1   **SUPPLEMENTARY MATERIAL**

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3   **Evaluation of a national data set for insights into sources, composition,**

4   **and concentrations of per- and polyfluoroalkyl substances (PFASs) in U.S.**

5   **drinking water**

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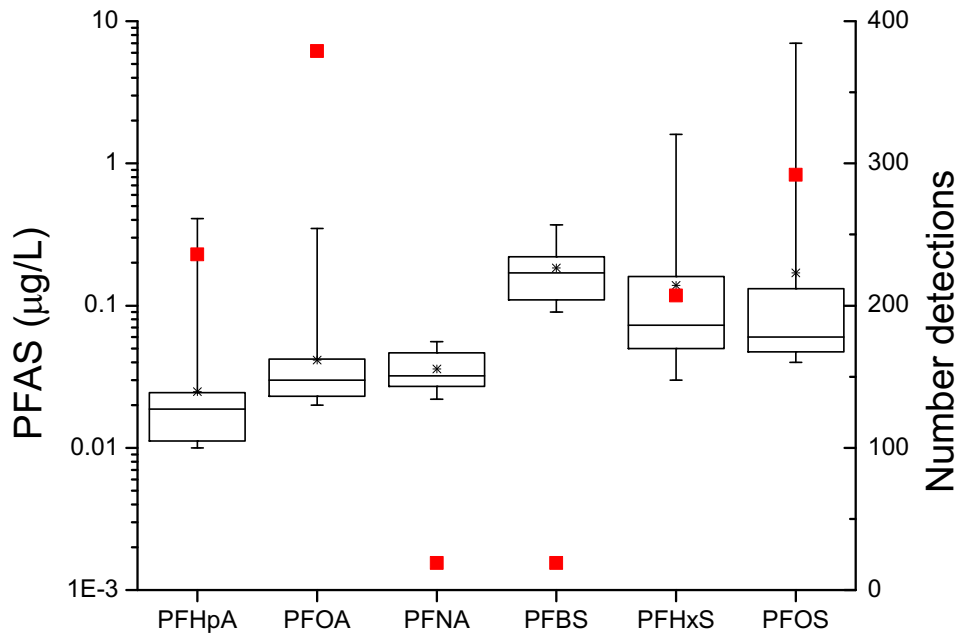
Figure S.1. Concentrations of each UCMR3 PFAS analyzed (left y-axis) and total number of detections (right y-axis). The lower and upper ends of the whisker represent minimum and maximum detections, respectively. Asterisks represent the average value. Lower and upper ends of the box represent the 25th and 75th percentile, respectively, and the center line is the median value. Red squares represent the number of detections per compound and correspond to the right-hand y-axis.

40 **Table S.1.** PFAS Co-Occurrence Matrix

	PFOS	PFOA	PFHxS	PFHpA	PFBS	PFNA
Odds Ratio for Association with Other PFAS						
PFOS	--	**216**	**876**	**295**	**371**	**46**
PFOA	**216**	--	**242**	**407**	**538**	**57**
PFHxS	**876**	**242**	--	**389**	**107**	**65**
PFHpA	**295**	**407**	**389**	--	**463**	**94**
PFBS	**371**	**538**	**107**	**463**	--	0
PFNA	**46**	**57**	**65**	**94**	0	--
Odds Ratio for Association with Other UCMR3 Compounds (Selected)						
1,4-Dioxane	**13.2**	**14.2**	**4.0**	**11.4**	**8.3**	**8.3**
HCFC-22	**5.0**	**4.5**	**3.7**	**3.0**	0	0
Chromium-6	**2.9**	**4.1**	**2.2**	**2.6**	-	1.2
1,2,3-TCP	0	0	0	0	0	0
Odds Ratio for Association with Groundwater (vs. Surface Water)						
Groundwater	**2.1**	**1.8**	**4.1**	1.0	**0.4**	**11.6**
Odds Ratio for Association with Large Systems (vs. Small Systems)						
Large Systems	**4.7**	**9.2**	**5.0**	**5.7**	-	1.7

41 Notes: (1) Reflects results of categorical analysis (chi square) on all samples collected during UCMR3 (i.e.,
42 not segregated by PWS where sample was collected); (2) All relationships in **bold** were significant based on
43 p-value < 0.05; (3) Odds ratio represents the odds that a detection will occur given a particular condition,
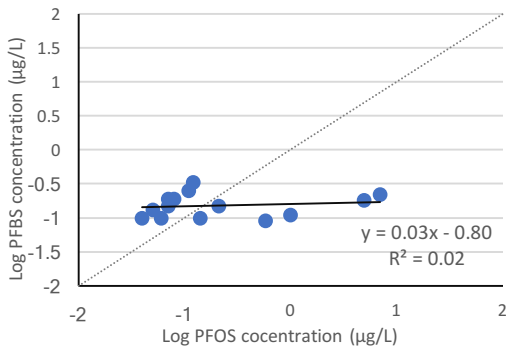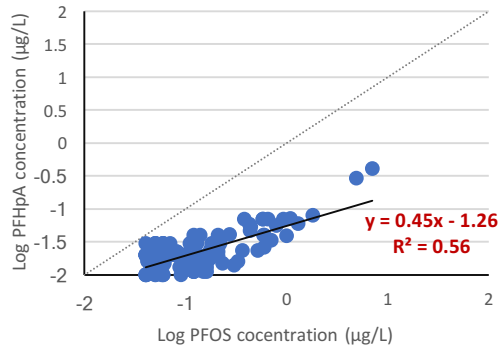44 compared to the odds of that outcome occurring in the absence of that condition.
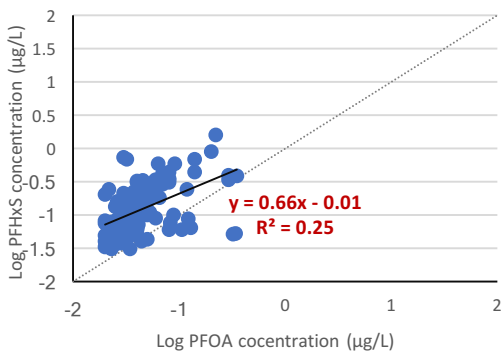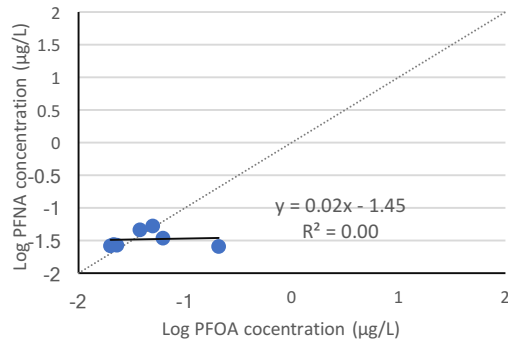
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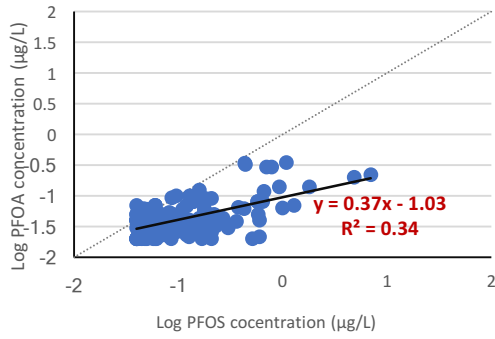
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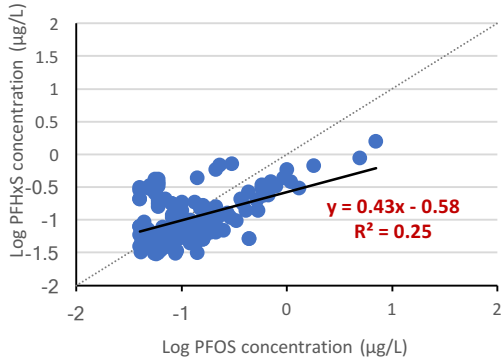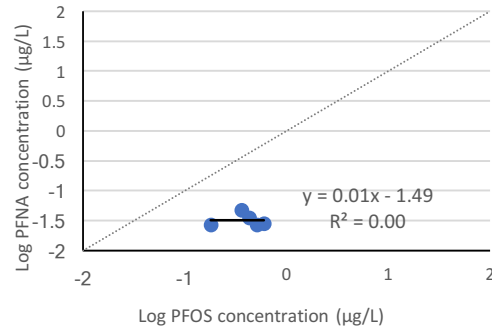
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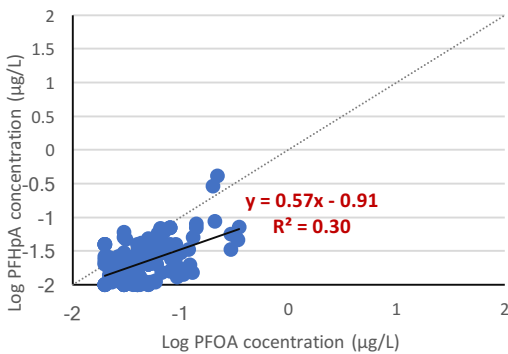
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57 **Figure S.2.** Concentration cross-plots for all PFASs in UCMR3 that include all
58 samples with detections shown relative to a 1 to 1 ratio. Results of linear
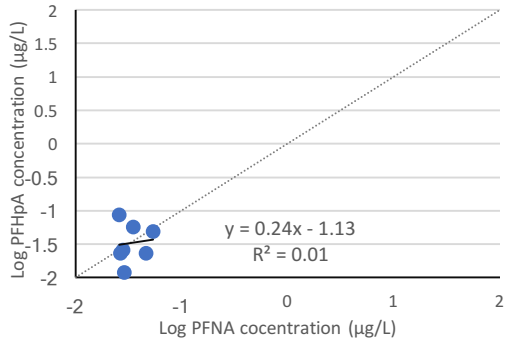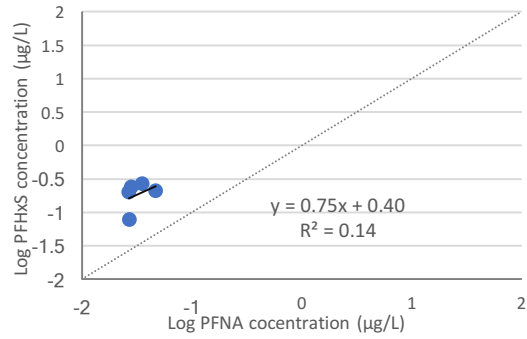59 regression are shown for reference purposes, and it should be noted that there
60 were no samples with detections of both PFBS and PFNA. Linear regression
61 equations and R^2 values shown in red represent slopes statistically different from
62 zero ($p<0.05$)
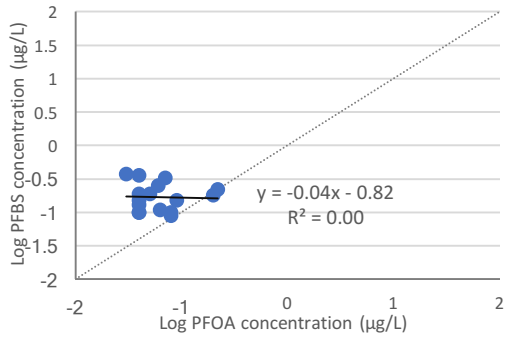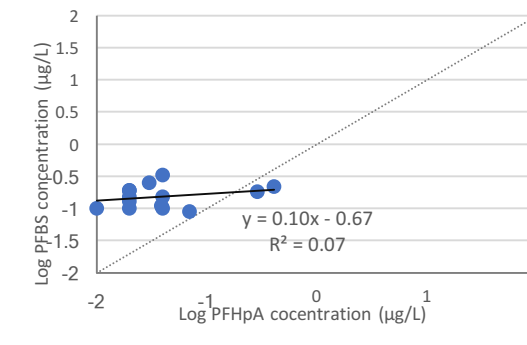
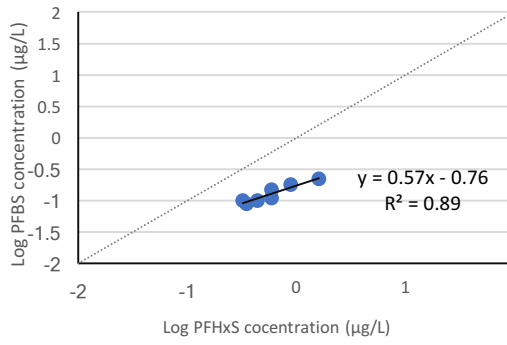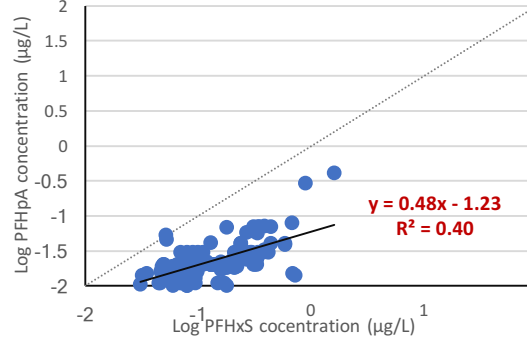**Figure S.2 *(continued)*.** Concentration cross-plots for all PFASs in UCMR3 that include all samples with detections shown relative to a 1:1 ratio. Results of linear regression are shown for reference purposes, and it should be noted that there were no samples with detections of both PFBS and PFNA. Linear regression equations and R^2 values shown in red represent slopes statistically different from zero ($p<0.05$)
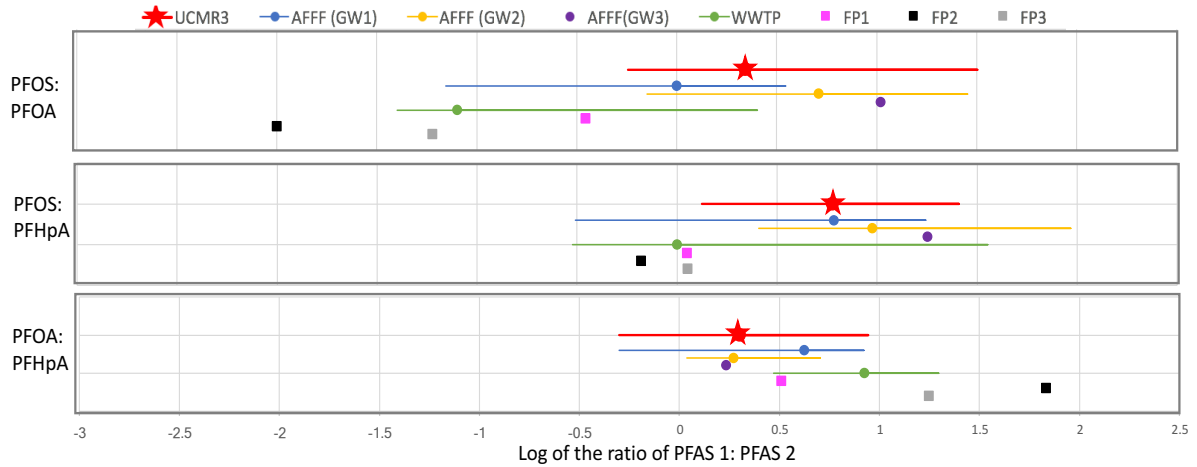
Figure S.3.   Logs of ratios of PFOS:PFOA, PFOS:PFHpA, and PFOA:PFHpA compared to ratios in previous studies. Lines represent the range of ratio values (where available) and circles denote median values. Squares denote ratios of maximum groundwater concentrations for sites where ranges were not reported. Sources types are AFFF-impacted groundwater (AFFF (GW)) (Anderson et al. 2016; Houtz et al. 2013; Hull et al. 2017), wastewater treatment plant effluent (WWTP) (Ahrens et al. 2009), and fluoropolymer manufacturing (FP) (Bach et al. 2017; Dauchy et al. 2012; Oliaei et al. 2013).

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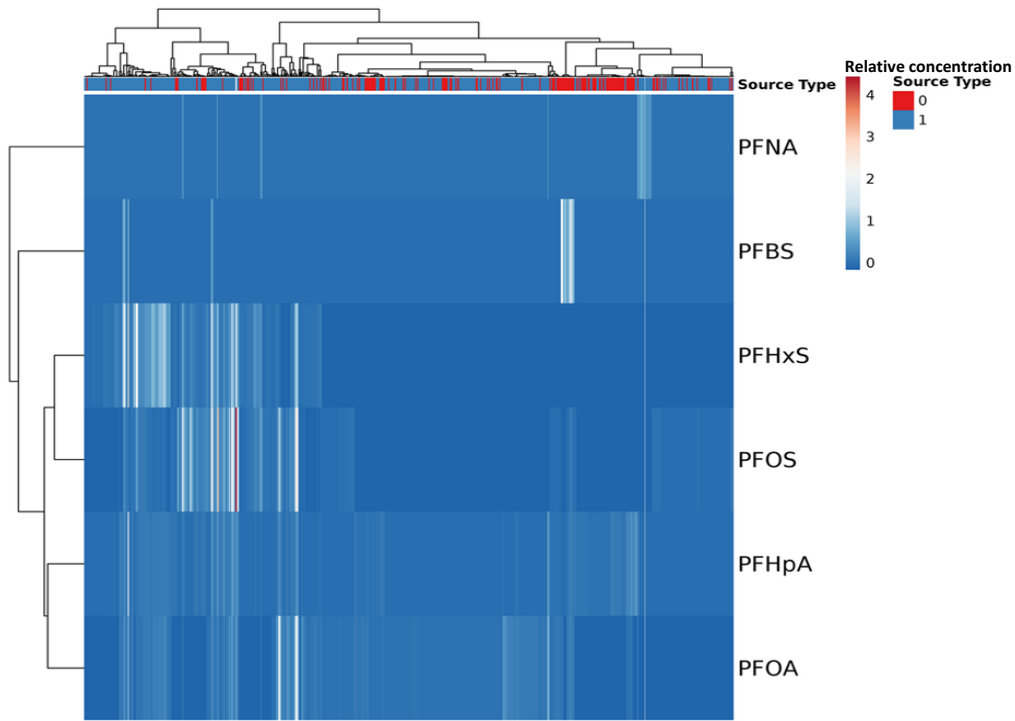
Figure S.4. Results of hierarchical cluster analysis using PFAS concentration data. Columns display log normalized concentrations of individual samples. Source type is either surface water (0; red) or groundwater (1; blue). Clustering of compounds is shown vertically; clustering of samples is shown across the top horizontally. Relative concentration levels are reflected in the heat map for each PFAS.

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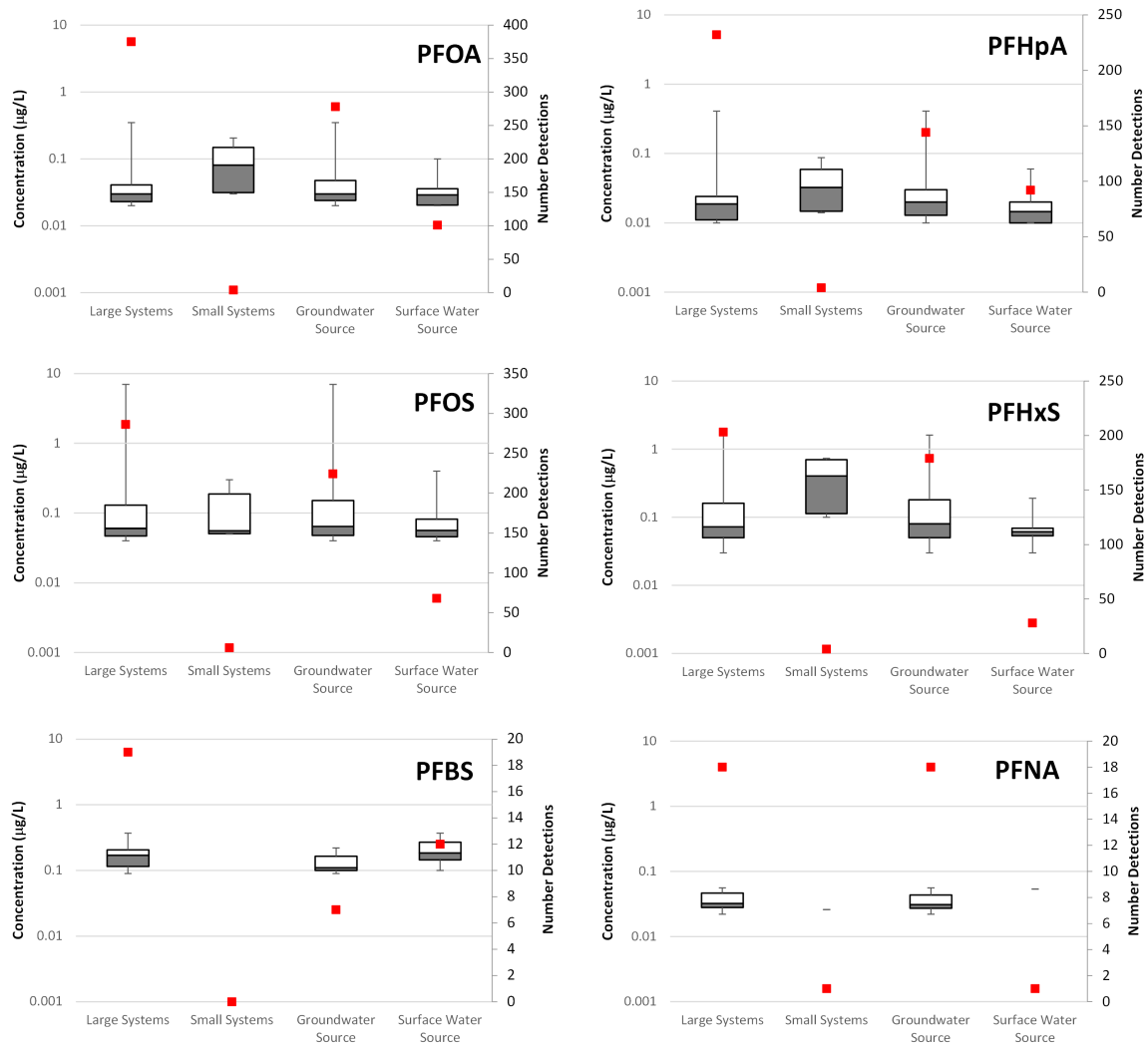
Figure S.5. Concentration of individual PFASs as function of system size and source water (box and whisker, left axis) and the number of detections in each category (red squares, right axis). Includes all samples with detections of one of more PFAS.

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a. Groundwater

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b. Surface water
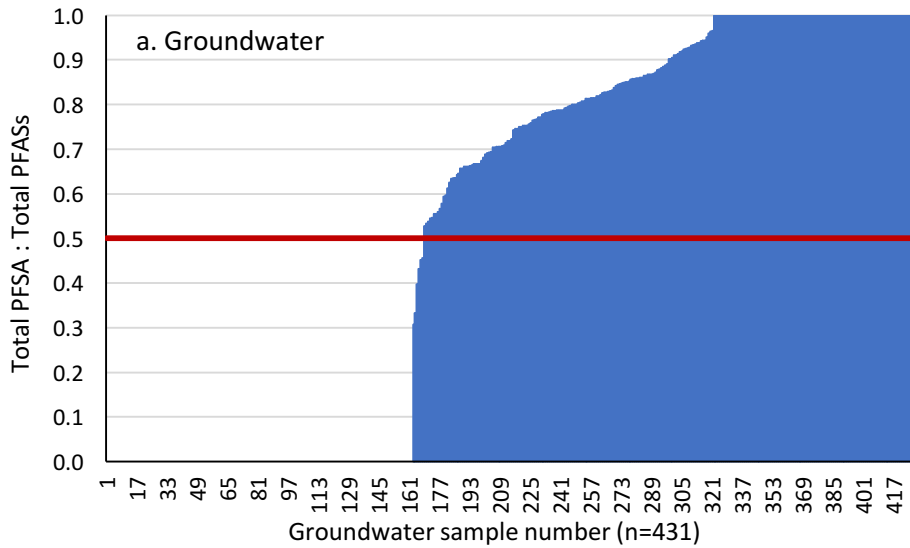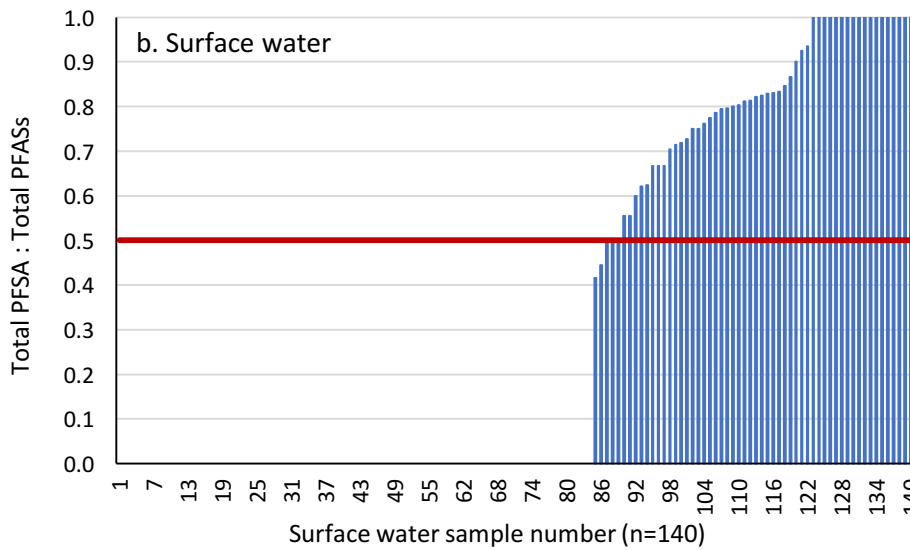
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122 **Figure S.6.** Influence of source water type on the composition of perfluoroalkyl
123 sulfonates (PFSAs) in UCMR3 samples. Each sample with detections was
124 assigned an arbitrary sample number (x-axis) and the ratio of total PFSAs
125 (PFOS+PFHxS+PFBS) to total PFASs (sum of all 6 UCMR3 PFASs) was plotted
126 on the y-axis. Samples with a ratio of 0 contained PFASs but no PFSAs (i.e.
127 they were comprised of 100% perfluoroalkyl carboxylates). Samples with ratios
128 greater than the red line are dominated by PFSAs (i.e. ratio of PFSA to PFASs
129 exceeds 0.5).

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S.1 Aggregation of data for temporal analysis. To investigate temporal trends,
sample data from all PWSs was aggregated and concentrations over time were
plotted for individual PFASs. Aggregation was necessary because individual
PWSs generally followed UCMR3-recommended monitoring protocol of 4 quarterly
samples for 1-year, but required samples may have been collected from a different
facility or sampling points within the system in each quarter. This limited the
number of data points and duration of the monitoring period, such that establishing
trends at the facility- or PWS-level was not possible. Linear regression was used
on the compound-specific aggregated dataset to determine if there was a temporal
concentration trend. For each PFAS, there was a high degree of variability, and
the resulting mix of slopes of regression lines were modest and included both
increasing and decreasing trends (data not shown).

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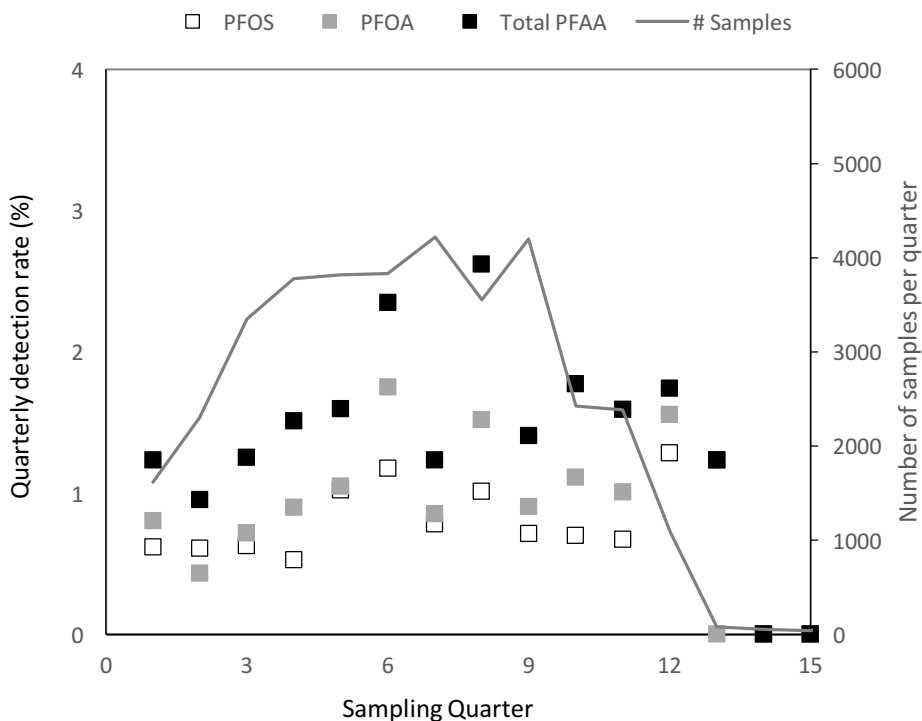
Figure S.7. PFAS, PFOA, and PFOS Occurrence Trend Using Quarterly Results
for All Samples. Data from all UCMR3 samples with analysis of one or more
PFAS were grouped by monitoring period (quarterly). The Mann-Kendall test
was used to evaluate the trend in detection frequency for data from the first 12
quarters (2013-2015). Data from the last 4 quarters (2016) were not used due to
the low number of samples per quarter (<2% of total). Squares are detection
rates and correspond to the left y-axis; lines are the number of samples per
quarter and correspond to the right y-axis.

S.2 Transience in PFAS detections. Given that PFASs included in UCMR3 were

all perfluoroalkyl substances, the assumption was that for a specific PWS with a

detection of a certain PFAS, the PWS in question would routinely detect that PFAS

in all samples collected during UCMR3 monitoring. Instead, individual PFASs

frequently were only detected in a single sample from a PWS (**Figure S.8**). The

percentage varied widely depending on the PFAS analyzed, ranging from 15% for

163   PFHxS to 64% for PFNA. A portion of this is explained by the fact that many PWSs

164   sampled multiple different facilities during UCMR3.   However, the lack of

165   consistent PFAS detection across all facilities associated with a PWS also

166   highlights the unexpectedly sporadic nature of many of these PFAS detections. In

167   some cases, this can likely be attributed to analytical variability, particularly for

168   concentrations that approach the MRL. Impacts of analytical variability and the

169   MRL are discussed in Section 4. However, sporadic detections may also be

170   attributable to release histories, PFAS fate and transport characteristics, and/or

171   hydrogeologic characteristics of the source aquifer or surface water body.

172   Uncertainty regarding the causes of this variation is an important knowledge gap,

173   especially in a regulatory capacity since it may inform the sampling approaches,

174   risk evaluation, and mitigation measures required to obtain data that fully represent

175   PFAS exposures in drinking water.

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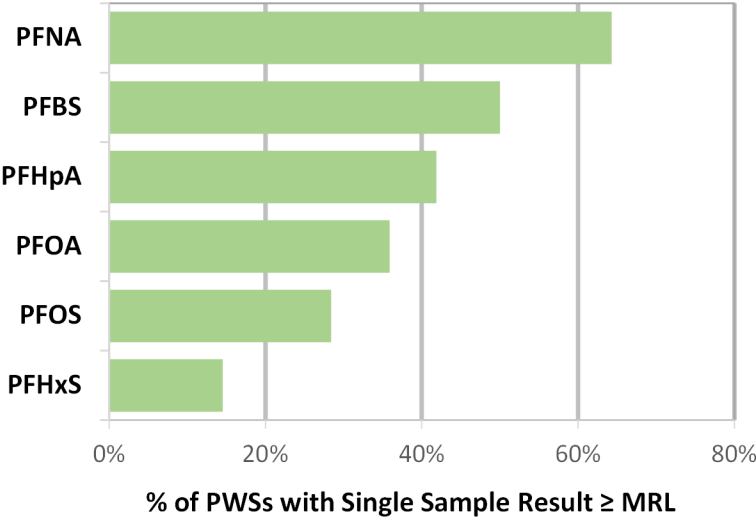
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178   **Figure S.8.** Percentages of public water systems with single sample detections of
179   PFASs.

180   **References Cited**

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