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Article

Locating carcinogenic per- and poly-fluoroalkyl substances in Santa Clarita groundwater

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SUMMARY

Per- and poly-fluoroalkyl substances (PFASs) are a widespread type of industrial waste notorious for their carcinogenic effects. Specifically, one of the main routes of human exposure is through drinking water. As drought conditions worsen around the world, preserving clean and safe water has become crucial, which necessitates pinpointing different water contamination pathways that can occur. Santa Clarita is an example of a water-stressed region that has been struggling with PFAS contamination and is unique in that residents are mostly reliant on groundwater. The region has a municipal landfill with multiple complaints filed about its leachate production. This study focused on identifying potential sources of the PFASs found in Santa Clarita's public wells. We hypothesized that the biggest source would be the leachate produced from a nearby municipal waste landfill infiltrating the groundwater. To identify the sources, we used a multivariate factor tool specialized for source apportionment called Positive Matrix Factorization. While our hypothesis was not confirmed with conclusive evidence, we identified three potential sources of PFASs, including domestic waste, fire extinguisher materials, and food packaging. These results indicated that many different physical sources, including leachate, may be contributing to the PFASs derived from domestic waste. We concluded that further research may be necessary to derive a more solid conclusion. Our study could inform California lawmakers about how to address the growing issue of PFASs in Santa Clarita.

INTRODUCTION

Per- and polyfluoroalkyl substances (PFASs) have been mass-produced since the 1950s for use in various products (1). PFASs are particularly known for their strong carbon-fluorine bonds, a chemical characteristic that makes them highly resistant to degradation by heat and water. This durability led to their widespread use in everyday items, such as non-stick pans and raincoats (1). Beyond cooking ware and clothing, the aerospace, automotive, construction, and electronics industries used PFAS as well (1). However, the same resistive properties that make PFASs so effective also cause them to bioaccumulate in both the human body and the environment (1). This is concerning when considering PFASs' highly toxic impacts on human health, such as increased risks of some cancers, high cholesterol, high blood pressure during pregnancy, obesity, and type two diabetes in females, as well as reduced vaccine effectiveness and infant birth weights (2, 3). Although discussions for advisories and action plans for dealing with PFASs in water began in the 2000s, it was not until April of 2024 that the first legally enforceable drinking water standard was implemented nationally in the United States (U.S.) (4).

People are exposed to PFASs in many ways, and one of the major pathways is through drinking water (5). PFASs enter public water systems in a variety of ways, including leachate contamination, which is when liquids secreted from landfills pollute a water source (6-9). Leachate pollution is especially detrimental for communities that rely on groundwater or other local water sources. As of 2021, groundwater supplies about 35% of the U.S.'s drinking water (10). However, reliance on groundwater is disproportionately high in drought-prone states like California, which alone accounts for 21% of total U.S. groundwater use (11). Groundwater became more important for water-stressed southern Californian cities, such as Santa Monica, as water importation, a historical solution, is costly and has a huge ecological footprint (12). With climate change intensifying, groundwater dependence has also risen in other states, including Texas, Idaho, Arizona, Utah, and Mississippi (11). Like Santa Monica, Santa Clarita, a wealthy suburban city in Los Angeles County, faces water stress, relying on local groundwater for half of its water supply (13). Currently, a single landfill, Landfill C, provides for all of the Santa Clarita Valley's municipal wastes. This landfill was controversial due to toxic air emissions (14, 15). Santa Clarita had 27 public wells where chemical concentrations could be measured, and many of these wells were close to the Santa Clara River, one of the main natural water sources that recharges the valley's drinking water supply (16). Because the Santa Clara River is connected to the two main water basins from which the community draws groundwater, contamination of the river would impact the drinking water directly (17, 18). To monitor risks like these, the California Water Department compiled data for 19 PFASs from the 27 wells since 1 March 2023, every quarter on GeoTracker, which is California's database for water information (19).

There are three main analytical strategies for source apportionment, i.e., identifying the sources of chemicals, in PFASs: Unmix, principal component analysis (PCA), and Positive Matrix Factorization (PMF). Unmix is an analysis method that works by identifying data points (specific samples at a given time and place) where a certain factor contributes zero contaminant (20-22). A study in the Hai River basin in China used Unmix to identify the sources of PFASs in self-measured surface water and sediments, finding metal plating, paper and textile manufacturing, fire extinguishing, and food packaging as the main sources of PFASs in the

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Hai River basin (22). Another method used for identifying the source of chemicals is PCA, which uses dimension reduction (a mathematical estimation) to reduce the amount of information while preserving the most information in order to identify a few factors that could explain the data (21, 23). It has been used in conjunction with hierarchical clustering to examine New York's surface water, effectively identifying textile mills, waste, and metal plating as the main sources of PFASs in New York's surface water (23).

However, Unmix and PCA-based methods are limited in different ways. For Unmix, a crucial assumption is that all sources will have a period without further contamination. This assumption, though appropriate for source apportionment of sediment and surface water, may not be fit for groundwater. Many different water systems drain into groundwater, so it may be impossible for Unmix to identify a time and place where a certain source does not contribute to the PFASs in groundwater, which could have created erroneous results. In contrast, PCA has typically been used in studies that collected their own data, which we were unable to do (23, 24). Public data sets sometimes contain missing data. This is best handled in PCA by pairwise deletion, which could affect data reliability (25). Pairwise deletion also restricts the data size for data sets with significant proportions missing.

In comparison, PMF enables users to input uncertainties to the model, which makes handling outliers and other "high-noise" points much easier (20). This is especially important for dealing with public data sets with missing data, as more data can be inputted without increasing errors. PMF determines the factors —hypothetical contamination sources— and their factor fingerprints, which represent the contributions of these sources to the overall PFAS species' concentration in the region (**Figure 1**). The user determines the number of factors using statistical tests that indicate the best number of factors, while the model itself is responsible for identifying each factor's contribution levels for each of the data species. The factors are spatially separated and affect different locations differently. They can, therefore, be related to real-world contamination sources through logical inference, associating factors with a specific physical source based on their PFAS composition and other factors (26-31). This method has been successful in diverse geographical contexts from the Arctic Ocean to the Chinese and South Korean coasts (26-31). A study on the sediments of the Arctic Ocean identified fluoropolymer manufacture, degradation of precursors from product use of textiles, and possibly local fire extinguisher materials (28). The second source, for instance, was identified through logical inference: PMF found a factor high in perfluoro-octane sulfonate (PFOS), a species of PFASs that tends to uniformly distribute in the environment in which they are placed, which was the case for the PFOS in the Arctic Ocean sediments. This supports their inference because the degradation of precursors should be uniform in their studied region. Other PMF-based studies on China and South Korea coastal areas, like the Bohai Sea and Yellow Sea, identified the same sources as those found in the earlier reviewed Unmix-based study on the Hai River Basin, a river connected to the Bohai Sea (22, 26). While the studies, using PMF and Unmix, identified the same sources, there were noticeable variations in the contributions from each source: the contribution of food packing/textile treatments was 17.7% in the Unmix study but was 33% in the PMF study.

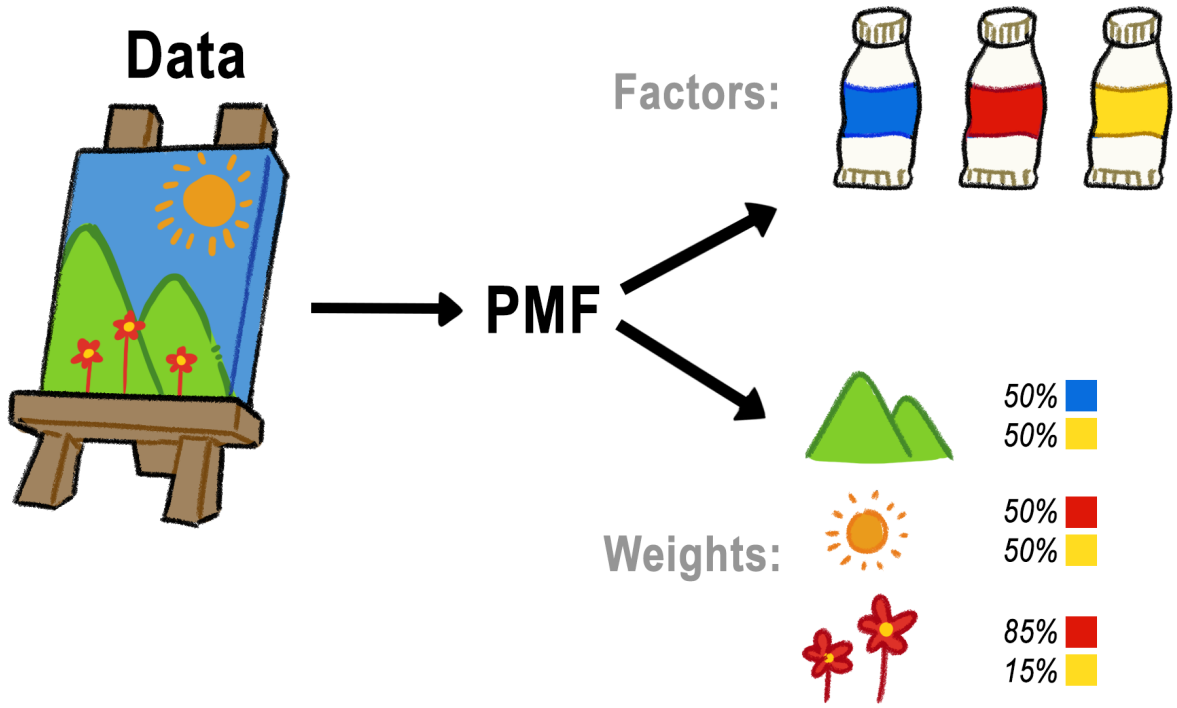

Figure 1. Positive Matrix Factorization's (PMF's) functions and capabilities. A complete data set represented by a painting, is split into factors and weights. The factors represent the raw "sources" used to create the data set, which are the three primary colors in this figure. The weights represent how much of these sources comprise the different parts of the data set.

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The differences between these studies can make targeting sources of PFASs much more difficult for lawmakers, which shows that more research should be carried out to improve our understanding of the sources of PFASs and the differences between source apportionment methods.

As a suburban city, Santa Clarita Valley was not a closely studied location for water quality as opposed to more densely populated cities in the county, such as Los Angeles. At least five different studies focusing on water quality have been conducted in Los Angeles; however, no water quality studies have been found for Santa Clarita and the Santa Clara River (32-36). Given the lack of studies focusing on this specific location, this study will be especially meaningful for its residents. As previously mentioned, landfill leachate can leach into groundwater. Thus, we hypothesized that a nearby municipal waste landfill, Landfill C, would be the biggest contributor to the PFAS contamination of Santa Clarita. We tested this hypothesis through generating a PMF model from the PFAS data of the public wells. The hypothesis that Landfill C would be the biggest contributor of PFASs in Santa Clarita was not supported by our findings. We found that there were three main factors contributing to the PFAS contamination in Santa Clarita: domestic waste, fire extinguisher materials, and food packaging, which contributed 45.4%, 17.1%, and 37.5%, respectively. These findings could inform future legislation regarding PFASs control in California (37-40). Understanding PFASs' sources will improve the community's health and potentially aid future studies.

RESULTS

We hypothesized that Landfill C would be the biggest contributor to PFAS contamination in the public wells of Santa Clarita. In order to investigate this, we retrieved quarterly data from our local water agency and additional data on PFAS concentrations within the wells recorded from 1 March 2023 to 2 May 2024 from California's GeoTracker website (19). We excluded data from five wells due to the unavailability of data after 1 March 2023. While we had data on 19 species of PFASs, we excluded 11 of them as they had concentrations less than the Minimum Detection Limit (MDL) for at least three-fourths of the wells. The remaining data used still included missing or below-MDL data points, which we treated to minimize their negative impact on the model's accuracy. After the initial PMF model was generated using this data, it was tuned and verified for accuracy using the analysis methods displacement and Fpeak (**Table S3, S4, Appendix**). The fitness of data for each PFAS species was also assessed through computing the coefficient of determination (r^2) values of each species, which were 0.850, 0.857, 0.840, 0.967, 0.993, 0.860, 0.964, and 0.883, for perfluorobutanoic acid (PFBA), perfluorobutane sulfonate (PFBS), perfluoroheptanoic acid (PFHpA), perfluorohexanoic acid (PFHxA), perfluorohexane sulfonate (PFHxS), perfluorooctanoic acid (PFOA), PFOS, and perfluoropentanoic acid (PFPeA) respectively, rounded to 3 d.p. An r^2 value greater than 0.65 was regarded as an indicator of good fitness of data (29). Using bootstrapping, we identified three factors, or contamination sources, responsible for the PFASs found within the wells (**Table S2, Appendix**).

To better understand the influences of each of the three factors, we analyzed how much each factor contributed to each PFAS species as well as the total PFASs (**Figures 2, 3**). As presented below, we categorized the levels of PFASs in each factor into high (>50%), medium (25-50%), and low (<25%) based on criteria adopted in previous studies (26-31).

Factor 1 was high in PFOS, comprising 45.4% of the total PFASs, and had a medium amount of every other PFAS (**Figures 2, 3**). Factor 2 was high in PFHxS, medium in PFBS, and made up 17.1% of the total PFASs (**Figures 2, 3**). Factor 3 was high in PFHxA, PFPeA, and PFHpA; medium in PFBA, PFBS, and PFOA. Factor 3 made up 37.5% of the total PFASs (**Figures 2, 3**).

We additionally determined if certain wells were more exposed to specific species of PFASs (**Figure 4**). We specifically looked for which wells were highest in PFOS (highest species in Factor 1), PFHxS (highest species in Factor 2), and PFHxA (second highest species in Factor 3). S8, N13, and the wells in the central Santa Clarita Region were very high in PFOS, PFHxS, and PFHxA, respectively.

To investigate Landfill C's impact on PFAS levels, we mapped out the location of the wells and Landfill C to analyze the relationship between their proximities and changes in PFAS levels (**Figure 5**). We also wanted to compare the PFAS concentrations in Landfill C to the Environmental Protection Agency's (EPA) recommended concentrations of PFASs. As of 10 April 2024, the EPA recommends less than 4 ng/L for PFOA and PFOS and less than 10 ng/L for PFHxS (41). The sum of the concentrations of the 8 PFAS species we studied (Σ8PFAS) in each well ranged from 10.5 ng/L (well: PT3; date: 2/15/2024) to 212 ng/L (well: S8; date: 6/21/2023), and the mean level was 84.68 ng/L since 1 March 2023 (**Table S1, Appendix and Figure S1, Appendix**). Many data points exceeded these recommended values: 96/107 data points exceeded the values for PFOA, 94/107 points for PFOS, and 38/107 points for PFHxS. We calculated the medians for PFOA, PFOS, and PFHxS, which were 13, 12, and 7.4 ng/L, respectively (**Table S1, Appendix**). The averages for PFOA, PFOS, and PFHxS were 13.4, 15.6, and 9.2 ng/L, respectively (**Table S1, Appendix**). This indicated that Santa Clarita's groundwater had PFAS concentrations that exceeded the EPA PFAS recommendations.

DISCUSSION

Through our study, we aimed to identify the main sources of PFAS contamination in Santa Clarita's public wells with the hypothesis that leachate from landfill C would be the biggest source. We processed available quarterly data of PFAS levels in wells recorded from 1 March 2023 to 2 May 2024 using a PMF model to identify factors (sources of contamination). Factors were attributed to a specific physical source through analyzing what key types of PFAS each factor contributed to. Our study revealed alarming PFAS contamination in Santa Clarita, particularly the central region, and, through PMF analysis, we identified three theoretical factors with distinct PFAS fingerprints. Factor 1 was high in PFOS, Factor 2 in PFHxS, and Factor 3 in PFHxA, PFPeA, and PFHpA. We used these features to identify the physical source of each factor through logical inference, with the detailed spatial variation of PFASs in Santa Clarita considered. Our findings did not support the hypothesis that Landfill C is the biggest contributor to PFASs in Santa Clarita.

The results showed that Factor 1 was mostly comprised of PFOS. This PFAS has mostly been used for metal plating, domestic waste, and food packaging (24, 26, 31). However, factors representing food packaging sites have been found to

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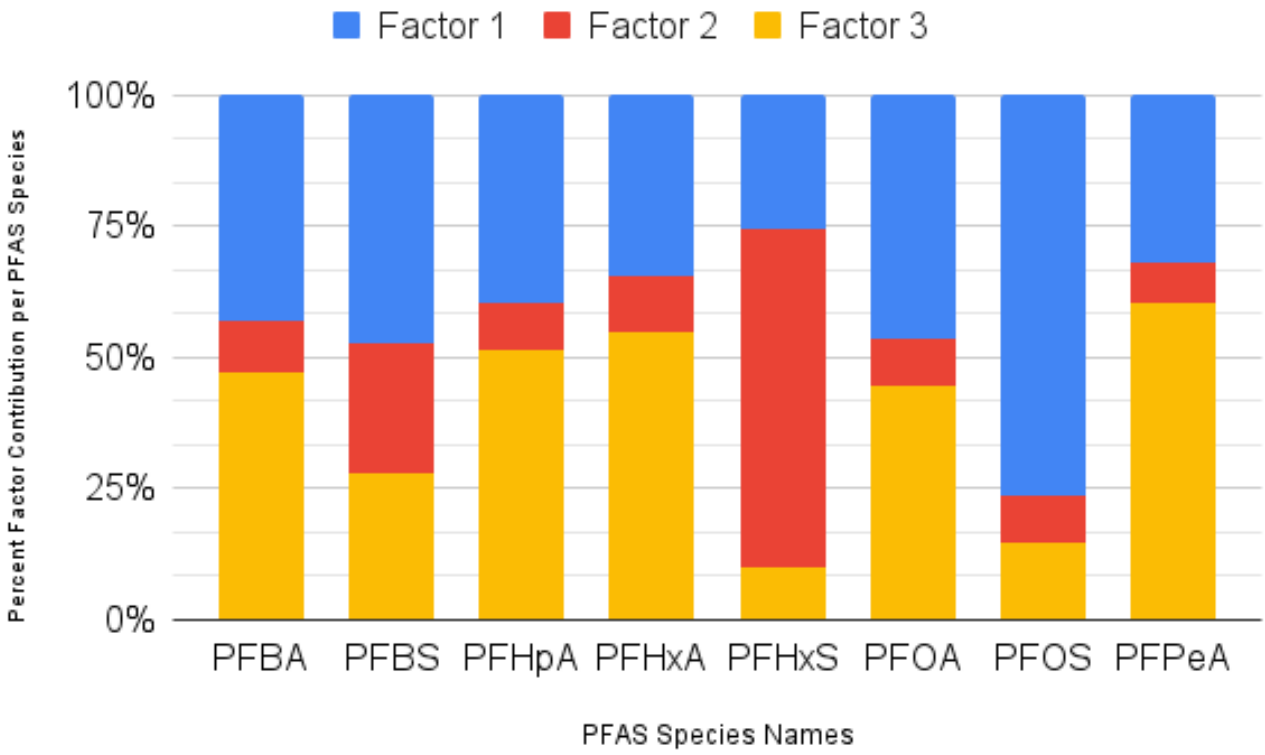

Figure 2. Contributions of three contamination sources to eight PFAS species. Each factor's, or contamination source's, contributions to each of the eight PFAS species including perfluorobutanoic acid (PFBA), perfluorobutane sulfonate (PFBS), perfluoroheptanoic acid (PFHpA), perfluorohexanoic acid (PFHxA), perfluorohexane sulfonate (PFHxS), perfluorooctanoic acid (PFOA), perfluorooctane sulfonate (PFOS), and perfluoropentanoic acid (PFPeA) are shown. Fpeak used was -0.3.

be high in PFBS and short-chain PFCAs, which are lacking in Factor 1 (42). Factor 1 was not high in PFBS and PFCAs, so Factor 1 was concluded not to be food packaging. However, it should be noted that excluding food packaging as the manifestation of Factor 1 does not exclude the possibility of food packaging representing Factor 2 or 3. Additionally, there were no metal plating plants near the center of Santa Clarita, where the highly contaminated S-8, N, N-7, N-8, T-7, and Valley Center wells were located. Thus, we believe that domestic waste represents Factor 1. This is a more viable conclusion because all of these contaminated wells are on the Santa Clara River, and the Santa Clara River collects water from many different sources of water in Santa Clarita Valley.

Factor 2 was mostly comprised of PFHxS. This species is linked with aqueous film-forming foams (AFFFs), which are commonly used in firefighting settings (28, 29). Specifically, AFFFs are components of Class B fire extinguishers, used to put out fires on flammable liquids and gases (43). Previously, Class B fire extinguishers were present in both field and training settings. However, as of 1 January 2022, they have been prohibited from training, which takes place in fire stations (44). However, these trainings still have effects today because PFASs have a half-life of 40 years in water; thus, trainings before 2022, gas fires, or liquid fires were likely the source of the AFFFs in Santa Clarita (45). Some class B fire extinguishers contain PFOS as well: airports and military bases are the most common examples (28). Because Santa Clarita does not have either of these facilities, it is logical that Factor 2 in our study lacked PFOS. Factor 2 likely represents Class B fire extinguisher use.

Factor 3 was predominantly composed of PHFxA, PFPeA, PFHpA, PFBA, and PFBS. These short-chain perfluoroalkyl carboxylic acids (PFCAs) have been used for food packaging (26, 27, 42). Other studies found this same combination (short-chain PFCAs and PFBS) and associated it with food packaging and textile treatment, so we assigned this factor to food packaging and textile treatment (26).

We also investigated the spatial variation of PFAS concentrations and found a particularly high concentration of PFASs within the central region of Santa Clarita. This unusual variation seems to have been caused by a combination of geographic and human factors. First, as the Santa Clara River flows from the east to the west, it encounters a rise in elevation, causing the water to move upstream in the central region. This upstream movement slows the flow, allowing more time for PFASs to seep into the groundwater (46). Additionally, as the river narrows westward, it is placed in a chokehold where PFASs can accumulate. Compounding these factors is the higher population density in the central area, where more waste is predicted to be generated. PFASs are commonly found in various consumer products and industrial processes (31). As a result, the waste produced by the local population contributes significantly to the elevated PFAS levels in the central region. Additionally, Santa Clarita is a watershed, which means that all the water in Santa Clarita Valley, such as rain drainage in pedestrian streets, connects to the Santa Clara River. In other words, any kind of waste in the valley that is thrown onto the streets travels down to the river whenever it rains (47).

Our initial hypothesis that Landfill C would be the biggest PFAS contributor was not supported by the results. The wells

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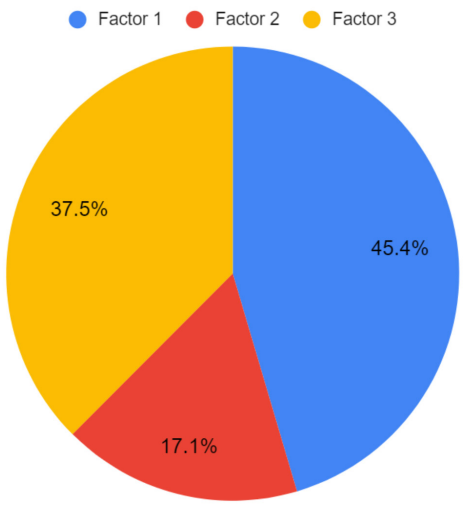

Figure 3. Each of three factor's contribution to the total PFASs in Santa Clarita groundwater. The numbers shown are the pt values, percentages of Santa Clarita's total PFASs that the factors contributed to.

near Landfill C (E17, E15, D, 207) were significantly lower in PFOS concentrations than the other wells, even though PFOS is the most common PFAS found in landfills (9). The central region wells contained the highest levels of PFOS, meaning Factor 1 most likely contributed to most of it, since Factor 1 was attributed to around 75% of all the PFOS in the wells (**Figure 2**). But since the river flows from east to west, and Landfill C is located in west Santa Clarita, it is highly unlikely that the landfill's leachate's chemicals traveled upstream to the central and eastern region, and instead, much more likely that the PFASs in the central and eastern region were sourced from the local domestic waste, AFFFs, food packaging and textile treatments. Hence, although Factor 1 was highest in PFOS, which is most prevalent in landfill leachate, it most likely did not represent Landfill C's influence. Instead, it likely represented non-landfill-sourced domestic waste, which could range from comparatively minor waste like pet waste to more hazardous pollutants like pesticides from irrigation runoffs (16). Since Factors 2 and 3 did not have considerable PFOS, it is even less likely that these factors represented Landfill C (24, 26, 31). Thus, Landfill C is not a major factor of PFASs based on the PMF results and is likely an insignificant source of PFASs in Santa Clarita.

Our findings, though, should be read with these limitations in mind: missing data or data points below MDL in the dataset, the relatively small size of the dataset, issues with the time of collection, and some unexplained phenomena. First, the dataset had data that was missing or below MDL. California's GeoTracker had an MDL of 2 ng/L, and 6% of the data was below this MDL. This is particularly problematic for PFHpA, which had 24.4% of data below MDL. 2.8% of the data points were missing; the issue was particularly problematic for PFPeA, which had 9.3% of its data points missing. Faulty data were treated based on practices from other similar research: missing data were replaced with the arithmetic mean of the corresponding species, and below MDL data were replaced with half the value of the detection limit (48, 49). Second, the data set was on the smaller side compared to other studies: we used 22 wells at a total of 107 data points for PMF, which was on the lower end for other studies using PMF for PFASs source apportionment, which used 29–812 wells (26-31). This was because, although there are 27 wells in the community, some were excluded due to the unavailability of data after 1 March 2023. Considering the spottiness and small size of our data, we suggest that future researchers collect their own data to minimize the impacts of these limitations. Third, there were two temporal issues with our data: our data spanned from March to May, which means that the spring season was weighted higher than other seasons. This is an issue because textile treatments are more prevalent in the winter months, when people wear more layers of clothing; this could have caused Factor 3 to be underrepresented in our study (5). Moreover, although the majority of the data was collected on a quarterly basis, some wells would occasionally have periods where they had more frequent or infrequent data collection intervals, which could have led us to overlook possible trends, such as the exact date at which L2A's PFASs concentrations began rapidly increasing between March 2023 and May 2024. Hence, we suggest that future research be carried out consistently throughout the year to represent the seasons equally. Fourth, we looked at a narrower breadth of PFASs than other studies. Although the public data covered 19 species of PFAS, our study was limited to 8 because data for 11 of the 19 PFAS species were missing or below MDL for at least 80% of the data. Similar studies analyzed an average of 14.2 and a range of 6 to 29 different PFAS species (26-31). The number of PFAS species we studied likely limited the scope of our source apportionment results. We believe this is because some sources of PFAS are highly associated with certain PFAS species that we were not able to include in our study: for example, 6:2Cl-PFESA and 8:2Cl-PFESA, which are highly associated with mist suppressants in chromium plating (26). Therefore, we believe future studies should look at more PFAS species. Lastly, a very high concentration of PFHxS, which our study associates with AFFFs, has been found at N13. A fire station is located 0.7 miles from this well; however, other wells are much closer to fire stations and have significantly lower PFHxS concentrations. This suggests that N13's high PFHxS concentration could not be due to the nearby fire station. N13 has had levels of PFHxS at around 30 ng/L since data collection began in August 2019; since fires are temporary events, the high levels of PFHxS at this location cannot be explained by fires either (19). Thus, we do not know what has caused this well to have elevated PFHxS concentrations. As a result, future researchers should try to have data collection go for as long as possible to see when changes started to happen, to make it easier to identify what causes changes in PFAS levels.

Despite these limitations, our study has significant implications for the Santa Clarita Valley community, California's state legislature, and the current understanding of PFAS and its sources. First, our study could help the residents of the Santa Clarita Valley because the area is not commonly the focus of studies, unlike cities like the neighboring Los Angeles. While at least five different water quality studies have been conducted for Los Angeles, there were none found for the Santa Clarita Valley, to our understanding (32-36). The findings of our study, particularly on the pathways of PFASs and the level of PFAS exposure within the community, will help inform local residents and the government about PFAS contamination and the actions they should take. Second, our

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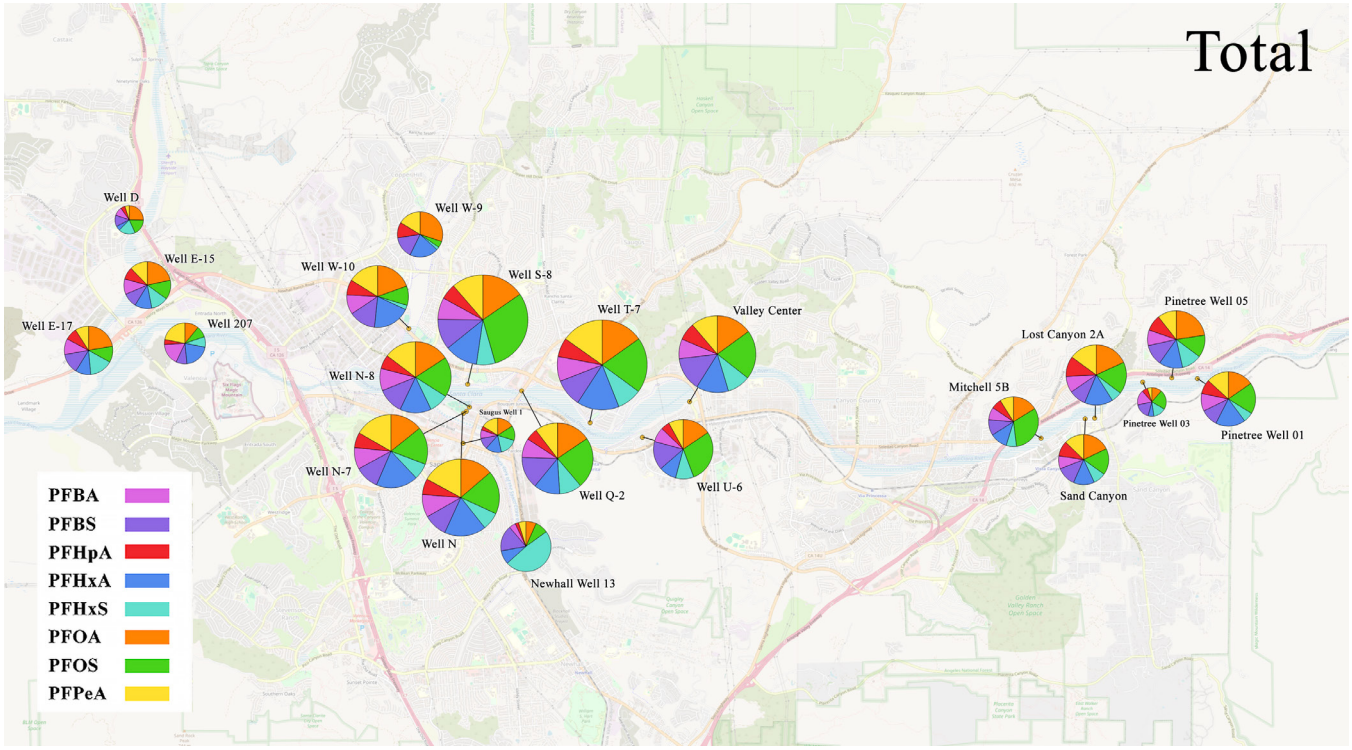

Figure 4. Locations of Santa Clarita wells with the concentrations of each PFAS species shown. In the map, we have attached pointer lines between the well locations (yellow dots) and their respective pie charts. Each color represents a specific PFAS species.

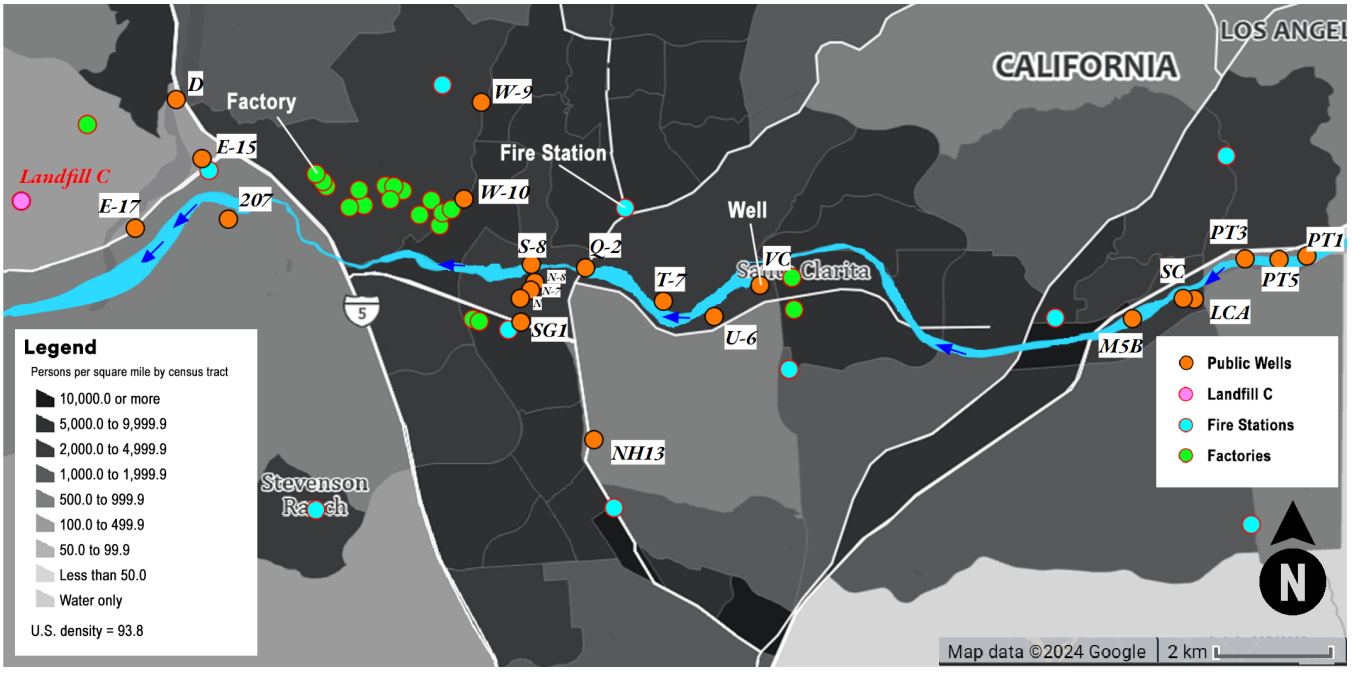

Figure 5. Locations of public wells used in the study, Landfill C, local fire stations, and factories. Landfill C is marked as a pink dot with a red outline and is located on the left side of the map. The wells are marked as orange dots with a black outline. The names of each well are shown on the map. The fire stations are marked as blue dots with a red outline. The factories are marked as green dots with a red outline. The silhouette of the Santa Clara River is highlighted in blue, with dark blue arrows indicating the direction of the flow. The grayscale tracts on the map indicate population density (55).

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study may aid California's lawmakers in creating laws that target the most prevalent source of PFAS. For example, due to the high contributions from non-landfill waste, lawmakers should raise the fines for littering and spread awareness of the fines to decrease the amount of trash ending up in the river. Lastly, our study provides more insight into PFASs and how PFAS sources interact with unique geographic features like the ones in Santa Clarita - the watershed and the river's connection to groundwater. Although these features were present in other studies on PFAS source apportionment — that of the Daliao River — they have not been investigated to the extent of our study, which showed that this geography had a significant impact on the PFAS sources of Santa Clarita (30). In our study, the watershed was a pathway for domestic waste, AFFFs, food packaging, and textile treatments entering the groundwater of Santa Clarita.

In conclusion, while our hypothesis of Landfill C contributing most to the PFASs in Santa Clarita was not supported by the results of this study, we identified domestic waste to be dominant as a contributor (45.4% of the total PFASs) to the PFASs in our community. Additionally, food packaging contributed 37.5% to the PFASs in Santa Clarita, and AFFFs contributed 17.1% to the PFASs in Santa Clarita. Our findings, therefore, raise awareness about PFASs for the community, support bills against these dominant sources of PFASs, and advance the knowledge of PFASs and their sources.

MATERIALS AND METHODS

Source of Data

PFAS concentrations within public wells (measured in ng/L) over the period from 1 March 2023 to 2 May 2024 were sourced from the California State Resources Control Board (19). From the original data set, which covered 27 wells and 19 PFAS species, information was retained for 8 species and 22 wells. The other five wells were excluded due to the unavailability of data after 1 March 2023. The excluded PFAS species had concentrations below the MDL for at least three-fourths of the wells. The eight PFASs considered in this study were PFOA, PFOS, PFHxS, PFHxA, PFBS, PFBA, PFHpA, and PFPeA. The final data set still contained missing data points or data points below the MDL, which were imputed based on the criteria described in the data pre-treatment section of the methods section to minimize negative impacts on the model accuracy. Additionally, some wells had more data points than others, although most of the data was collected every quarter (**Figure S1**).

Positive Matrix Factorization (PMF)

Positive Matrix Factorization (PMF) is a model that assists users in source apportionment through analyzing multiple variables. PMF organizes the inputted data (X) into matrices — the matrix of source contributions (G), the matrix of factor profiles (F), and the leftover matrix (E) (52). This model can be represented by equation 1:

$$x = (G \cdot F) + E \qquad [1]$$

where X is the matrix of the inputted data points, G is the matrix of source contributions, F is the matrix of factor profiles and E is the residual matrix (52).

The specific PMF model we used was the latest version of the EPA's PMF (44). For PMF, besides the original concentration values, uncertainties are also necessary. We derived uncertainties (u) using the following equations, adopted from existing studies with similar data sizes and/or similar experimental designs (26, 27, 53).

Equation 2 was used if the minimum detection limit exceeded the input concentration:

$$u = \frac{5MDL}{6} \qquad [2]$$

We used Equation 3 when the concentration exceeded the MDL:

$$u = \sqrt{(ErrorFraction \cdot Concentration)^2 + (0.5 \cdot MDL)^2} \qquad [3]$$

Error fraction is the assumed error on the concentration data, which can be calculated by using the relative standard deviations, and concentration is the reported concentration value.

To identify the error fraction, we found the relative standard deviations of the numbers that we put into PMF. Since our Relative Standard Deviation (RSD) was 30.9%, the error fraction was set to 0.2 (53).

Data Pre-treatment and Model Development

Missing data values were filled by the arithmetic mean of the corresponding species, and the uncertainty for that missing value was that arithmetic mean multiplied by a factor of four (48, 49). For values under 2 ng/L, which is the MDL, we input 1 ng/L for the concentration to PMF; a study on the Bohai Sea used the same method of inputting half the MDL as the concentration to avoid producing inaccurate results (27) (**Figure S1**). Of the 64 data points below MDL, 62 had an MDL of 2 ng/L, while 2 had an MDL of 1.8. Since so few did not have an MDL of 2 ng/L, we used 2 ng/L for all data points. After the base model was created through PMF, various parameter tests and analyses were run to ensure the model's stability and accuracy (**Appendix**).

Signal to Noise Ratios

S/N ratio was automatically calculated in the EPA PMF program with the formula:

$$\frac{S}{N} = \sqrt{\sum s_i^2 / \sum n_i^2} \qquad [4]$$

where S/N is Signal to Noise ratio, s_i is signal, and ni is noise of a PFAS species (54). The percentage of data below MDL was calculated by dividing the number of data points below MDL by the number of normal values. The percentage of missing data was calculated by dividing the number of missing values by the number of normal values. Data with S/N ratios between 0.2 and 2 had their uncertainties increased fourfold to downsize their impacts on the model (26).

Data Post-treatment

After the model was finalized, we calculated the percentages of how much each of the three factors contributed to the total PFASs (**Equation 5**). We used the equation:

$$p_t = \frac{\sum(p_s \cdot c_{st})}{\sum c_{st}} \qquad [5]$$

where pt is the percentage of total PFASs that the factor contributed, ps is the percentage of one of the PFAS species that the factor contributed to (**Figure 2**), cst is the total concentration of that PFAS species,

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adding the concentrations of all wells together.

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Appendix

Species (ng/L)	S/N	Minimum (ng/L)	Median (ng/L)	Maximum (ng/L)	Mean (ng/L)	S.D. (ng/L)	Number of missing data	Number of data below MDL	Normal Values	Percent of data below detection (%)	Percent of missing values (%)
PFBA	2.56	1	7.3	18	7.7	4.3	5	5	97	4.7	4.7
PFBS	3.04	1	8.8	25	10.3	6.2	0	5	102	4.7	0
PFHpA	1.98	0.9	6.1	16	5.9	3.9	9	21	86	19.6	8.4
PFHxA	3.02	1	8.8	35	11.9	8.6	0	6	101	5.6	0
PFHxS	2.78	1	7.4	37	9.2	7.2	0	7	100	6.5	0
PFOA	3.19	1	13	36	13.4	8.2	0	6	101	5.6	0
PFOS	3.17	1	12	82	15.6	13.4	0	1	106	0.9	0
PFPeA	2.52	0.9	9.2	29	10.7	7.7	10	11	86	10.3	9.3

Table S1. Overall statistics of raw data. Basic statistical metrics for each PFAS species such minimum, median, maximum, mean, and standard deviation (S.D.) are shown. Additional extra details to assess the quality of the raw data is provided such as the signal-to-noise ratio (S/N), number of missing data, number of data below MDL, normal values, percent of data below minimum detection limit (MDL), and percent of missing values are included. The number of missing data is included inside of the number of data below MDL.

	Factor 1	Factor 2	Factor 3	Factor 4	Factor 5	Factor 6	Unmapped
Boot Factor 1	100	0	0	0	0	0	0
Boot Factor 2	2	67	6	5	15	5	0
Boot Factor 3	0	2	96	0	2	0	0
Boot Factor 4	1	0	4	88	7	0	0
Boot Factor 5	0	0	0	0	100	0	0
Boot Factor 6	2	2	8	8	9	71	0

	Factor 1	Factor 2	Factor 3	Factor 4	Factor 5	Unmapped
Boot Factor 1	90	4	0	2	4	0
Boot Factor 2	2	92	0	1	5	0
Boot Factor 3	1	1	98	0	0	0
Boot Factor 4	6	7	1	70	16	0
Boot Factor 5	0	0	0	0	100	0

	Factor 1	Factor 2	Factor 3	Factor 4	Unmapped
Boot Factor 1	100	0	0	0	0
Boot Factor 2	0	100	0	0	0
Boot Factor 3	19	6	75	0	0
Boot Factor 4	0	2	1	97	0

	Factor 1	Factor 2	Factor 3	Unmapped
Boot Factor 1	96	1	3	0
Boot Factor 2	0	100	0	0
Boot Factor 3	0	0	100	0

	Factor 1	Factor 2	Unmapped
Boot Factor 1	68	32	0
Boot Factor 2	0	100	0

Table S2. Bootstrapping data for 2-6 factors. Results from bootstrapping conducted with 100 runs and a minimum R-correlation of 0.6 (n=5). We looked for over 80% mapping of each factor onto itself. Bootstrap analysis was used to find the optimal factor solution. Bootstrap analysis is a diagnostic tool that helps to identify if a small set of data in the original model is disproportionately influencing the solution (53). In Bootstrapping, a set of samples with replacement are randomly selected first. Then, PMF is executed on this bootstrapped sample with estimated factor profiles as well. Afterwards, hundreds of bootstrapped data sets are analyzed to determine the precision of PMF profiles, shown in terms of percentage mapped (48). Based on these results, the best number of factors are trimmed down. We conducted the Bootstrap Analysis for our model for 2 through 6 numbers of factors. For our model, the best solution came down to 3 factors because it was the only one with all factors over 80% mapping (53). This set up a framework for our source apportionment by providing us a clear number of factors to look for. However, we will need to further test this number of factors through displacement analysis to confirm whether or not this solution worked.

Strength	dQ (Robust)	Q (Robust)	%dQ (Robust)	Q (Aux)	Q (True)	Converged
-0.5	38.6	400.3	9.64	32.8	367.6	Yes
-0.3	**15.2**	**376.9**	**4.03**	**12.5**	**364.4**	**Yes**
0.3	44.4	406.2	10.94	43.2	363.0	Yes
0.5	133.1	494.9	26.90	129.5	365.4	Yes
-1.0	136.6	498.4	27.42	110.1	388.3	Yes

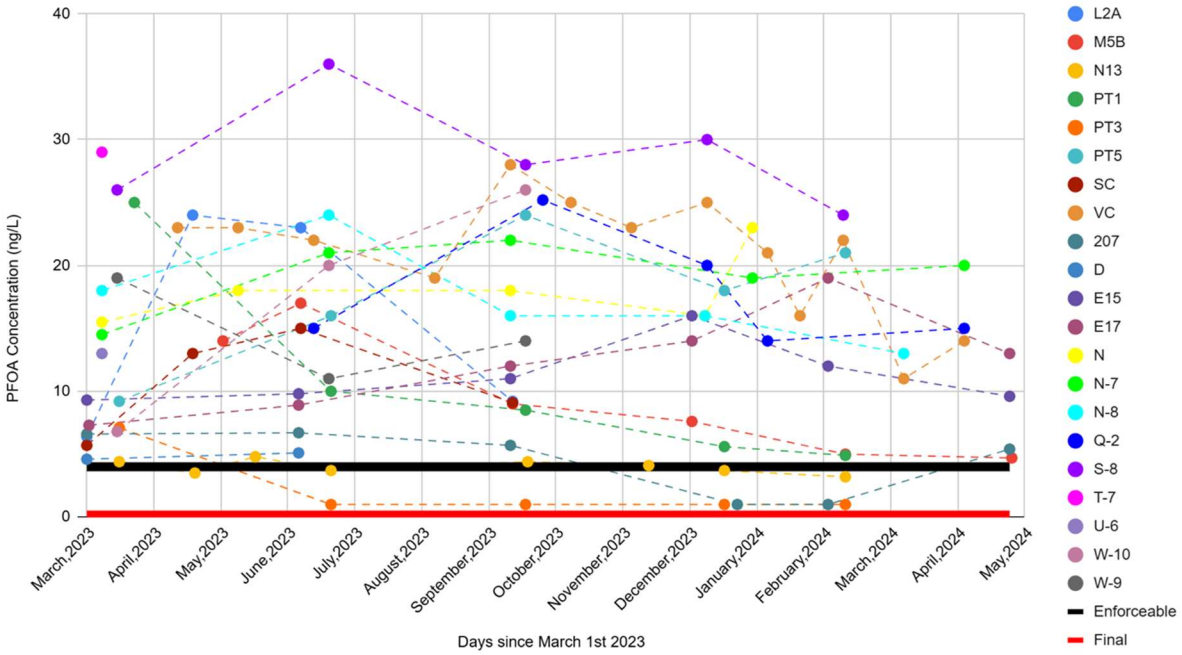
Table S3. Fpeak model run summary. The results from the Fpeak run using -.5, -.3, .3, .5, and -1 strengths for Fpeak rotations are shown. To find the best Fpeak rotation to use, we had to put values until we found the value with the highest Q(robust) while converging and having less than a 5% dQ(robust) (53). The Fpeak parameter was tuned to make trends in the model more visible. In matrix factorization, rotational tools like Fpeak are a helpful tool in finding clearer trends within the data matrix by viewing it in different "orientations." Specifically, Fpeak is a rotational method that forces the rows and columns of the 2 matrices derived through PMF (F and G) to be added and/or subtracted from each other (48). We tested the model's stability with 5 different Fpeak parameters: -1.0, -0.5, -0.3, 0.3, 0.5. For our model, the highest Q(robust), while being under 5% %dQ(robust), was at an Fpeak strength of -.3. Hence, for our final model, an Fpeak of -0.3 was used (**Figure 1**). This helped our study by increasing the accuracy of the model - an increase in Q value is an increase in accuracy of the model.

		Factor Swaps	
dQmax=4	0	0	0
dQmax=8	0	0	0
dQmax=15	0	0	0
dQmax=25	*0*	*0*	*0*

Table S4. Displacement data for 3-factor solution. A table of the amount of times factors swapped are shown. Displacement is an analysis method that is used to tune the selected solution to higher precision by testing its sensitivity to small influences. Utilizing DISP analysis helps identify weak factors. The error was 0, and the largest drop in Q is -.028. Since the error was 0, the largest drop in Q was small and no factors swapped, the 3-factor solution was chosen as the best number of factors (53). This method was the final step in verifying our model after all the tuning was complete. It confirmed to us that our model was stable enough to trust.

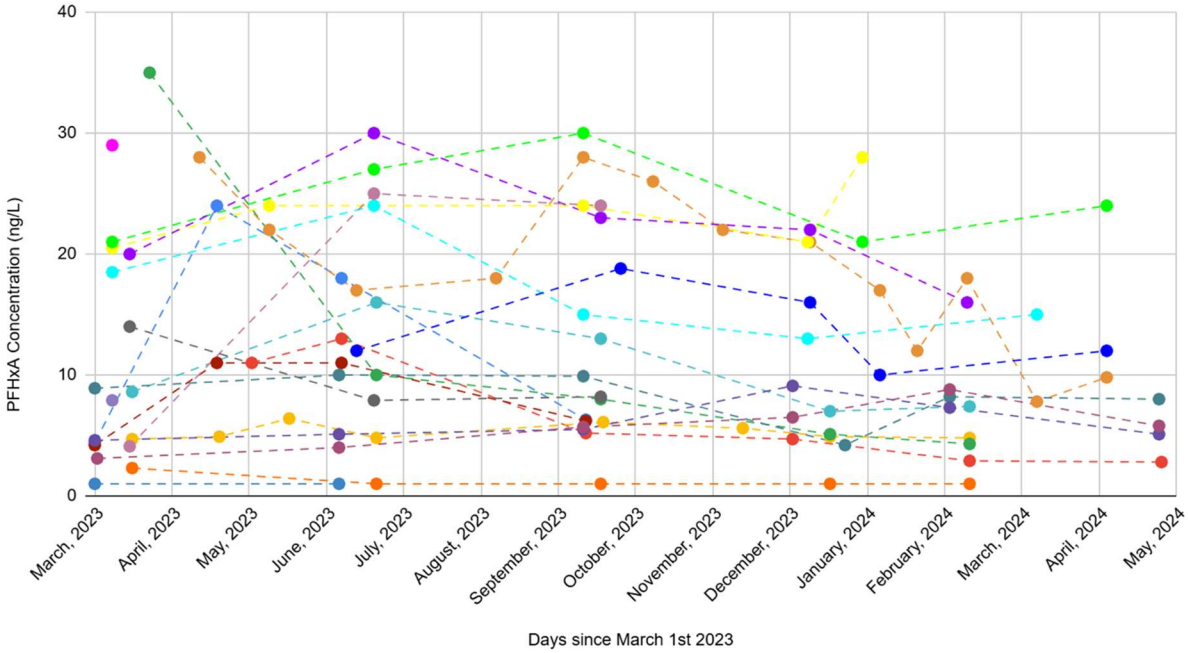
PFOA vs Time

a



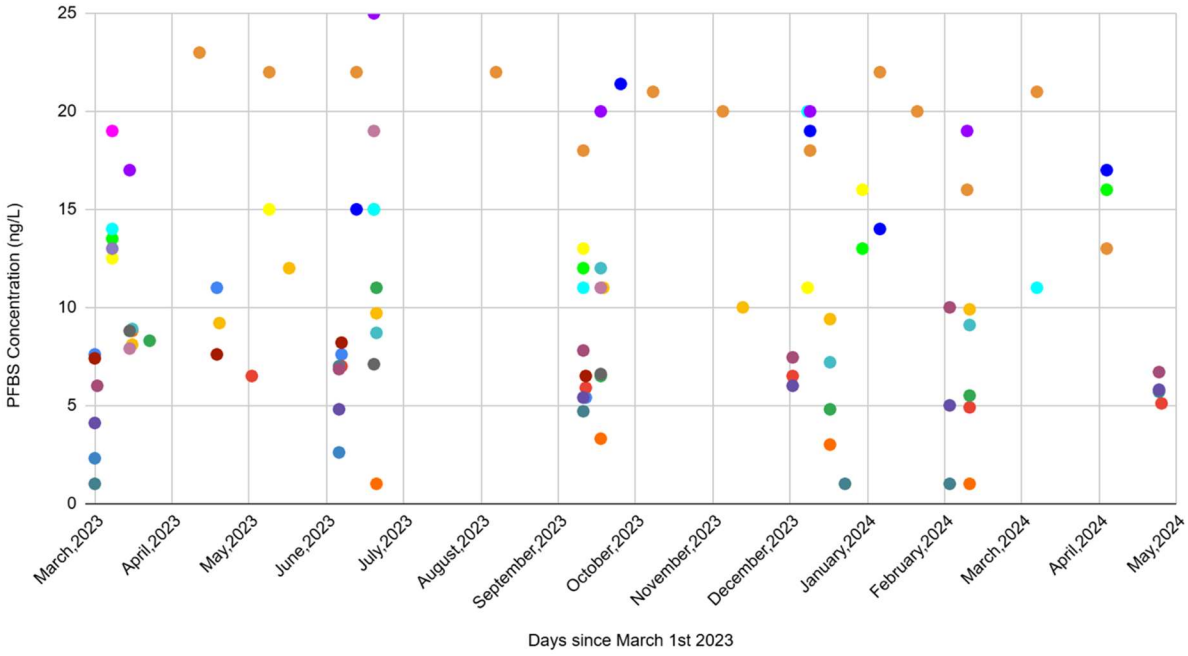
PFHxA vs Time

b



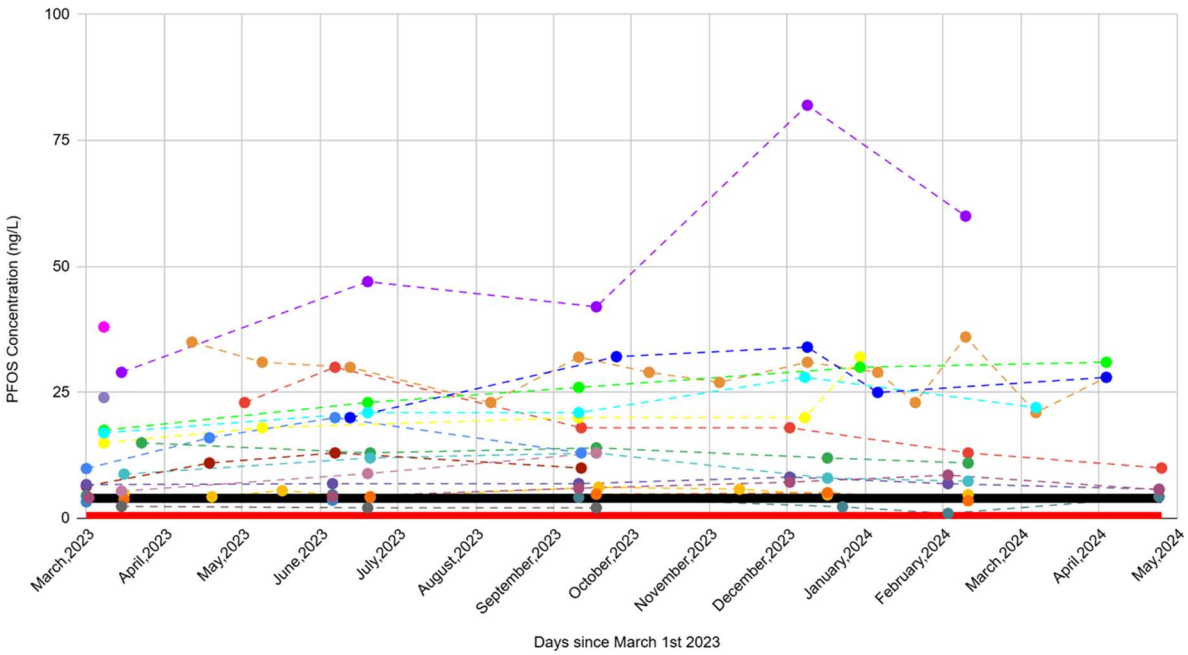
PFBS vs Time

c



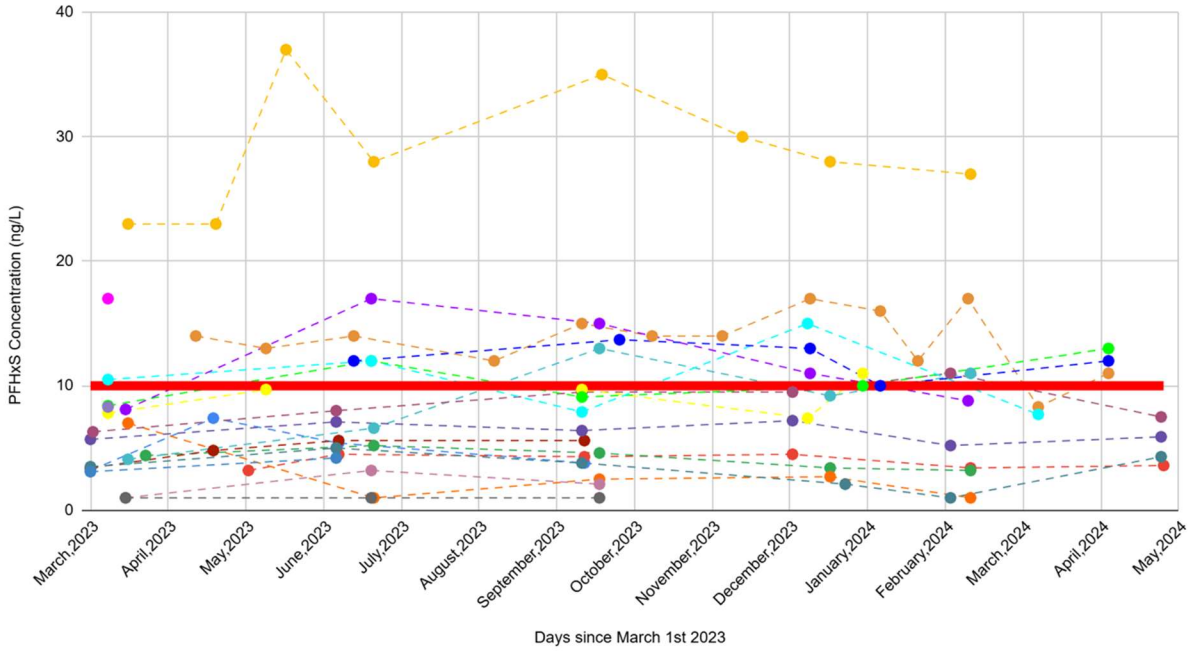
PFOS vs Time

d



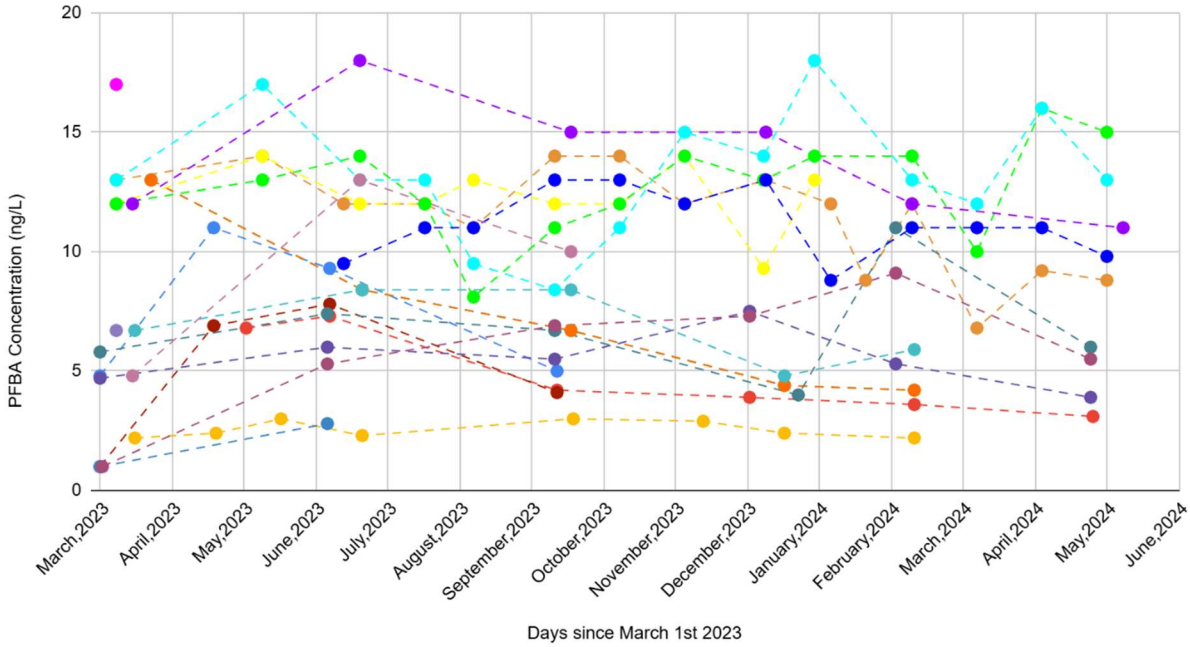
PFHxS vs Time

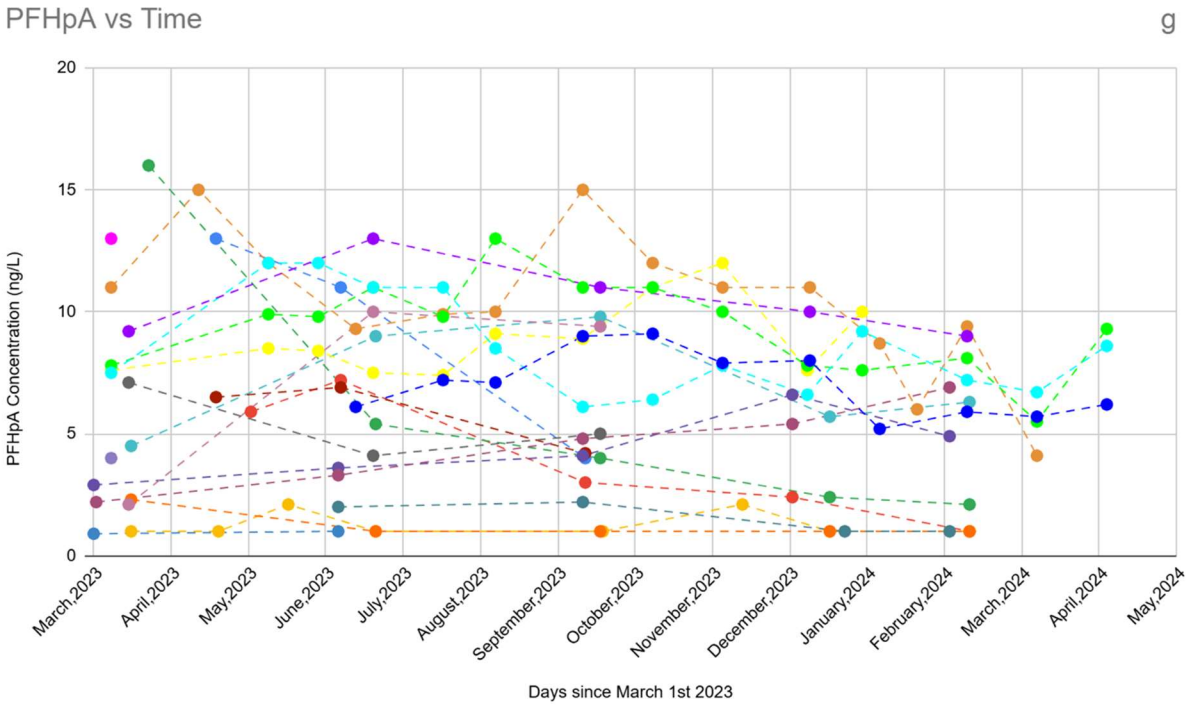
e



PFBA vs Time

f





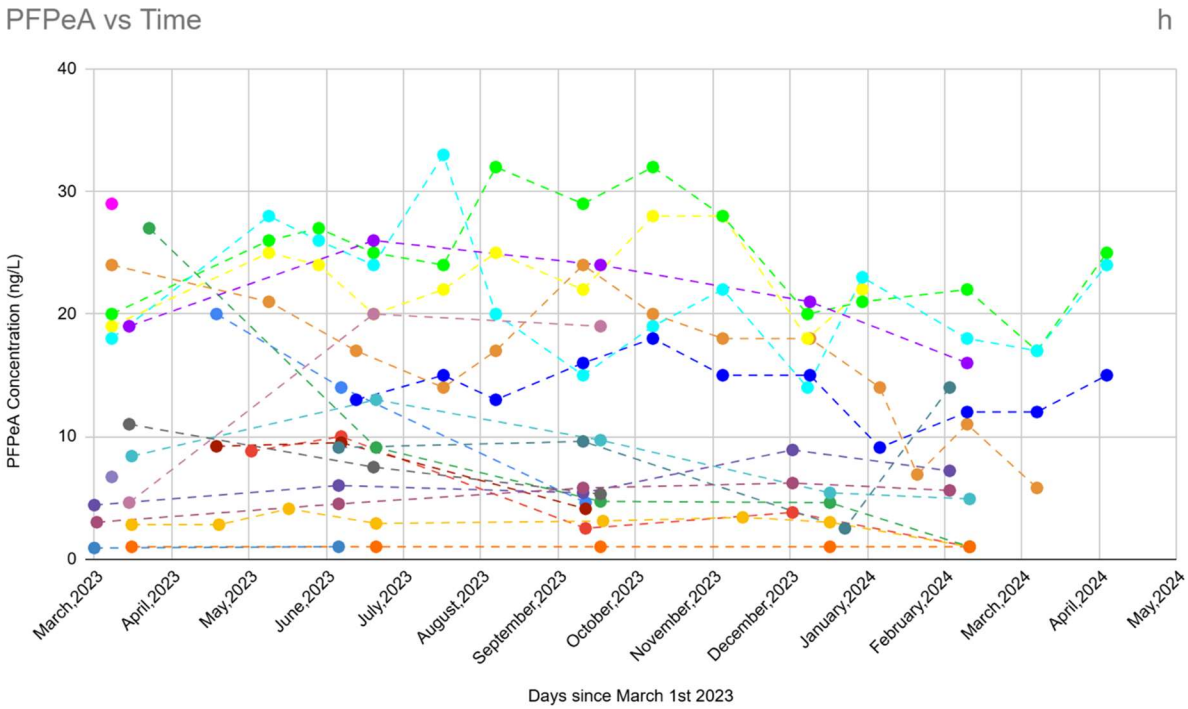


Figure S1. Modified data from each well over time, measured in months from March 2023 to May 2024. From (a) to (h) are different PFAS species shown. All wells share the legend shown on (a). The horizontal black line represents the EPA's enforceable goal concentration, and the red line represents their final concentration. The dotted line between data points should not be used to infer concentration between measurements, but instead to aid in identifying which well was which. Not every well had data on every species.