

2016

# Detection of Poly- and Perfluoroalkyl Substances (PFASs) in U.S. Drinking Water Linked to Industrial Sites, Military Fire Training Areas, and Wastewater Treatment Plants

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## Citation/Publisher Attribution

Hu, X. C., Andrews, D. Q., Lindstrom, A. B., Bruton, T. A., Schaider, L. A., Grandjean, P., Lohmann, R., Carignan, C. C., Blum, A., Balan, S. A., Higgins, C. P., & Sunderland, E. M. (2016). Detection of Poly- and Perfluoroalkyl Substances (PFASs) in U.S. Drinking Water Linked to Industrial Sites, Military Fire Training Areas, and Wastewater Treatment Plants. *Environ. Sci. Technol. Lett.*, 3(10), 344-350.

Available at: <http://dx.doi.org/10.1021/acs.estlett.6b00260>

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2 **industrial sites, military fire training areas and wastewater treatment plants**

3

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26 The authors declare no competing financial interest.

27 **Abstract**

28 Drinking water contamination with poly- and perfluoroalkyl substances (PFASs) poses risks to  
29 the developmental, immune, metabolic, and endocrine health of consumers. We present a spatial  
30 analysis of 2013-2015 national drinking water PFAS concentrations from the U.S.  
31 Environmental Protection Agency's (US EPA) third Unregulated Contaminant Monitoring Rule  
32 (UCMR3) program. The number of industrial sites that manufacture or use these compounds,  
33 military fire training areas, and wastewater treatment plants are all significant predictors of PFAS  
34 detection frequencies and concentrations in public water supplies. Among samples with  
35 detectable PFAS levels, each additional military site within a watershed's 8-digit hydrologic unit  
36 is associated with a 20% increase in PFHxS, a 10% increase in both PFHpA and PFOA, and a  
37 35% increase in PFOS. The number of civilian airports with personnel trained in the use of  
38 aqueous film-forming foams (AFFFs) is significantly associated with the detection of PFASs  
39 above the minimum reporting level. We find drinking water supplies for 6 million U.S. residents  
40 exceed US EPA's lifetime health advisory (70 ng/L) for PFOS and PFOA. Lower analytical  
41 reporting limits and additional sampling of smaller utilities serving <10,000 individuals and  
42 private wells would greatly assist in further identifying PFAS contamination sources.

43

44

## 45 **Introduction**

46 Poly- and perfluoroalkyl substances (PFASs) are a large group of persistent  
47 anthropogenic chemicals used in industrial processes and commercial products over the past 60  
48 years.<sup>1</sup> Widespread use and extreme resistance to degradation have resulted in the ubiquitous  
49 presence of these compounds in the environment. The 2011-2012 U.S. National Health and  
50 Nutrition Examination Survey reported detectable serum PFAS concentrations in virtually all  
51 individuals (97%).<sup>2,3</sup> Human PFAS exposure has been linked to cancer, elevated cholesterol,  
52 obesity, immune suppression, and endocrine disruption.<sup>4-6</sup> Health concerns in the early 2000s  
53 prompted manufacturers in Europe and North America to phase out production of some long-  
54 chain PFASs.<sup>7-10</sup> Declines in production of these compounds have been offset by increases in  
55 developing regions such as Asia.<sup>8</sup> Limited available data suggest widespread exposure to  
56 replacement (short-chain) PFASs may also adversely affect human health.<sup>11,12</sup>

57 Human PFAS exposure includes dietary sources, household dust, air, and drinking  
58 water.<sup>13,14</sup> Exposure from drinking water is a serious concern due to the high aqueous solubility  
59 of many PFASs.<sup>15,16</sup> Relatively low PFAS concentrations can lead to elevated exposures in the  
60 general population.<sup>17</sup> Elevated PFAS concentrations in U.S. drinking water have been reported in  
61 numerous regions,<sup>15,16,18,19</sup> especially near industrial sites that produce or use them.<sup>6,16,20</sup> For  
62 example, perfluorooctanoic acid (PFOA) concentrations 190-fold higher than the lifetime health  
63 advisory (70 ng/L) recommended by the U.S. Environmental Protection Agency (US EPA)<sup>21</sup>  
64 were measured in drinking water near a fluorochemical facility in Washington, West Virginia  
65 where PFOA was used in fluoropolymer production.<sup>18</sup>

66 Many civilian airports and military fire training areas have been contaminated by PFASs  
67 contained in aqueous film-forming foams (AFFFs) that are widely used during firefighting

68 training activities. Groundwater and surface waters surrounding these sites containing PFAS  
69 concentrations that are three to four orders of magnitude higher than the US EPA health advisory  
70 level for drinking water have been reported.<sup>22, 23</sup> Wastewater treatment plants (WWTPs) are  
71 another important PFAS source because these compounds are not removed by standard treatment  
72 methods<sup>24</sup> and labile precursors biodegrade, increasing concentrations in effluent relative to  
73 influent.<sup>25, 26</sup> Land application of approximately half of the biosolids generated by WWTPs may  
74 contribute to human exposure through subsequent contamination of water, food, livestock, and  
75 wildlife.<sup>27</sup>

76         Understanding nation-wide PFAS exposures from drinking water is important for  
77 identifying potentially vulnerable populations. However, previous studies have mainly focused  
78 on individual point sources of PFAS contamination and site-specific drinking water exposures.<sup>15,</sup>  
79 <sup>16</sup> Here we develop a statistical framework for investigating whether increased PFAS  
80 concentrations in drinking water are associated with the number of point sources within a  
81 watershed (represented by an 8-digit hydrologic unit code, from here on abbreviated HUC). We  
82 used publicly available drinking water concentration data for six PFASs from the US EPA’s third  
83 Unregulated Contaminant Monitoring Rule (UCMR3), including: perfluorobutane sulfonic acid  
84 (PFBS), perfluorohexane sulfonic acid (PFHxS), perfluoroheptanoic acid (PFHpA), PFOA,  
85 perfluorooctane sulfonic acid (PFOS), and perfluorononanoic acid (PFNA) (Table S1).<sup>28</sup> We  
86 discuss the utility of the UCMR3 database for identifying PFASs sources to U.S. drinking water  
87 supplies, locations of vulnerable populations, and priorities for future monitoring.

88

## 89 **Methods**

### 90 **Drinking water data**

91 Our analysis included analytical results for six PFASs in 36,149 drinking water samples  
92 from the US EPA’s UCMR3 program collected between January 2, 2013 and December 9,  
93 2015.<sup>28</sup> Samples cover all 4,064 public water supplies serving > 10,000 individuals. Data are  
94 also available for 800 public water supplies serving <10,000 individuals but this represents only  
95 a small fraction (0.5%) of the 144,165 in this category. Minimum reporting levels (MRLs) for  
96 the six PFASs analyzed are listed in Table S1.

97 One limitation of the UCMR3 database is that national data on system intakes for public  
98 water supplies are classified,<sup>29</sup> making it difficult to place them within a specific hydrological  
99 network. We therefore extracted the zip codes for areas served and aggregated data within 8-digit  
100 HUCs<sup>30</sup> to capture the most detailed hydrologic information that exceeds the spatial resolution of  
101 PFAS data (zip code areas). We used the highest reported PFAS concentrations when multiple  
102 systems were located within a single zip code and/or when multiple zip code areas were located  
103 within the same HUC.

104

### 105 **PFAS point sources**

106 Our spatial analysis (Figure S1) included point source information for: (a) 16 industrial  
107 sites listed in the US EPA’s 2010/2015 PFOA Stewardship Program (Table S2);<sup>31</sup> (b) 8572  
108 WWTPs;<sup>32</sup> (c) 290 military fire training areas that contain 664 military fire training sites;<sup>33</sup> and  
109 (d) 533 civilian airports that are compliant with Title 14 Code of Federal Regulations, Part 139  
110 for personnel trained in the use of AFFF (from here on referred to as “AFFF certified  
111 airports”).<sup>34</sup> PFASs produced and/or used vary across industrial sites and not all compounds were  
112 associated with all sites. For example, a fluorochemical manufacturing facility in Decatur,  
113 Alabama, produced both PFOS and PFOA,<sup>35</sup> while only PFOA was used in the manufacturing



114 process of another fluorochemical production facility in Parkersburg, West Virginia.<sup>36</sup> We  
115 conducted a sensitivity analysis to examine the potential production misclassification bias by  
116 limiting industrial sites to include the ones that only produced or used each specific compound  
117 (Table S3). We used the Google Maps application program interface (API) to geocode  
118 coordinates based on addresses. Potentially important PFAS sources such as landfills, biosolids,  
119 and small industrial PFAS users could not be included in this analysis because comprehensive  
120 geospatial data are not available.

121

## 122 **Spatial and statistical analysis**

123 We used ArcMap 10.3.1 (ESRI) to explore statistical differences between the number of  
124 point sources in 8-digit HUCs with PFAS levels above and below detection. We developed a  
125 multivariate spatial regression model for watersheds with detectable PFASs that adjusts for  
126 correlations and co-location among point sources. A natural log transformation was used to  
127 normalize the distribution of individual PFASs. PFNA and PFBS were excluded from the spatial  
128 regression analysis due to low detection frequency (15 and 14 out of 1601 watersheds,  
129 respectively). We used Moran's I statistic to test for spatial dependence in the model residuals  
130 from an ordinary least square (OLS) regression and correct for spatial dependence in the final  
131 spatial regression model. Akaike Information Criterion<sup>37</sup> was used to compare the OLS and  
132 spatial regression models, where a lower value implies a better model fit. A series of cross-  
133 validation tests were also completed to assess the predictive capacity and stability of the final set  
134 of models. The OLS and spatial regression models were constructed using GeoDa 1.6 software,<sup>38</sup>  
135 and cross-validation was implemented in R version 3.1.3.

136

137 **Results and Discussion**

138 **PFASs in U.S. drinking water**

139 PFASs were detected at or above the MRLs in 194 out of 4,864 public water supplies,  
140 serving 16.5 million residents in 33 different states, three American territories (American Samoa,  
141 Northern Mariana Islands and Guam), and the Salt River Pima-Maricopa Indian Community.  
142 Drinking water from 13 states accounted for 75% of detections, including, by order of frequency  
143 of detection: California, New Jersey, North Carolina, Alabama, Florida, Pennsylvania, Ohio,  
144 New York, Georgia, Minnesota, Arizona, Massachusetts and Illinois (Figure 1). Detection  
145 frequencies for PFASs across the 4,864 public water supplies were 2.2% for PFOA, 2.0% for  
146 PFOS, 1.7% for PFHpA, 1.1% for PFHxS, and <0.003% for others.

147 Many detectable PFAS concentrations in the UCMR3 database are above chronic  
148 drinking water and water quality standards for other regions (i.e., surface water European Union:  
149 PFOS <1 ng/L; drinking water Sweden: sum of 7 PFASs < 90 ng/L; ground water State of New  
150 Jersey: PFNA <10 ng/L; drinking water State of Vermont: sum of PFOS and PFOA <20 ng/L).<sup>39-</sup>  
151 <sup>42</sup> A recent analysis developed a benchmark-dose for immunotoxicity in children and suggested a  
152 drinking water limit of approximately 1 ng/L for PFOS and PFOA.<sup>26</sup> Data from rodents that  
153 measured sensitive endpoints such as mammary gland development support a similar level.<sup>26</sup>

154 Six million people were served by 66 public water supplies that have at least one sample  
155 at or above the US EPA's 2016 health advisory for PFOS and PFOA (70 ng/L individually or  
156 combined). Concentrations ranged as high as 349 ng/L for PFOA, 1,800 ng/L for PFOS, and 56  
157 ng/L for PFNA.

158 The detection frequency in drinking water sourced from groundwater was more than  
159 twice that from surface water (Table S4). Long-chain PFASs<sup>43</sup> (PFHxS, PFOS, PFOA, PFNA)

160 were more frequently detected in groundwater and short-chain compounds (PFHpA, PFBS) were  
161 detected more frequently in surface waters. This may be due to both the original mode of  
162 environmental release (as an aerosol, application to soil, aqueous discharge) and the inverse  
163 relationship between PFAS mobility and chain length.<sup>44</sup> The MRLs (10-90 ng/L) in the UCMR3  
164 database are up to two orders of magnitude higher than the limit of quantitation in most  
165 published studies,<sup>45-49</sup> and more than 10 times higher than the drinking water limit (1 ng/L)  
166 suggested by human and animal studies.<sup>26, 50</sup> Since PFASs are detectable in virtually all parts of  
167 the environment,<sup>5, 7, 9, 13, 14, 20, 44, 51</sup> we infer that the large fraction of samples below reporting  
168 limits (Table S4) is driven in part by high MRLs.

169

#### 170 **Sources surrounding locations with detectable PFASs**

171 Our analysis indicates point sources are significantly more abundant in HUCs with  
172 detectable PFASs (two-sided t-test,  $p < 0.05$ , Table 1, Figure S2). This includes drinking water  
173 samples from 1601 of the 2158 total U.S. HUCs. For example, HUCs with detectable PFOA  
174 levels (8% of the total) have more industrial sites, military fire training areas, AFFF certified  
175 airports, and WWTPs than those with concentrations below detection. These trends are  
176 observable across all PFASs. Similarly, HUCs with point sources have higher detection  
177 frequencies for PFASs (Table S5). For example, 10.4% of the HUCs with no military fire  
178 training areas have a detection of any PFAS, but this percentage increases to 28.2% for HUCs  
179 with at least one. One caveat is that imprecise information on public water supply intakes can  
180 cause misclassification bias. Systems that draw water upstream from point sources, such as  
181 Minneapolis and St. Paul in Minnesota, may not actually be affected as indicated the by  
182 aggregated spatial analysis.

183

## 184 **Results of the spatial regression model**

185         Spatial regression modeling explains 38-62% of the variance in drinking water  
186 concentrations for the four PFASs considered (Table 2). Each additional industrial site within a  
187 HUC is associated with an 81% increase in PFOA ( $p<0.001$ ), which is the strongest statistical  
188 association across compounds and point sources. Increasing PFOS concentrations are positively  
189 associated with the number of industrial sites but this relationship is not statistically significant  
190 ( $p=0.124$ ). The small number of sites that have manufactured or used PFOS likely accounts for  
191 the lack of a statistically significant relationship.

192         The number of military fire training areas within each HUC is positively associated with  
193 increasing levels of all PFOS, PFOA, PFHxS and PFHpA, and is statistically significant for  
194 PFHxS ( $p=0.045$ ) and PFOS ( $p=0.007$ ). Each additional military fire training area within the  
195 same HUC is associated with a 20% increase in PFHxS ( $p=0.002$ ), 10% increase in PFHpA  
196 ( $p=0.155$ ), 10% increase in PFOA ( $p=0.111$ ) and 35% increase in PFOS ( $p<0.001$ ). AFFFs  
197 typically contain relatively high concentrations of PFOS and PFHxS and their polyfluorinated  
198 precursors compared to other perfluorinated carboxylates,<sup>23, 52-54</sup> which is consistent with these  
199 statistical results.

200         We find a small but significant increase in PFOS and PFOA (2%,  $p<0.01$ ) with each  
201 additional WWTP within the same HUC. This is consistent with the greater abundance but  
202 smaller quantities of PFASs released by WWTPs.<sup>55</sup> Similarly, results from Valsecchi et al.<sup>51</sup>  
203 show PFAS releases from WWTPs are important but less significant than fluoropolymer  
204 manufacturing facilities in Italy. The number of WWTPs may also be a proxy for other  
205 population-driven PFAS sources.

206 The number of AFFF certified airports is not significantly associated with PFAS  
207 concentrations in the current dataset. This may reflect misclassification bias because the  
208 certification used to identify airports indicates eligibility but not actual use of AFFF. The  
209 UCMR3 database contains limited data for smaller drinking water systems where localized  
210 reports of contamination from airports have been most abundant.<sup>22, 56</sup>

211

### 212 **Current data limitations and future monitoring efforts**

213 The UCMR3 database has several limitations that restrict its predictive power for  
214 identifying U.S. drinking water supplies likely to contain elevated levels of PFASs.  
215 Classification of geospatial data on intakes for public water supplies limits the spatial resolution  
216 of the current dataset and associated statistical models to a radius of 50 km (median radius of  
217 watersheds).<sup>57, 58</sup> Many of the impacted drinking water systems are groundwater systems and  
218 contaminated groundwater plumes are often much smaller than 50 km.<sup>23, 53, 59</sup>

219 Geospatial data are lacking for many potentially important PFAS point sources such as a  
220 wide-range of industries, landfills, biosolids application, and other AFFF-impacted sites where  
221 relatively smaller volumes of AFFF were released<sup>27, 54, 60-67</sup> Data on PFAS releases from smaller  
222 industrial facilities (e.g., plastics, textiles, paper, lubricants) are usually withheld as confidential  
223 business information and little information on airborne emissions is available for characterizing  
224 the importance of atmospheric releases and potential long-range transport. For example,  
225 biosolids application resulted in one of the largest PFAS drinking water contamination in  
226 Europe<sup>68</sup> but could not be included in this analysis because U.S. use data are not available on a  
227 national scale.

228 Sources not included in our spatial analysis are represented by the highly significant  
229 lambda ( $\lambda$ ) coefficients (Table 2). Areas with high model residuals (greater than 1.5 standard  
230 deviation) mean that current information on sources cannot fully explain the high observed  
231 PFAS concentrations. The map of model residuals (Figure S3) can thus be used to guide high  
232 priority sampling regions in future work.

233 We found statistically greater abundance of point sources in watersheds with detectable  
234 PFASs, including AFFF certified airports. However, multivariate spatial regression models did  
235 not show a significant association between AFFF certified airports and concentrations of PFASs  
236 in nearby drinking water. Other studies have reported elevated PFAS concentrations in  
237 groundwater wells adjacent to AFFF certified airports.<sup>22</sup> Small drinking water systems and  
238 private wells may be disproportionately affected by PFASs originating from AFFF use at civilian  
239 airports but representative data for these small drinking water systems are not included in the  
240 UCMR3 program.<sup>69</sup>

241 Approximately 44.5 million U.S. individuals rely on private drinking water wells<sup>70</sup> and  
242 52 million individuals rely on smaller public water supplies (< 10,000 served). The UCMR3  
243 program includes 0.5% testing incidence for smaller public water supplies<sup>71</sup> and no testing of  
244 private wells, meaning that information on drinking water PFAS exposures is therefore lacking  
245 for almost 1/3 of the U.S. population.

246

## 247 **Acknowledgements**

248 We acknowledge financial support for research at Harvard from the Smith Family Foundation  
249 and a private donor. We thank Marcia Castro (Harvard) for her feedback on an earlier version of  
250 this manuscript and Jahred Liddie (Harvard) for his assistance with the sensitivity analysis. T.B.

251 was supported by the U.S. National Institute for Environmental Health Sciences (NIEHS)  
252 Superfund Research Program (Grant P42 ES004705) and the Superfund Research Center at  
253 University of California, Berkeley. This article was reviewed in accordance with the policy of  
254 the National Exposure Research Laboratory, U.S. Environmental Protection Agency, and  
255 approved for publication.

256

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477

478 Table 1. Mean abundance of point sources within 8-digit hydrologic unit codes (HUCs) with  
 479 drinking water PFAS concentrations above and below method reporting limit in the UCMR3  
 480 program.

Compound	Mean abundance <sup>a</sup> within 8-digit hydrologic unit codes			
	Major industrial sites <sup>b</sup>	Military fire training areas	AFFF certified airports	WWTP <sup>c</sup>
<i>PFBS</i>				
<90 ng/L (n=1587)	0.01	0.15	0.29	
>90 ng/L (n=14)	0.21	0.71	0.50	
<i>p-value</i> <sup>d</sup>	0.206	0.105	0.148	
<i>PFHxS</i>				
<30 ng/L (n=1507)	0.01	0.13	0.27	
>30 ng/L (n=94)	0.06	0.60	0.63	
<i>p-value</i>	0.056	<0.001	<0.001	
<i>PFHpA</i>				
<10 ng/L (n=1509)	0.01	0.13	0.26	
>10 ng/L (n=92)	0.09	0.57	0.67	
<i>p-value</i>	0.016	<0.001	<0.001	
<i>PFOA</i>				
<20 ng/L (n=1473)	0.01	0.13	0.26	
>20 ng/L (n=128)	0.05	0.52	0.56	
<i>p-value</i>	0.038	<0.001	<0.001	
<i>PFOS</i>				
<40 ng/L (n=1487)	0.01	0.13	0.26	
>40 ng/L (n=114)	0.05	0.54	0.57	
<i>p-value</i>	0.064	<0.001	<0.001	
<i>PFNA</i>				
<20 ng/L (n=1586)	0.01	0.15	0.28	
>20 ng/L (n=15)	0.13	1.13	1.13	
<i>p-value</i>	0.366	0.014	0.008	

508 <sup>a</sup> Mean abundance is calculated as the mean numbers of point sources within HUCs with PFASs  
 509 above or below-detection.

510 <sup>b</sup> Only the major industrial sites participating in US EPA 2010/2015 PFOA Stewardship Program  
 511 were included.

512 <sup>c</sup> Wastewater treatment plant.

513 <sup>d</sup> Two-sample t-test *p-values*.

514  
 515  
 516

517 Table 2. Spatial regression models for drinking water PFAS concentrations as a function of  
 518 abundance of point sources.  
 519

Compound	Major industrial sites <sup>a</sup>	MFTAs <sup>b</sup>	AFFF certified airports	WWTPs <sup>c</sup>	$\lambda^d$	$R^2$
<i>PFHxS</i>						
Coefficient <sup>e</sup>	24%	20%	-13%	1%	94%	0.62
<i>p-value</i> <sup>f</sup>	0.249	0.002	0.073	0.045	<0.001	
<i>PFHpA</i>						
Coefficient	10%	10%	-2%	0.5%	72%	0.40
<i>p-value</i>	0.569	0.155	0.761	0.436	<0.001	
<i>PFOA</i>						
Coefficient	81%	10%	-6%	2%	52%	0.38
<i>p-value</i>	<0.001	0.111	0.353	0.006	<0.001	
<i>PFOS</i>						
Coefficient	46%	35%	-6%	2%	79%	0.46
<i>p-value</i>	0.124	<0.001	0.512	0.007	<0.001	

520  
 521 <sup>a</sup> Only the major industrial sites participating in US EPA 2010/2015 PFOA Stewardship Program  
 522 were included.

523 <sup>b</sup> MFTA = military fire training area.

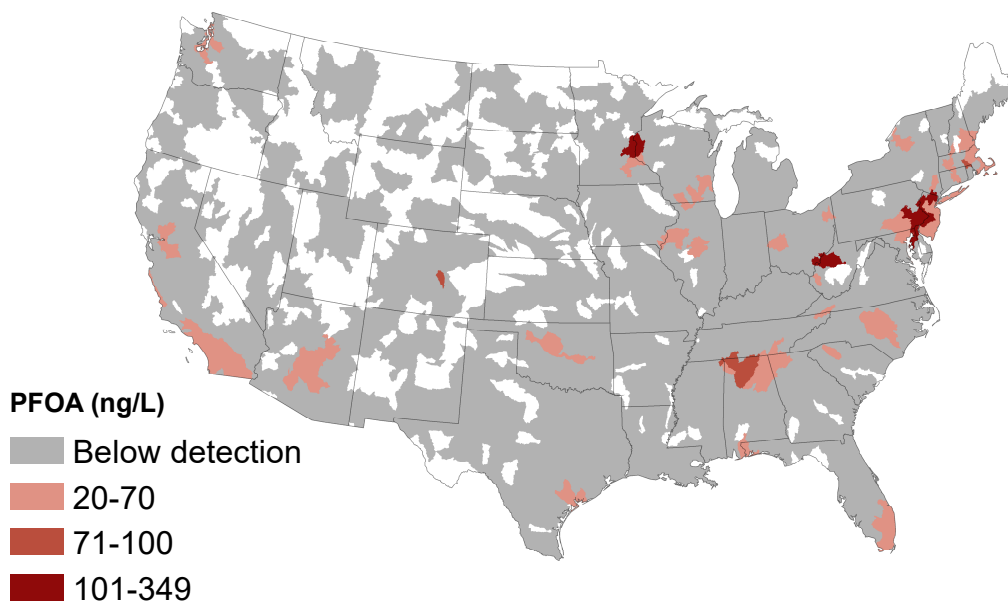
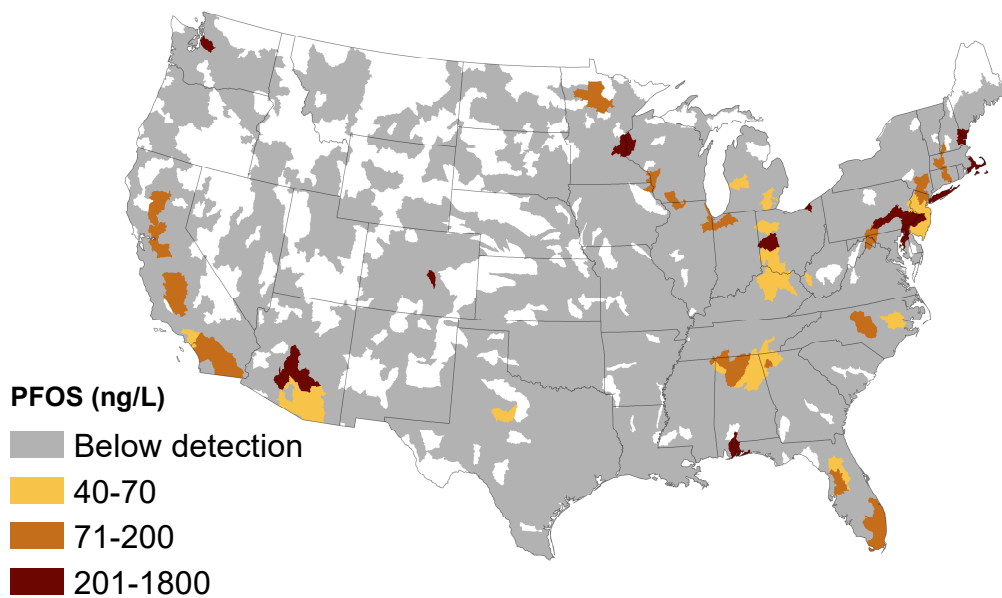
524 <sup>c</sup> WWTP = wastewater treatment plant.

525 <sup>d</sup> Coefficient for the spatial error term characterizing spatial influence.

526 <sup>e</sup> Results have been transformed to reflect expected changes in drinking water concentrations per  
 527 increase in the abundance of different sources. Positive coefficients in the results indicate  
 528 increasing concentrations with increasing abundance of point sources within the same hydrologic  
 529 unit.

530 <sup>f</sup> *p*-values for spatial error regression model. The spatial error term is used to incorporate spatial  
 531 autocorrelation structures into a linear regression model.

532  
 533



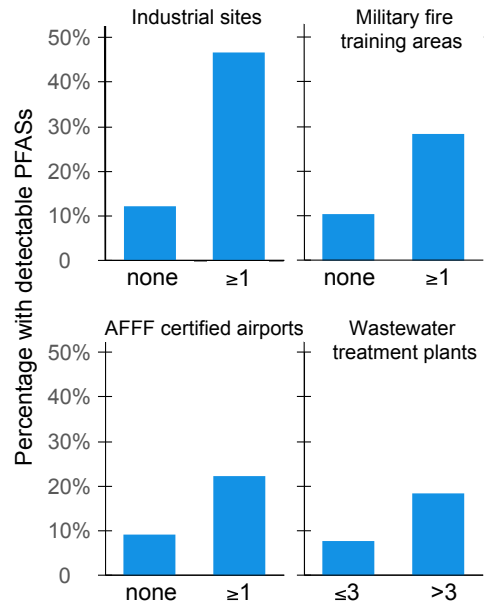
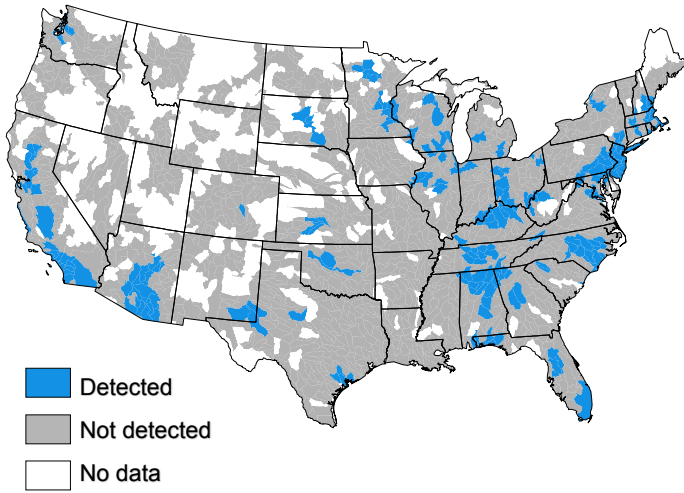
534

535 Figure 1. Hydrologic unit codes (8-digit HUCs) used as a proxy for watersheds with detectable  
 536 PFOA and PFOS in drinking water measured in the US EPA's UCMR3 program (2013-2015).

537 Blank areas represent regions where no data are available.

538

### Hydrological units with detectable PFASs



539

540 TOC Art