**	**0.16**	**0.0040**	<0.0062
HV1_1	092019-092619	2180	**0.95**	<0.021	**0.76**	<0.028	<0.016	<0.029	<0.012	<0.008	**0.016**	**0.031**	<0.0055	<0.0046
SW1	092719-100319	1955	**0.96**	<0.024	**1.63**	<0.031	<0.017	<0.033	<0.013	<0.0092	**0.573**	**0.047**	**0.012**	<0.0051
SW2	100419-101019	1922	**1.36**	<0.024	**2.35**	<0.031	<0.018	<0.033	<0.014	**0.17**	**0.081**	**0.043**	**0.0085**	**0.070**
SW3	101119-101719	1922	**1.20**	<0.024	**0.85**	<0.031	<0.018	<0.033	<0.014	<0.0094	**0.10**	**0.063**	<0.0062	<0.0052
SW4	101819-102419	1901	**1.27**	<0.024	**2.59**	<0.032	**0.088**	<0.034	<0.014	<0.0095	**0.047**	**0.042**	0.0029	<0.0053
SW5	102519-103119	1923	**1.13**	<0.024	**2.13**	<0.031	<0.018	<0.033	<0.014	<0.0094	**0.064**	**0.068**	**0.0040**	<0.0052
SW6	110119-110719	1861	**1.42**	<0.025	**0.36**	<0.032	<0.018	<0.034	<0.014	<0.0097	**0.27**	**0.044**	0.0023	<0.0054
SW7	110819-111419	1820	**1.51**	<0.025	<0.023	**0.104**	<0.019	<0.035	**0.45**	**4.16**	**0.033**	**0.035**	**0.0065**	**0.097**
SW8	111519-112119	1850	**1.28**	<0.025	<0.023	0.004	<0.018	<0.035	<0.014	**0.49**	**0.061**	**0.029**	**0.0033**	**0.062**
SW9	112219-112819	1872	**1.12**	<0.025	<0.022	<0.032	<0.018	<0.034	<0.014	<0.0096	**0.021**	**0.031**	<0.0064	<0.0053
SW10	112919-120519	1864	**1.42**	<0.025	<0.023	0.026	<0.018	<0.034	<0.014	**0.20**	**0.054**	**0.027**	0.0008	**0.054**
SW11	120619-121219	1838	**1.60**	<0.025	<0.023	**0.138**	<0.019	<0.035	<0.014	**0.32**	**0.068**	**0.050**	**0.012**	**0.058**
SW12	121319-121919	1856	**1.69**	<0.025	<0.023	0.039	<0.018	<0.035	<0.014	**0.69**	**0.13**	**0.055**	**0.029**	**0.055**
SW13	122019-122619	1828	**1.43**	<0.025	<0.023	0.020	<0.019	**0.33**	<0.014	**0.08**	**0.038**	**0.043**	**0.0040**	**0.058**
SW14	122719-010220	1818	**1.85**	<0.025	<0.023	0.016	**0.030**	<0.035	<0.014	<0.0099	**0.059**	**0.045**	**0.0091**	**0.068**
SW15	010320-010920	1725	<0.0046	<0.027	<0.024	<0.035	**0.322**	**1.02**	**0.43**	**1.32**	**0.13**	**0.034**	**0.019**	**0.081**
SW16	011020-011620	1799	**1.91**	<0.026	<0.023	0.014	<0.019	0.02	<0.015	**0.21**	**0.017**	**0.040**	0.0024	**0.074**
SW17	011720-012320	1818	**1.98**	<0.025	<0.023	0.050	<0.019	**3.20**	**0.58**	**3.13**	**0.071**	**0.034**	**0.0046**	**0.069**

SW18	012420-013020	1810	**1.85**	<0.025	<0.023	<0.033	<0.019	<0.035	**0.0081**	**2.83**	**0.062**	**0.045**	**0.017**	**0.056**
SW21	021420-022020	1708	**2.02**	<0.027	<0.025	0.0058	<0.020	<0.038	**0.16**	**3.20**	**0.38**	**0.13**	**0.026**	**0.073**
SW22	022120-022720	1729	**1.76**	<0.027	<0.024	0.0032	<0.020	<0.037	<0.015	<0.010	**0.025**	**0.049**	0.0017	<0.0058
SW23	022820-030520	1628	**1.93**	<0.028	<0.026	<0.037	**0.032**	<0.039	<0.016	<0.011	**0.015**	**0.050**	0.0013	**0.066**
SW24	030620-031220	1653	**1.76**	<0.028	<0.025	<0.036	<0.021	<0.039	<0.016	**0.10**	**0.030**	**0.072**	<0.0073	**0.070**
SW25	031320-031920	1672	**1.74**	<0.028	<0.025	0.0032	<0.020	<0.038	<0.016	<0.011	**0.053**	**0.049**	**0.010**	**0.065**
SW26	032020-032620	1672	**1.60**	<0.028	<0.025	<0.036	<0.020	<0.038	<0.016	**0.25**	**0.20**	**0.039**	**0.0040**	**0.059**
NE1	092719-100319	1902	**0.93**	<0.024	**1.51**	<0.032	<0.018	<0.034	<0.014	<0.0095	**0.039**	**0.030**	**0.011**	<0.0053
NE2	100419-101019	1914	**1.25**	<0.024	**2.34**	<0.031	<0.018	<0.033	<0.014	**0.14**	**0.019**	**0.026**	<0.0063	<0.0052
NE3	101119-101719	2089	**1.09**	<0.022	**1.04**	<0.029	**0.037**	<0.031	<0.012	<0.0086	**0.11**	**0.053**	0.0008	**0.064**
NE4	101819-102419	2073	**1.15**	<0.022	**1.82**	<0.029	**0.049**	**0.23**	<0.013	<0.0087	**0.086**	**0.032**	0.0028	<0.0048
NE5	102519-103119	2114	**1.08**	<0.022	**2.07**	<0.028	<0.016	<0.030	<0.012	<0.0085	**0.10**	**0.035**	<0.0057	<0.0047
NE6	110119-110719	1983	<0.0040	**0.034**	**1.83**	<0.030	<0.017	<0.032	<0.013	<0.0091	<0.0050	<0.0050	<0.0061	<0.0050
NE7	110819-111419	1949	<0.0041	**0.137**	<0.022	<0.031	<0.017	<0.033	<0.013	**3.41**	**0.14**	**0.018**	**0.010**	0.024
NE8	111519-112119	2137	<0.0037	**0.21**	<0.020	<0.028	<0.016	<0.030	<0.012	**0.35**	**0.62**	**0.030**	**0.016**	0.015
NE9	112219-112819	2050	<0.0039	**0.08**	<0.021	<0.029	<0.017	<0.03	<0.013	**0.38**	**1.88**	**0.070**	**0.033**	<0.0049
NE10	112919-120519	1863	<0.0043	**0.099**	<0.023	<0.032	<0.018	**0.23**	<0.014	**0.43**	**0.22**	**0.017**	**0.034**	0.023
NE11	120619-121219	1839	<0.0044	**0.059**	<0.023	<0.033	<0.019	**0.37**	<0.014	**0.55**	**0.24**	**0.015**	**0.029**	0.025
NE12	121319-121919	1841	<0.0043	<0.025	<0.023	<0.033	<0.019	<0.035	<0.014	<0.0098	**0.0079**	0.008	<0.0065	<0.0054
NE13	122019-122619	1828	<0.0044	<0.025	<0.023	<0.033	<0.019	**0.26**	<0.014	**0.31**	**0.25**	**0.055**	**0.040**	0.020
NE15	010320-010920	1788	<0.0045	<0.026	<0.024	<0.034	<0.019	<0.036	<0.015	**0.34**	**0.063**	**0.023**	**0.0089**	0.022
NE18	021420-022020	1858	<0.0043	<0.025	<0.023	<0.032	<0.018	<0.034	<0.014	**0.11**	**0.10**	**0.050**	**0.032**	<0.0054
NE19	022120-022720	1720	<0.0047	<0.027	<0.024	<0.035	<0.020	**0.42**	<0.015	**0.29**	**0.36**	**0.24**	**0.093**	0.021
NE20	022820-030520	1652	<0.0048	<0.028	<0.025	<0.036	<0.021	**0.45**	<0.016	**1.20**	**0.20**	**0.029**	**0.012**	0.029
NE21	030620-031220	1644	<0.0049	<0.028	<0.026	<0.037	<0.021	**0.14**	<0.016	**0.50**	**0.10**	**0.033**	**0.16**	0.020
NE22	031320-031920	1662	<0.0048	<0.028	<0.025	<0.036	<0.021	**0.096**	<0.016	**0.27**	**0.02**	**0.026**	**0.0036**	<0.0060
NE23	032020-032620	1613	<0.0050	<0.029	<0.026	<0.037	<0.021	**0.091**	<0.016	**0.20**	**0.11**	<0.0062	**0.0072**	<0.0062
Field Measurement Detection Limit (pg/m³)[b]			0.0021	0.012	0.046	0.083	0.0089	0.059	0.0068	0.0047	0.0026	0.013	0.0032	0.034
Average Field Blank (pg/m³)			0.00	0.00	0.019	0.0079	0.00	0.0080	0.00	0.00	0.00	0.003	0.00	0.0032
Analytical Precision (CV, %)[c]			7.5	0.2	6.2	16.3	12.9	15.4	7.7	30.9	3.6	3.6	3.8	12.2

[a]: Reported concentrations were blank corrected.
[b]: 3σ of the field blank divided by a sample air volume of 1901 m^3 (i.e., sampling flow rate of 220 L min^{-1}, sampling duration of 6 days).
[c]: Pooled coefficient of variation: pooled standard deviation divided by the mean value of duplicate sample analyses. For paired data, $\sigma_{pooled} = [\Sigma di^2/2n]^{1/2}$, where: σ_{pooled} is the standard deviation, d is the difference between paired i values, and n is the number of pairs.

Table S4. Descriptive statistics of PFAS $PM_{2.5}$ measurements at the SW and NE sites (DF: Detection Frequency).

		SW (N=26)			NE (N=20)		
Compound Name		DF (%)	Min (pg/m^3)	Max (pg/m^3)	DF (%)	Min (pg/m^3)	Max (pg/m^3)
Legacy PFAS	PFBA	84.6	0.16	0.96	75.0	0.084	0.75
	PFHxA	88.5	0.0054	6.1	25.0	0.0047	5.0
	PFHpA	0.0	0.0064	0.0087	5.0	0.0066	0.71
	PFNA	0.0	0.0046	0.0062	5.0	0.0047	0.24
	PFDoA	3.8	0.0055	0.29	0.0	0.0026	0.047
	PFHxDA	23.1	0.021	1.9	10.0	0.022	5.3
	PFODA	0.0	0.018	0.025	15.0	0.019	2.8
	PFHxS	3.8	0.024	0.73	15.0	0.024	0.22
	PFOS	96.2	0.0051	3.2	95.0	0.0053	4.7
	PFDoS	7.7	0.050	1.4	5.0	0.051	3.6
Emerging PFAS	PMPA	96.2	0.0046	2.0	25.0	0.0037	1.2
	PFO2HxA	0.0	0.021	0.029	30.0	0.022	0.21
	NVHOS	30.8	0.022	7.0	30.0	0.020	2.3
	PEPA	7.7	0.0032	0.14	0.0	0.028	0.037
	PFO3OA	19.2	0.016	0.32	10.0	0.016	0.049
	GenX	11.5	0.017	3.2	45.0	0.030	0.45
	PFO4DA	19.2	0.0081	0.58	0.0	0.012	0.016
	PFO5DoA	53.8	0.0083	4.2	70.0	0.0085	3.4
	Nafion BP1	100.0	0.015	0.57	95.0	0.0050	1.9
	Nafion BP2	100.0	0.027	0.16	85.0	0.0050	0.24
	Nafion BP4	61.5	0.0008	0.029	70.0	0.0008	0.16
	Hydro Eve	65.4	0.0046	0.097	5.0	0.0047	0.064

Table S5. Spearman correlation analysis for paired SW and NE site measurements.

Compound Name		Spearman Correlation, two-tailed, α=0.01	
		SW vs. NE	
		Correlation Coefficient	P-value
Legacy PFAS	PFBA	**0.618**	**0.004***
	PFHxA	0.289	0.217
	PFHpA	N/A	N/A
	PFNA	N/A	N/A
	PFDoA	N/A	N/A
	PFHxDA	-0.165	0.487
	PFODA	N/A	N/A
	PFHxS	N/A	N/A
	PFOS	**0.600**	**0.005***
	PFDoS	-0.053	0.826
Emerging PFAS	PMPA	**-0.516**	**0.020***
	PFO2HxA	N/A	N/A
	NVHOS	**0.975**	**0.000***
	PEPA	N/A	N/A
	PFO3OA	0.348	0.132
	GenX	0.046	0.848
	PFO4DA	N/A	N/A
	PFO5DoA	0.344	0.138
	Nafion BP1	**-0.595**	**0.006***
	Nafion BP2	0.229	0.332
	Nafion BP4	-0.313	0.178
	Hydro Eve	0.215	0.362
Total Legacy PFAS		**0.487**	**0.029***
Total Emerging PFAS		0.277	0.238
Total PFAS (legacy+emerging)		**0.483**	**0.031***

*: statistically significant at at α= 0.01 for Spearman Correlation Test.

Table S6. Spearman correlation coefficients among PFAS species with detection frequency >20%.

	PFBA	PFHxA	PFOS	PMPA	NVHOS	GenX	Hydro Eve	Nafion BP4	Nafion BP1	PFO5DoA	Nafion BP2
PFBA	1										
PFHxA	**-.406****	1									
PFOS	0.156	**-.516****	1								
PMPA	0.235	**.597****	**-.388****	1							
NVHOS	**-.748****	**.437****	**-.339***	-0.031	1						
GenX	**.320***	**-.465****	0.212	**-.357***	-0.284	1					
Hydro Eve	**.558****	0.067	-0.083	**.504****	**-.448****	0.101	1				
Nafion BP4	**.357***	**-.479****	0.119	**-.337***	**-.418****	**.391****	0.198	1			
Nafion BP1	0.045	-0.24	0.006	**-.313***	-0.081	0.285	0.004	**.652****	1		
PFO5DoA	**.631****	**-.537****	0.059	-0.087	**-.594****	**.379****	**.502****	**.623****	**.392****	1	
Nafion BP2	-0.006	**.353***	-0.051	**.442****	0.085	-0.21	0.176	0.145	0.247	-0.157	1

** significant at α=0.01(two-tailed), * significant at α= 0.05 (two-tailed).

Table S7. Paired upwind and downwind sampling dates.

Sampling Started	10/4/2019	11/1/2019	11/8/2019	11/15/2019	12/6/2019	12/20/2019	2/14/2020	2/21/2020	2/28/2020	3/6/2020
Wind Direction	NE	NE	NE	NE	NE	NE	NE	SW	SW	SW
Downwind Site	SW	SW	SW	SW	SW	SW	SW	NE	NE	NE

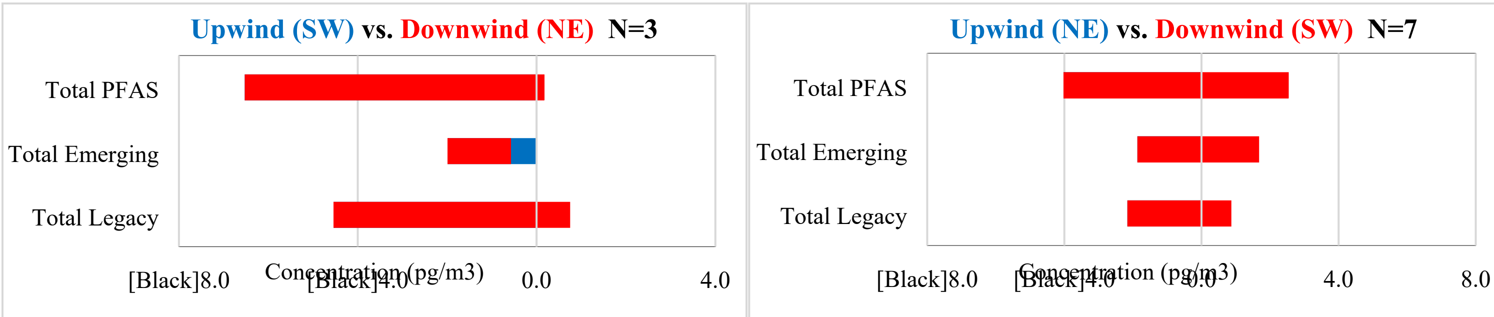
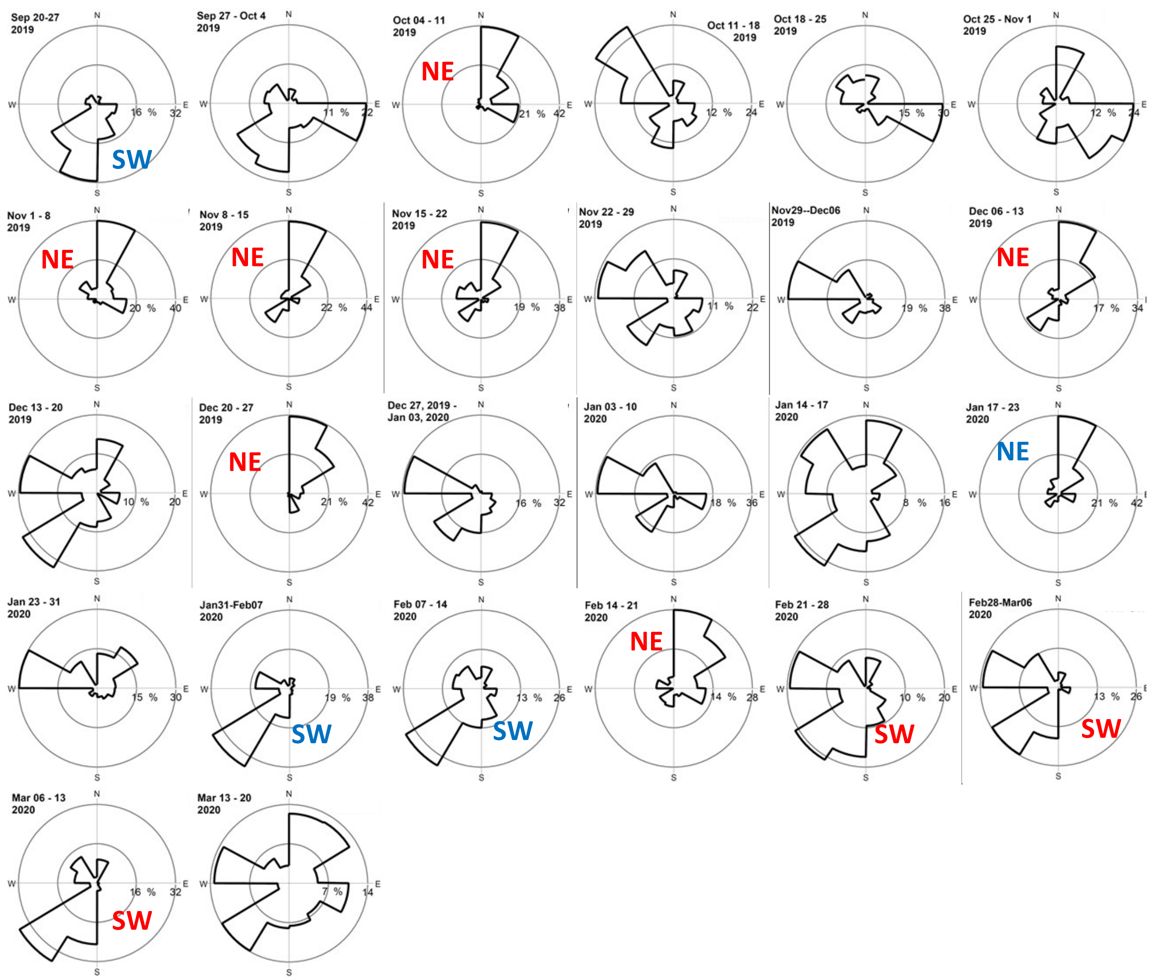


Figure S1. Upwind and downwind PFAS $PM_{2.5}$ concentrations.



Red: Paired NE and SW PFAS Measurement

Blue: Only SW PFAS Measurement

Figure S2. Weekly wind roses for the SW and NE sites.

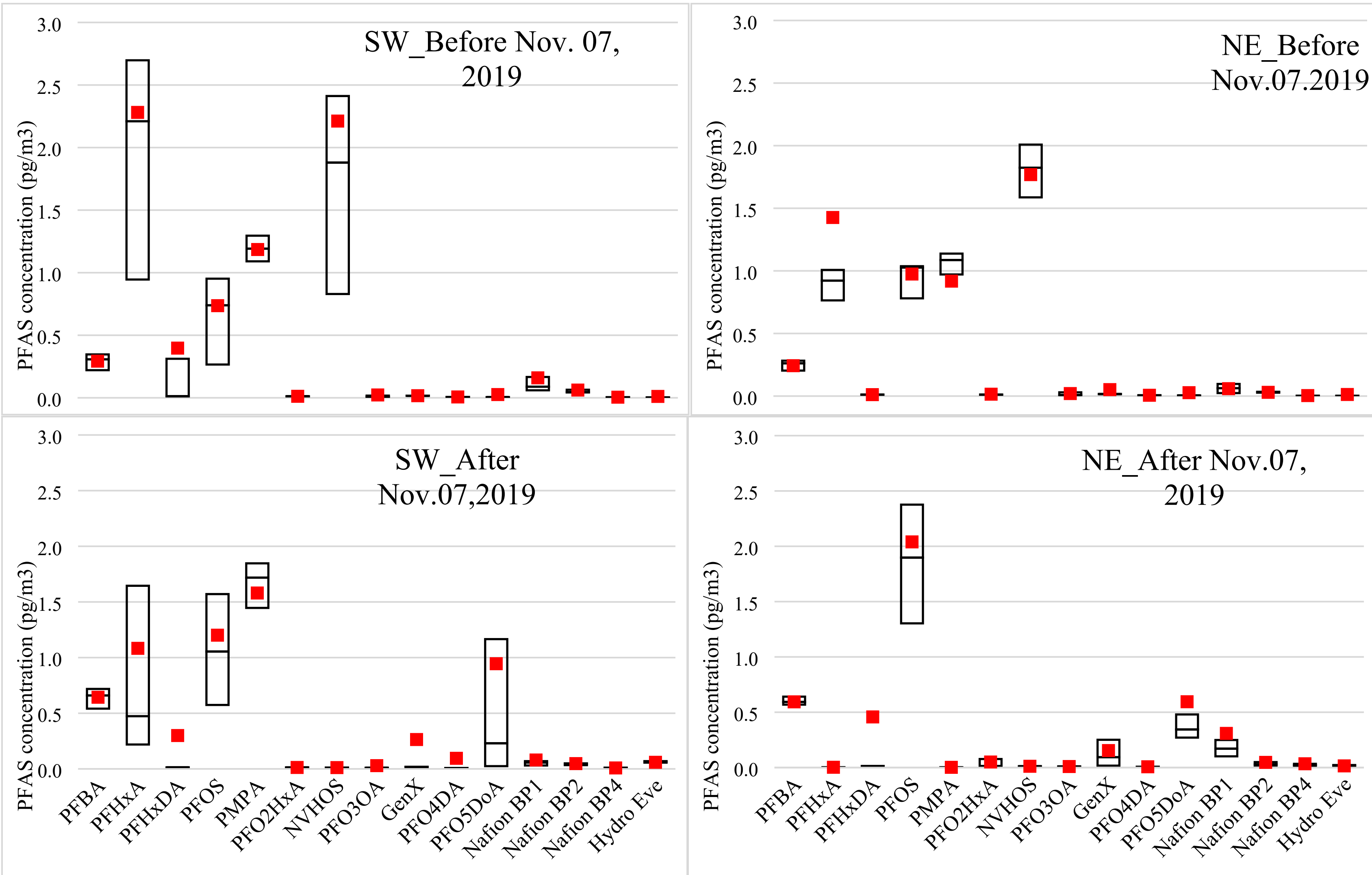


Figure S3. Box-plot of weekly-averaged PFAS species concentrations in $PM_{2.5}$ (pg/m^3) for all PFAS species with detection frequencies >10%, measured at SW and NE sites, before and after November 07, 2019. The top, central, and bottom of each box represent the third quartile, median, and the first quartile, respectively. Red squares represent the mean value.

Potential Sources of Individual PFAS. For four decades, the Chemours' Fayetteville Works has released thousands of pounds of GenX and other PFAS annually to the environment.[1] According to emissions data submitted by the Chemours' Fayetteville Works facility to NC DEQ, [2, 3] PFAS emissions to air averaged 45,390 kg/year during 2012-2016, and the total PFAS emissions at the Fayetteville Works facility were 109,393 kg/year in 2017. It is noteworthy that several control actions were taken by the Chemours' Fayetteville Works facility to reduce PFAS emissions as required by NC DEQ since 2018.[4] For example, carbon adsorber units were commissioned and scrubbers were upgraded in order to meet the requirements of the consent order. A thermal oxidizer/4-stage scrubber system designed for 99.99% PFAS removal was installed and was fully operational December 27, 2019.[5] Two removal efficiency tests (one in February, 2020, and the other one in January, 2021) were performed by Chemours since the installation of the thermal oxidizer.[6, 7] These tests concluded that the thermal oxidizer reached the >99.99% removal requirement based on measurement of five target PFAS compounds: HFPO monomer, GenX, HFPO-DAF (hexafluoropropylene dimer acid fluoride), COF2 (carbonyl difluoride), and Fluoroether E-1 (heptafluoropropyl-1,2,2,2-tetrafluoroethyl ether). Below we provide some information of potential sources about selected PFAS compounds we detected.

PFBA and PFHxA: PFBA and PFHxA are sometimes used as substitutes for longer-chain PFAS (e.g., PFOA).[8, 9, 10] PFBA was frequently detected at both sites (85% at SW site and 75% at NE site). Similarly, PFHxA was frequently detected at the SW site (88%). PFBA and PFHxA are both positively correlated with several longer-chain PFAS in this study (Table S6), suggesting they are used (precursor) or generated (product or byproduct) in the manufacturing process.

PFOA: the ammonium salts of PFOA are mainly used in the production of fluoropolymers and can readily dissociate into PFOA in the environment. Therefore, direct industrial emissions are a source of PFOA.[11, 12] PFOA has been measured and identified as the predominant PFAS species near fluorochemical facilities in the US before 2015,[13] or in other countries (e.g. Italy, Spain, Japan, and China)[14-19] where there are still ongoing PFOA production to meet market demands without regulations on PFOA emissions.[20] Continued occurrence of PFOA contamination in surface water has been seen tens of kilometers away from manufacturing plants at least 10 years after dramatic reductions in air emissions,[21, 22] suggesting long-term impacts of legacy contamination, manufacturing byproducts or secondary formation in the local environment. Eight of the main fluoropolymer manufacturers in the U.S. participated in the U.S. Environmental Protection Agency (EPA) 2010/2015 Stewardship Program in 2006[8] to completely cease the production and use of PFOA and related compounds by 2015.[23] At the Chemours' Fayetteville Works facility, PFOA was produced during 2002 to 2013, and was gradually phased out while using GenX as a replacement.[24] Reported emission data from the Chemours Company-Fayetteville Works facility and measurement of process/non-process wastewater measured inside the facility are consistent with the phase out of PFOA production at Chemours.[25, 26] PFOA was below detection limits in $PM_{2.5}$ samples in our study.

PFOS: Unlike PFOA, which has been discharged primarily from point sources at manufacturing facilities, PFOS has more widely dispersed emission sources.[11] Though PFOS and PFOA were phased out of production in the US in the 2000s, PFOS was still detected at higher concentrations in the nearfield samples in our study than in regional background samples, suggesting either that PFOS is emitted as a chemical impurity during manufacturing, emitted from previously contaminated land through volatilization or re-entrainment of contaminated soil, or formed from other PFAS through atmospheric/environmental chemistry. A PFAS emission inventory provided by the Fayetteville Works facility for 2012-2016 reported no detectable PFOS emitted and the emissions of some potential PFOS precursors (e.g., sulfonamide substances) were low. Higher $PM_{2.5}$ concentrations of PFOS than PFOA measured during our study are consistent with recent studies around this same area that analyzed drinking and surface water levels of PFOS and PFOA. These recent studies found that PFOS water concentrations were higher than PFOA.[27, 28] PFOS was the only C8 legacy PFAS measured in ambient $PM_{2.5}$ samples collected in this area.

GenX: the ammonium salt of HFPO-DA, "GenX", has been used as a substitute for PFOA since 2013, but emissions have been recently reduced through regulatory controls.[29] Its release as an airborne contaminant has been previously verified.[29] GenX was first detected in the Cape Fear River in 2012,[30] and Chemours Company-Fayetteville Works facility acknowledged that GenX produced as a byproduct was released into the Cape Fear River through the Fayetteville Works wastewater treatment plant starting in 1980. High levels of GenX have been measured in local drinking water wells and in groundwater samples collected from both upstream and downstream of the Chemours' Fayetteville Works.[28, 31, 32] This suggests that GenX was emitted into the air and delivered to these waters by atmospheric wet or dry deposition.[2, 31, 33] Similarly, Brandsma et al.[34] detected GenX in plant material collected 3 km downwind from a fluoropolymer manufacturing facility in the Netherlands, indicating that GenX undergoes near-field atmospheric deposition. Due to the control technologies requested by NC DEQ in 2018 and 2019,[4] reported GenX emissions from the Chemours' Fayetteville Works were reduced to 1.31 kg/year in 2019[35] and 0.75 kg/year in 2020,[36] while the value was 1044 kg/year in 2017.[37] The detection frequency for GenX in $PM_{2.5}$ samples analyzed during our study was relatively low, although the detection limits were not as low as some species. In our study, GenX was measured in 12 out of total 46 SW+NE samples near the Chemours' Fayetteville Works facility, with $PM_{2.5}$

concentrations mostly <0.5 pg/m^3, with the exception of two spikes in January 2020 with concentrations >1 pg/m^3. However, it should be stated here we did not measure the gas-phase GenX levels during this study.

NVHOS: NVHOS was measured in moderately high concentrations in September and October 2019 at both near-field sampling sites (0.02 to 7.0 pg/m^3) and below detection limits after those two months, suggesting either the success of control efforts to remove it or a change in manufacturing activities in early November.

Nafion BP1, Nafion BP2 and Nafion BP4: These compounds were detectable in almost all the $PM_{2.5}$ samples collected at both near-field sites. Since one main group of the manufacturing products at the Chemours' Fayetteville Works are Nafion membranes and films, it is not surprising that we observed these Nafion byproducts frequently in $PM_{2.5}$. Notably, these same three Nafion byproducts have been recently measured in pine needles collected within 2 miles of this facility,[38] indicating the existence and contamination of these three byproducts in the surrounding area of the facility.

PMPA: PMPA was detected in 96% of all the $PM_{2.5}$ samples collected at the SW near-field sampling site. PMPA and Nafion byproducts (1, 2, and 4) have also been detected in high concentrations in process and non-process wastewater collected from the facility, suggesting PMPA originates from the Chemours Fayetteville Works facility.[25, 39]

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