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PFAS in California Groundwater: Distribution, Facility Sources, and Environmental Factors Correlations

THESIS

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ABSTRACT OF THE THESIS

PFAS in California Groundwater: Distribution, Facility Sources, and Environmental Factors Correlations

by

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Master of Science

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Professor Christopher Olivares Martinez, Chair

Poly- and perfluoroalkyl substances (PFAS) have been widely used since the 1940s. They are ubiquitous and have posed a threat to human health. Because of that, researchers have shown high interest in PFAS contamination in the environment and are trying to find effective ways for PFAS removal, primarily focusing on specific compounds like perfluorooctanoic acid (PFOA) and perfluorooctanesulfonic acid (PFOS). The limited understanding of PFAS presence in groundwater, along with the diverse subclasses of PFAS and their occurrence after the discharge from various industrial facilities, has significantly challenged our ability to assess risks. These complexities hinder the thorough evaluation of PFAS subclasses' impact on groundwater quality after the related facilities discharge. 38 PFAS in California groundwater from 2016 to 2022 were analyzed in this study. Statistical and spatial analyses were performed to visualize the distribution of groundwater PFAS in California and examine the relationship between PFAS and the facilities and environmental factors such as weather, soil, and air quality. I found that airports in California contributed significantly to PFAS groundwater contamination and then came bulk fuel terminal/ refinery and cleanup program sites. In addition, I identified the three highest

concentration PFAS of each industrial facility. These analyses demonstrate the Greater Los Angeles Area is a hotspot for PFAS-contaminated groundwater. PFOS is the most frequently occurring PFAS in all the facility types. Also, this research summarizes all the environmental factors associated with PFAS and the top three highest concentrations of PFAS in all the facilities. I compared the results of this study with those of other researchers regarding PFAS in groundwater at airports and landfills, which exhibit similar trends. The recognition of PFAS subclasses provides a new understanding of PFAS contamination in California caused by industry. It aims to offer targeted approaches to the removal of specific PFAS for future study.

1. INTRODUCTION

Poly- and perfluoroalkyl substances (PFAS) are a class of more than 4,000 highly fluorinated aliphatic compounds manufactured for numerous industrial and consumer applications[1]. Due to the strength of the covalent carbon-fluorine (C-F) bonds, PFAS has the property of high chemical stability and also are both hydrophobic and oleophobic[2], they are also recognized as being extremely recalcitrant[3,4], which means it will take a long time for PFAS to break down and degrade. As a result, PFAS are also known as "Forever chemicals"[5,6]. From 2000-2020, PFAS was found in many countries, and the top 10 of them that have considerable concern about PFAS are the US, China, Canada, Sweden, Germany, Norway, Spain, Denmark, Japan, and the United Kingdom[7].

In light of the fact the C-F bond provides PFAS with unique physicochemical properties such as resistance to greasy, stains, water, and heat, PFAS have been widely used since the 1940s in several commercial applications, such as aqueous film-forming foam (AFFF), food packaging, waterproof clothes and furniture, personal care products, cosmetics, carpet, paper industry, and textiles field[8–10]. The historical trajectory of PFAS manufacturing and emissions spans a considerable expanse of time. As a result, PFAS is widely spread in groundwater, drinking water, surface water, air, and soil[6,11–22]. Prior studies show PFAS were detected in PM2.5 samples[23,24]. Studies have found PFAS occurrence in surface water, groundwater, and surface water near airport fighting training areas, metal industries, chrome plating industries, landfills, wastewater treatment plants, textile facilities, and fluoropolymer manufacturing[8,23,25–29]. According to Clara et al.[8], the metal industry, such as galvanizing, has the highest emission of PFOS, and the paper industry has the highest emission of PFOA[25]. PFAS will be discharged into groundwater and surface water and then cause contamination after the industrial wastewater discharge[30,31].

According to previous studies, PFAS will accumulate in the environment and then harm human health for a prolonged period, which is of great concern. Additionally, PFAS are associated with increases in blood cholesterol and blood pressure, high blood uric acid concentration, chronic kidney disease, kidney cancer, a negative correlation with infant weight, influence neuro-development, and reduction of the effectiveness of some vaccines such as rubella[32–37]. Besides, PFAS is bioaccumulative and toxic in mammals[8]. Furthermore, studies have shown that PFAS are found almost everywhere that humans can easily directly contact PFAS from food, soil, PFAS-treated furniture, drinking water, and air. PFAS will accumulate in the human body[38–43]. Some research also points out that PFAS have been found in human and animal plasma[44,45].

Nowadays, manufacturers use short-chain PFAS instead of long-chain PFAS. Short-chain PFAS are defined as perfluoroalkyl carboxylic acids (PFCA) with less than six carbons and perfluorosulfonic acid (PFSA) with less than five carbons[34]. Also, some facilities stopped the use of long-chain PFAS. For example, 3M will exit all PFAS manufacturing by the end of 2025[34,46]. Since the 2000s, short-chain PFAS has been used as a substitute for long-chain PFAS in a large number of manufacturing industries due to their shorter half-life in the human body, which will undermine the harm of PFAS to the environment and humans. As a result, some presume short-chain PFAS to be less toxic than long-chain PFAS[36,47,48]. Some studies show short-chain PFAS are more persistent in groundwater and soil than long-chain PFAS[49,50]and short-chain PFAS have a high mobility, which makes it easy to transport to the groundwater and eventually contaminate the groundwater [51,52]. PFOA and PFOS are the most studied long-chain PFAS; perfluorobutanoic acid (PFBSA) and perfluorobutane sulfonamide (PFBSA) are the most common short-chain PFAS widely detected in drinking water[53]. Also, different properties like water repellent are

shown by different lengths of PFAS. Taken together, these results suggest that research has mainly focused on the common PFAS, such as PFOA and PFOS in drinking water. Although groundwater, as some drinking water source, has shown high concentrations of PFAS in China[54]. Furthermore, PFAS in surface water, soil, and atmospheric deposition are potential sources of contamination in groundwater[55].

According to the research on California serum PFAS levels[56], contamination in the human body can be seen as a pervasive phenomenon in California. Also, although the usage of PFAS is being limited, some PFAS subclass contamination remained at the same level for decades, one of the reasons is the long half-life of the PFAS[56]. So, the issue that PFAS is commonly present in humans must be resolved. PFAS contaminates groundwater quality from not only the discharge of wastewater from facilities but also surface water and soil[55,57,58]. More importantly, more than 50% of the global population relies on groundwater[59], so PFAS contamination in California would easily influence drinking water and people's health. Though it is an incontrovertible fact of the contamination by PFAS, the research on PFAS in California groundwater is limited.

Furthermore, the relationship between PFAS and air, soil, and weather factors has been sparsely studied. To eliminate the health risks that would cause groundwater to run into drinking water and eventually influence the human body, it is necessary to find out the PFAS transport and distribution after discharge by the facilities into groundwater. The objective of this thesis is to find out the most influenceable facility to groundwater PFAS contamination, the more accurate recognition of the PFAS contribution of each facility, mainly focus on the PFAS and PFAS subclasses distribution in groundwater related to according to industry facilities and discover the correlations between PFAS and other environmental factors in California, identify

which kind of facility has the highest risk to groundwater quality, to informing further clear PFAS risk assessment of each type of facility and the groundwater contamination and better help the policymaking.

The study investigates the distribution of PFAS in California groundwater, and evaluates the relationship between PFAS and other environmental variables, considering the increasing health issues posed by PFAS contaminants, this study applied different analysis methods to provide a clear introduction of PFAS in California groundwater.

2. Methods

The dataset used in this study is based on the study by Dong et al.[60], which was originally published in additional material to their paper and stored in Excel format. The dataset was imported into Excel and used for further studies. This dataset was chosen because it provided detailed information about California's PFAS-related and environmental-related data from 2016 to 2022, I used Excel to filter relevant variables, such as the sample site information, PFAS tested concentrations, facility information, and the environment-related data were kept in the new sheet used in this study. From the research by Dong et al., the PFAS sampled location and concentration data were obtained from the Groundwater Ambient Monitoring and Assessment (GAMA), the Water Boards' data GeoTracker portals (GeoTracker), and the Environmental Working Group (EWG). The datasets for the facilities-related data collection were from EPA PFAS Analytic Tools, Toxics Release Inventory (TRI) Program Basic Data, EPA Detailed Facility Report, and Environmental Working Group (EWG). The National Cooperative Soil Survey (USDA) Soil Characterization Database was collected from the dataset for soil data. The air quality and weather data were from the National Centers for Environmental Information (NOAA), Purple Air, and EPA Outdoor Air Quality Data. The samples' locations are shown in Figure 2.1. All the units of the above data are in Table 2.1.

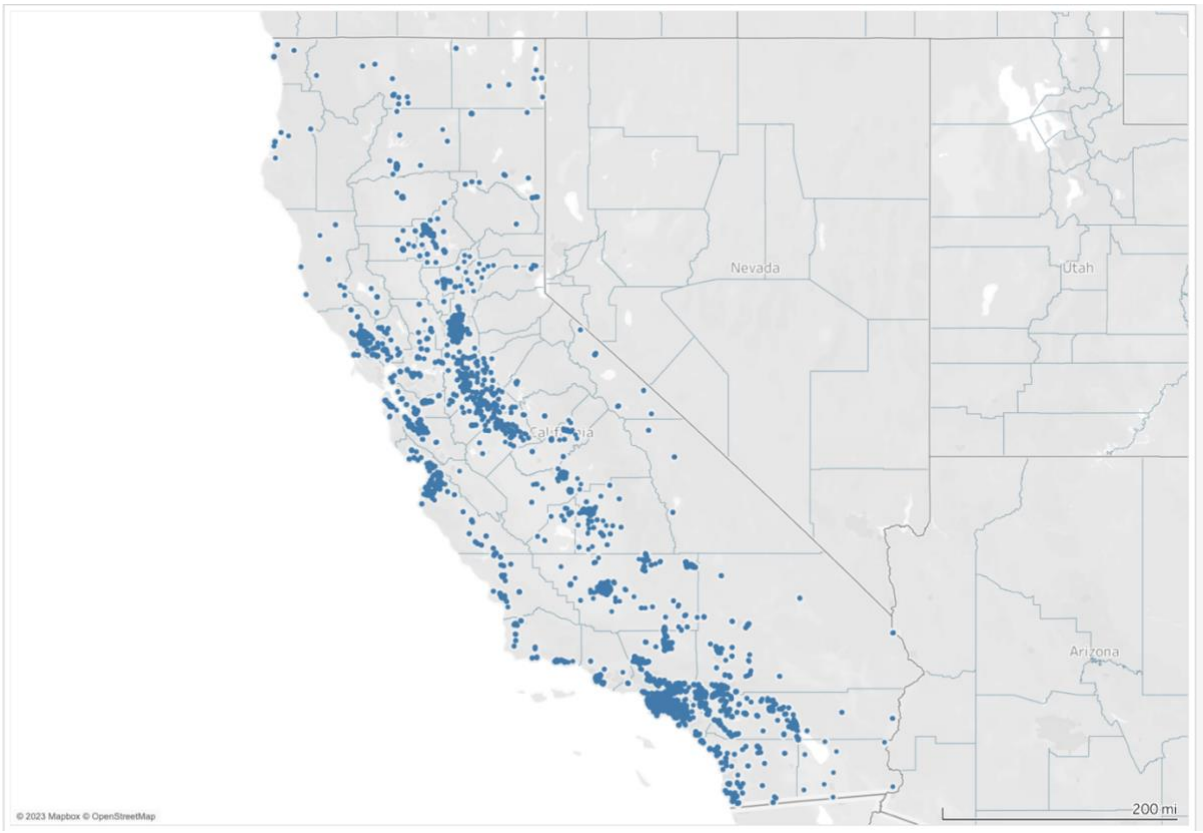

Figure 2.1. The sampling sites in California of the datasets used in this study

Table 2.1. The analytes and the full name of the analytes

Analyte	Name
PFBTA	Perfluorobutanoic acid
PFPA	Perfluoropropionic acid
PFHA	Perfluorohexanoic acid
PFHPA	Perfluoroheptanoic acid
PFOA	Perfluorooctanoic acid
PFNA	Perfluorononanoic acid
PFNDCA	Perfluorodecanoic acid
PFUNDCA	Perfluoroundecanoic acid
PFDOA	Perfluorododecanoic acid

PFTRIDA	Perfluorotridecanoic acid
PFTEDA	Perfluorotetradecanoic acid
PFHXDA	Perfluorohexadecanoic acid
PFODA	Perfluorooctadecanoic acid
3:3FTCA	3:3 fluorotelomer carboxylic acid
5:3FTCA	5:3 fluorotelomer carboxylic acid
7:3FTCA	7:3 fluorotelomer carboxylic acid
4:2FTS	4:2 fluorotelomer sulfonic acid
6:2FTS	6:2 fluorotelomer sulfonic acid
8:2FTS	8:2 fluorotelomer sulfonic acid
10:2FTS	10:2 fluorotelomer sulfonic acid
PFBSA	Perfluorobutane sulfonamide
PFPES	Perfluoropentane sulfonic acid
PFHXSA	Perfluorohexane sulfonic acid
PFHPSA	Perfluoroheptane sulfonic acid
PFOS	Perfluorooctanesulfonic acid
PFNS	Perfluorononane sulfonic acid
PFDSA	Perfluorodecane sulfonic acid
PFOSA	Perfluorooctanesulfonamide
ETFOSE	N-Ethyl Perfluorooctane sulfonamide ethanol
ETFOSA	N-Ehtyl Perfluorooctane sulfonamide
NETFOSAA	N-Ethyl Perfluorooctane sulfonamidoacetic acid
MEFOSE	N-Methyl Perfluorooctane sulfonamide ethanol
MEFOSA	N-Methyl Perfluorooctane sulfonamide
NMEFOSAA	N-Methyl Perfluorooctane sulfonamidoacetic acid

ADONA	4,8-Dioxa-3H-Perfluorononanoic acid
HFPA-DA	Hexafluoropropylene Oxide Dimer Acid
11ClPF3OUDS	11-Chloroeicosafluoro-3- oxaundecane-1-sulfonic acid
9ClPF3ONS	9-ChlorohexB2:B39adecafluoro-3- oxanonane-1-sulfonic acid
PFAS_total	The sum of 38 PFAS concentration
PFOA+PFOS	The total concentration of PFOA and PFOS
Short-chain PFCA	PFBTA, PFPA, PFHA, PFHPA
Long-chain PFCA	PFOA, PFNA, PFNDCA, PFUNDCA, PFDOA, PFTRIDA, PFTEDA, PFHXDA, PFODA
Short-chain PFSA	PFBSA, PFPES
Long-chain PFSA	PFHXSA, PFHPSA, PFOS, PFNS, PFDSA, PFOSA
Fluorotelomer carboxylic acids	3:3FTCA, 5:3FTCA, 7:3FTCA
Fluorotelomer sulfonic acids	4:2FTS, 6:2FTS, 8:2FTS,10:2FTS
Perfluoroalkane sulfonamido substances	ETFOSE, ETFOSA, NETFOSAA, MEFOSE, MEFOSA, NMEFOSAA

The dataset I used collected included 38 PFAS species, 14 facility types, and 26907 data in total, which have data from April 2016 to June 2022. The 38 PFAS analyzed are shown in Table 2.1. The 14 facility types were MSW Landfill, Cleanup Program Sites, Wastewater Treatment Plants, Airport, Chrome Plating, Bulk Fuel Terminal/Refinery, Fluoropolymer Manufacturers, Military Cleanup Site, Chemicals, Other Landfill, No Facility Within 50 km, Semiconductors and Related Devices, Other and Industrial. To ensure the facilities, soil, air, and weather condition data correspond to the PFAS data, I used Python to extract the PFAS-related data into a whole sheet and deleted the unrelated data; ArcGIS and Excel were applied to merge the data related to the study purpose into an entire excel sheet. According to all PFAS subclass

concentrations, I used the Excel function SUM to calculate the PFAS species groups like PFCA and PFSA concentration.

To reveal the data distribution and trend. I use Excel and SPSS for descriptive analysis. As for the mean and standard division bar chart, I applied Excel 2016 pivot table to calculate the mean of total PFAS and each PFAS subclass, and formula STDEV to calculate the standard deviation and then to create the bar chart of every facility and PFAS the mean and standard deviation chart. To illustrate the relationship between the amount of facility and PFAS concentration, I applied SPSS to create the scatter plot of the two variables. To compare the distribution of PFAS across multiple facilities, Python, Excel pivot tables, and add-ins solver were employed in the matter of diagnostic analysis and exploratory data analysis. SPSS, seaborne, matplotlib, and Pandas were applied to calculate the Pearson and Spearman correlation coefficient, and also depict the heatmaps. The average of every PFAS was generated by a pivot table used to make the stacked bar chart. Regarding the spatial analysis of PFAS, air, and weather data, Tableau was utilized to demonstrate the distribution and connection better. In the maps created by Tableau, each circle's area symbolized the concentration or magnitude.

3. Results and discussion

3.1Multivariate analysis of California groundwater PFAS concentrations

3.1.1 Temporal variation of PFAS concentration in California groundwater

Figure 3.1 Fluctuations in total PFAS yearly concentrations of four sites. The vertical axis represents the total PFAS concentration, and the horizontal axis shows the sampling dates; the unit of the PFAS concentration is part per trillion (ppt). The PFAS subclasses amount tested, and species considered are the same within the four sites.

PFAS was widely detected in California groundwater; two sites have continuous PFAS concentration records from 2016 to 2022, and two sites have records from 2017 to 2022. The line charts show the PFAS concentration trends during the seven (or six) years. All four sites are sampled in drinking water wells. Site 1 (latitude: 33.3060, longitude: -117.3474) and site 2 (latitude: 33.3081, longitude: -117.4516) are both near a wastewater treatment plant located in San Diego County. Site 3 (latitude: 33.8635, longitude: -117.8253) is near an MSW landfill in Orange County, and Site 4 (latitude: 33.5694, longitude: -121.3091) is near a cleanup program site. The PFAS concentration in Site 1 and 2 increased from 2016 to 2017, and the concentration

in 2018 for these two sites was the lowest; after that, the total PFAS concentration began to rise again. For Site 3, the concentration of total PFAS increased from 2017 to 2018 and then fluctuated back and forth between 80 to 160 ppt. The overall trend for Site 4 is downward, with the inevitable fluctuations within the annual range. I am exploring the nexus of PFAS with various facilities and PFAS intrinsic association. Cause the concentration change with the time between Site 1 and Site 2 is the same, while the trend of the PFAS concentration of Site 3 and Site 4 is different from all the other sites, the trend of the total PFAS change is relevant to the facility types nearby.

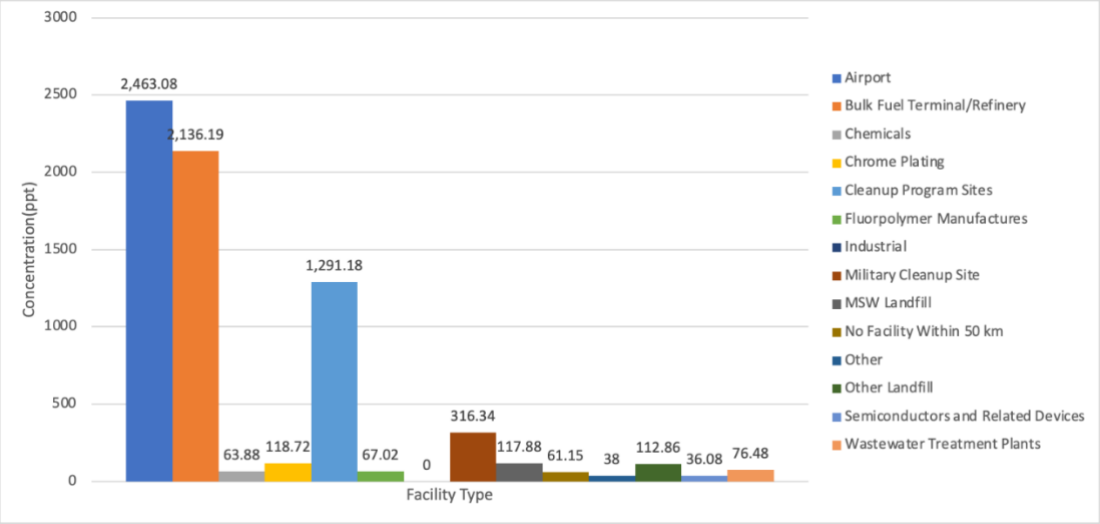

Figure 3.2. Total groundwater PFAS concentrations in 14 facilities nearby

Figure 3.2 summarizes the total PFAS contamination from all facility types. The total number of facility types is 14; one of them is no facility within 50km. From the graph, it is evident that the concentration of total PFAS in the airport is the highest, which is 2463.08 ppt. In descending order of concentration, the sequence is bulk fuel terminal/refinery, cleanup program sites, military cleanup site, chrome plating, MSW landfill, other landfill, wastewater treatment plants, fluoropolymer manufacturers, chemicals, no facility within 50 km, other, semiconductors

and related devices, industrial. Besides, at the sites near industrial facilities, the datasets show PFAS was not detected, so the concentration shown on the plot is zero. With the intention of discovering the contribution and comparing the PFAS subclasses within all the facility types, stacked bar charts and 100% stacked bar charts were applied in the study. The concentration used to plot the stacked bar is the average of every PFAS subclass.

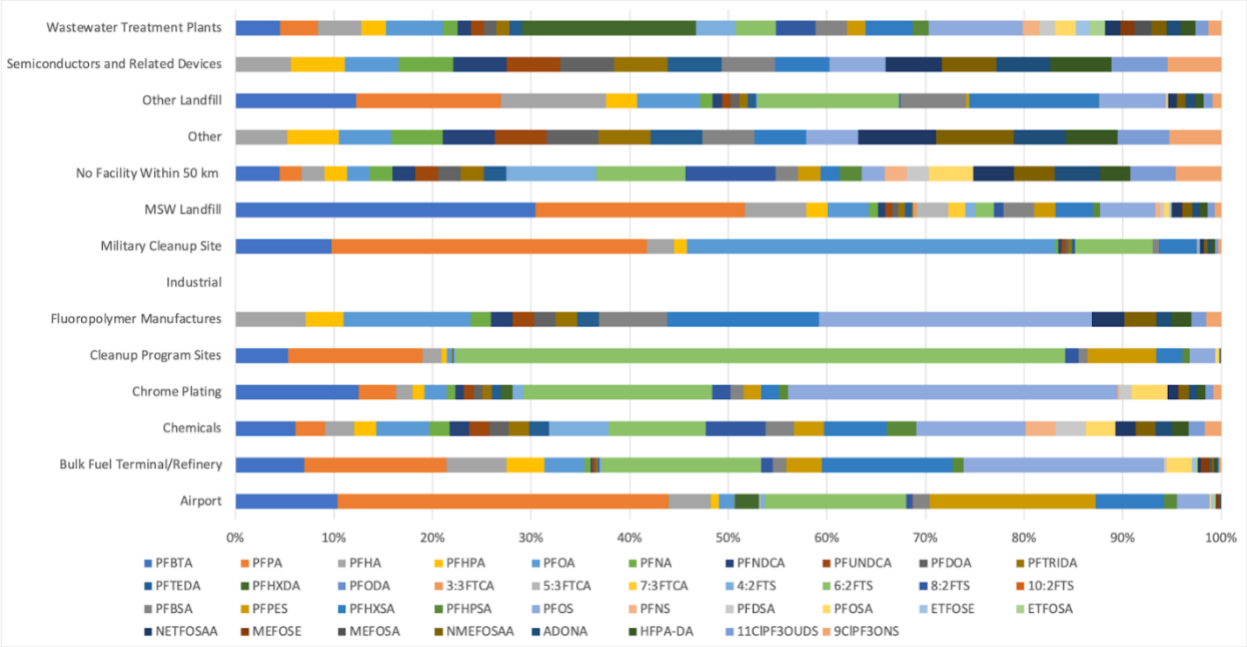

Figure 3.3. The PFAS profile (% contribution) in each facility, different colors shows different facility types

As Figure 3.3 shows, no matter which kind of facility it is, perfluorohexanoic acid (PFHA), perfluoroheptanoic acid (PFHPA), PFOA, perfluorononanoic acid (PFNA), perfluorodecanoic acid (PFNDCA), perfluoroundecanoic acid (PFUNDCA), perfluorododecanoic acid (PFDOA), perfluorotridecanoic acid (PFTRIDA), perfluorotetradecanoic acid (PFTEDA), perfluorobutane sulfonamide (PFBSA), perfluorohexane sulfonic acid (PFHXSA), PFOS, N-Ethyl perfluorooctane sulfonamidoacetic acid (NETFOSAA), N-Methyl perfluorooctane sulfonamidoacetic acid (NMEFOSAA), 4,8-Dioxa-3H-

perfluorononanoic acid (ADONA), hexafluoropropylene oxide dimer acid (HFPA-DA), 11-Chloroeicosafluoro-3- oxaundecane-1-sulfonic acid (11ClPF3OUDS) and 9-Chlorohexadecafluoro-3- oxanonane-1-sulfonic acid (9ClPF3ONS) are consistently detected in the groundwater. For the airport, perfluoropropionic acid (PFPA) tops the list in terms of PFAS; as for bulk fuel terminal/ refinery, PFOS has the highest content; for the chemicals, PFOA holds the highest concentration; for the chrome plating industry, PFOS is the dominant compound; As for Cleanup program sites, PFNA contains a 62% proportion of total PFAS, which also accounts for the largest proportion of a facility type. For fluoropolymer manufacturing, the predominant part is PFOS; in terms of military cleanup sites, the primary segment is 4:2 fluorotelomer sulfonic acid (4:2 FTS); regarding MSW landfill, perfluorobutanoic acid (PFBTA) is the most significant fraction, while other landfill's principal parts are PFOS and 6:2 fluorotelomer sulfonic acid (6:2 FTS). For Wastewater treatment plants, the main constituent is perfluorohexadecanoic acid (PFHXDA). Industrial's PFAS data are all equal to 0. The distribution of PFAS in these three categories, "Semiconductors and related devices", "no facility within 50 km" and "other" appears to be very uniform because the amount of data is relatively small.

According to the total PFAS contamination severity, the classification also falls into the following three classes: first class: facility with the greatest impact; second class: medium-impact facilities; and third class: facilities with modest impact.

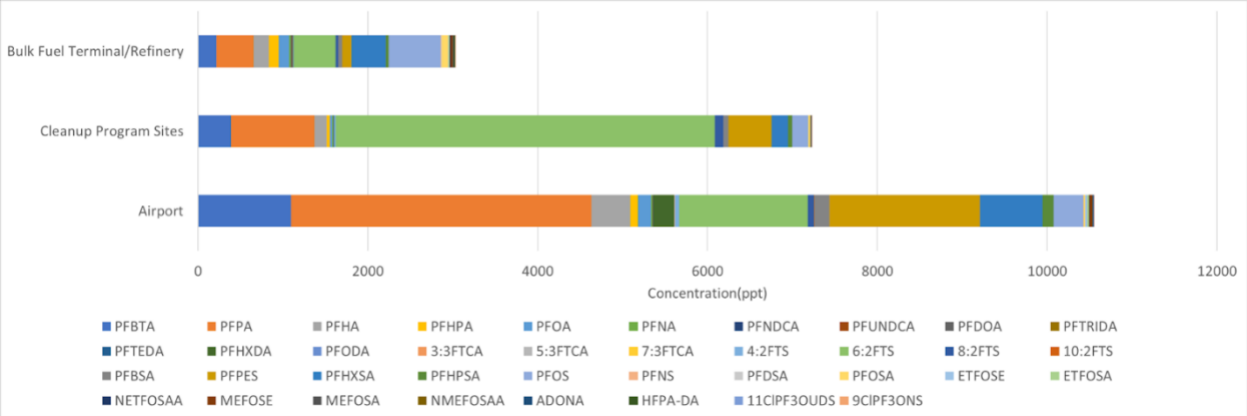

Figure 3.4. Class 1: The facilities with the most significant impact on California groundwater PFAS contamination PFAS profile

The first classification shown in Figure 3.4 indicates the total concentration of the PFAS average on bulk fuel terminal/ refinery, cleanup program sites, and airport. The three facilities have a concentration of PFAS ranging from 2000 ppt to above 10000 ppt. The highest facility in class 1 is the airport, with the greatest concentration of the component PFPA.

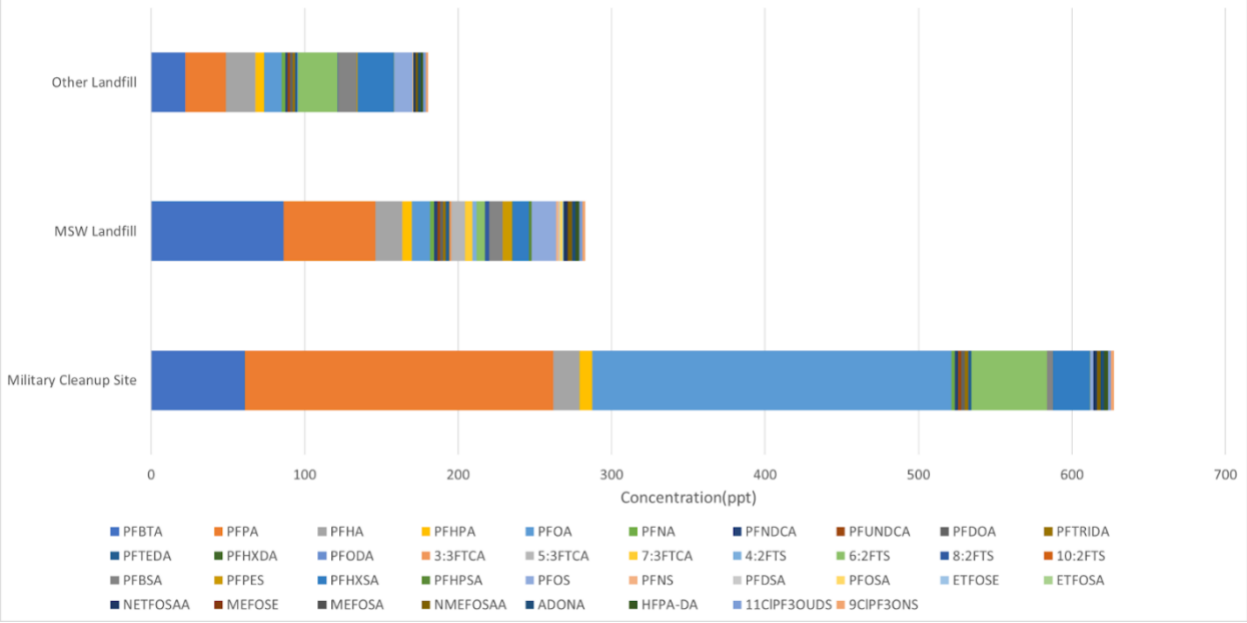

Figure 3.5. Class 2: The facilities with a medium impact on California groundwater PFAS contamination PFAS profile

The results shown in Figure 3.5 show that the range of the sum of all PFAS' averages on other landfill, MSW landfill, and military cleanup sites is from 150 ppt to above 600 ppt. The highest facility in class 2 is the military cleanup site, with the greatest concentration of the component PFOA.

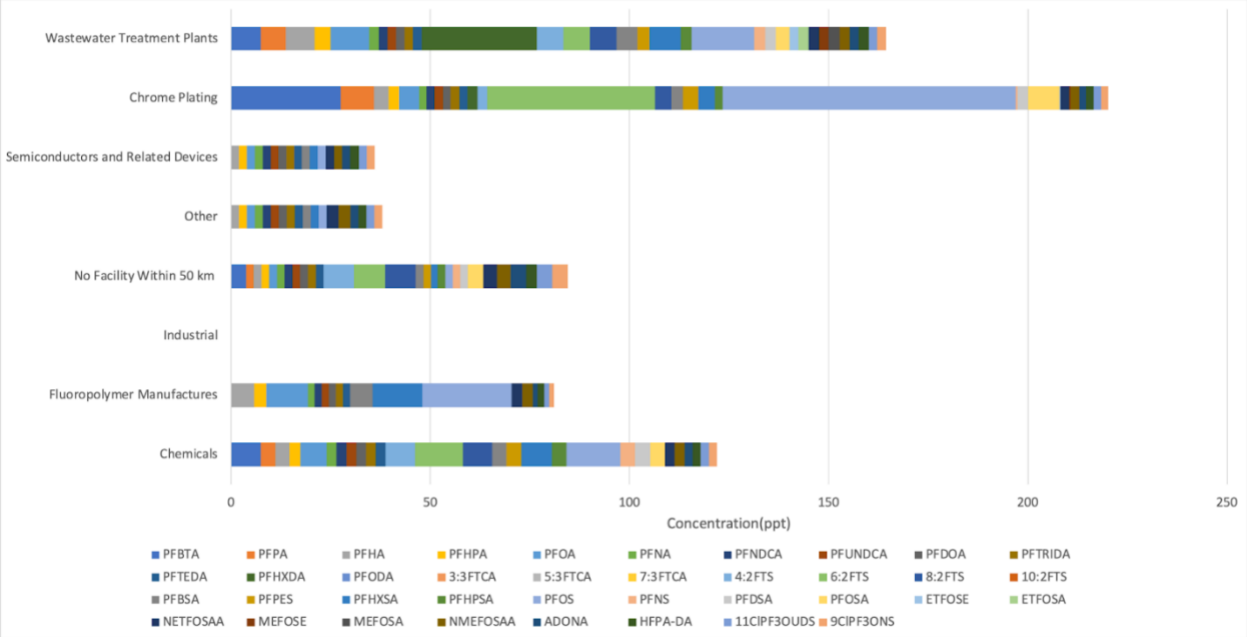

Figure 3.6. Class 3: The facilities with a modest impact on California groundwater PFAS contamination PFAS profile

The eight facility types shown in the third class have a range from 0 ppt to 250 ppt. In the last class, chrome plating is the highest concentration facility type, and the highest concentration of the component of chrome plating is PFOS.

While the 100% stacked bar contributes to illustrating the percentage of each PFAS subclass to the total PFAS, the regular stacked bar highlights the magnitude of the subclass concentration. I divided different facilities into three groups according to the total PFAS concentration to better see the distribution and amount of PFAS and PFAS subclasses of each facility type. Facilities in the first class include the airport, bulk fuel terminal/ refinery, and cleanup program sites; class 2 consist of military cleanup sites, MSW landfill, and other landfill.

The third class of facilities covers wastewater treatment plants, chrome plating, semiconductors, and related devices; other, no facility within 50 km, industrial, fluoropolymer manufactures, and chemicals. The 100% stacked bar chart and the regular stacked bar charts also demonstrate that Airports in California contributed the most to PFAS concentration.

Because of the different orders of magnitude of the pollution of the three classes, the PFAS issues from tier 1 facilities are the highest priority due to the greatest contamination concentration from the tier and the sum of the mean values of the PFAS subclasses all above 2000 ppt.

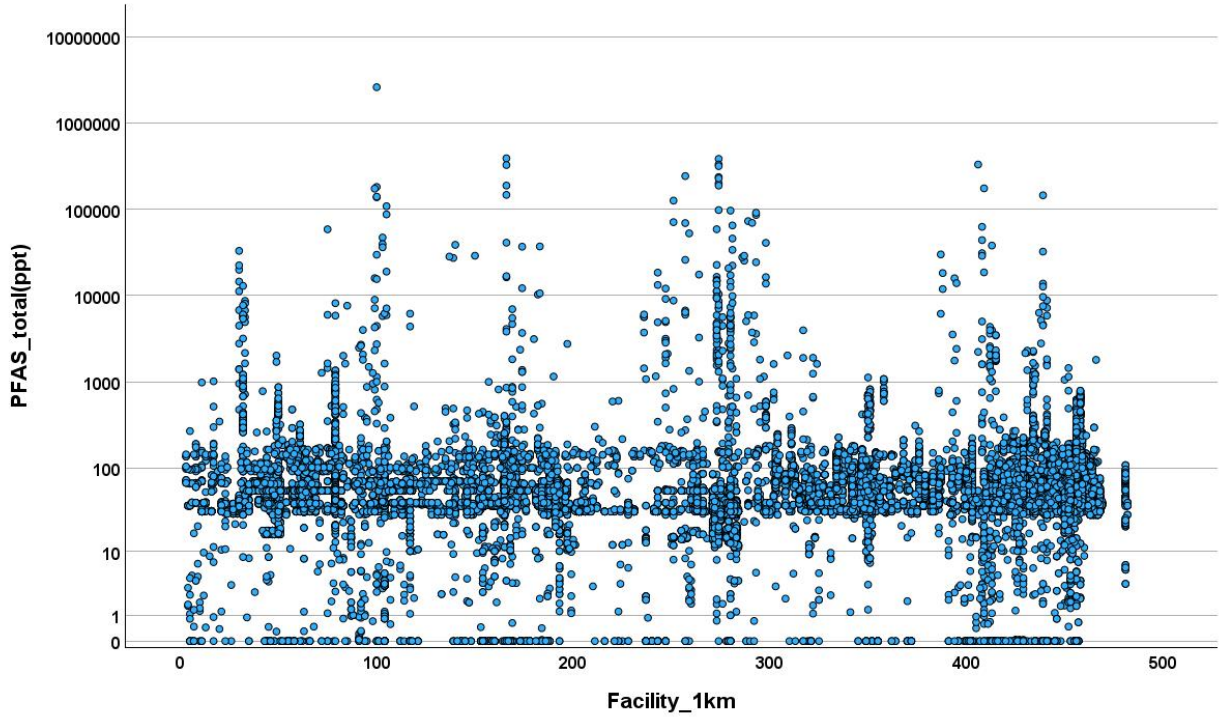

Figure 3.7. The scatter plots between the number of facilities within 1 km of each groundwater well and total PFAS concentration

The x-axis represents the number of facilities, and the y-axis represents the concentration of total PFAS. A logarithmic scale is used in the scatter plot. This result shown in Figure 3.7 is inconsistent with previous conjectures of a positive correlation between facility number and

PFAS concentration—no overall tendency for an increase or decrease of the PFAS concentration with the number of facility changes. Most of the concentrations of PFAS near the facilities are around 100 ppt, and there are concentrations of PFAS up to 100,000 ppt despite the distance between facilities. Calculated by the SPSS, the Spearman's correlation coefficient is 0.12, and the p-value is $5.20*10^{-81}$, which also proves that the relationship between the PFAS concentration and the facility number nearby is limited.

3.1.2 The Pearson and Spearman correlative heatmaps of PFAS interactions with environmental factors

Pearson's correlation coefficient(R) describes the direction and degree to which one variable is linearly related to another, while Spearman's rank correlation coefficient (Rho) is a non-parametric measure of correlation between variables that assess how well an arbitrary monotonic function could describe the relationship between two variables. According to the definition of the coefficient range[61,62], Spearman's rho and Pearson's r meanings are shown in Table 3.1. Because the focus of this part of the study is to investigate the relationship between PFAS and factors of air, soil condition, and weather, only coefficients greater than 0.4 ($p<0.05$) were selected for the study of the linkages within PFAS, and coefficients greater than 0.2 ($p<0.05$) were selected for the study of the relationship between factors air soil condition and weather.

Table 3.1. The meaning of Spearman and Pearson's correlation coefficient range

Spearman and Pearson's Correlation Coefficient (Positive or Negative)	Meaning
0.00 to 0.19	A very weak correlation
0.20 to 0.39	A weak correlation

0.40 to 0.69	A moderate correlation
0.70 to 0.89	A strong correlation
0.90 to 1.00	A very strong correlation

According to the work of Buck et al. and Camdzic D et al.[46,63], I classify the PFAS into 11 groups. They are short-chain PFCA, long-chain PFCA, fluorotelomer carboxylic acids, fluorotelomer sulfonic acids, short-chain PFSA, long-chain PFSA, perfluoroalkane sulfonamido substances, ADONA, HFPA-DA, 9ClPF3ONS, and 11ClPF3ONS. To emphasize the holistic nature of each group, I will be more focused on the relationship between the groups. The heatmap of the correlation analysis heatmap of PFAS with soil, air, and weather metrics is shown in Figure 3.8.

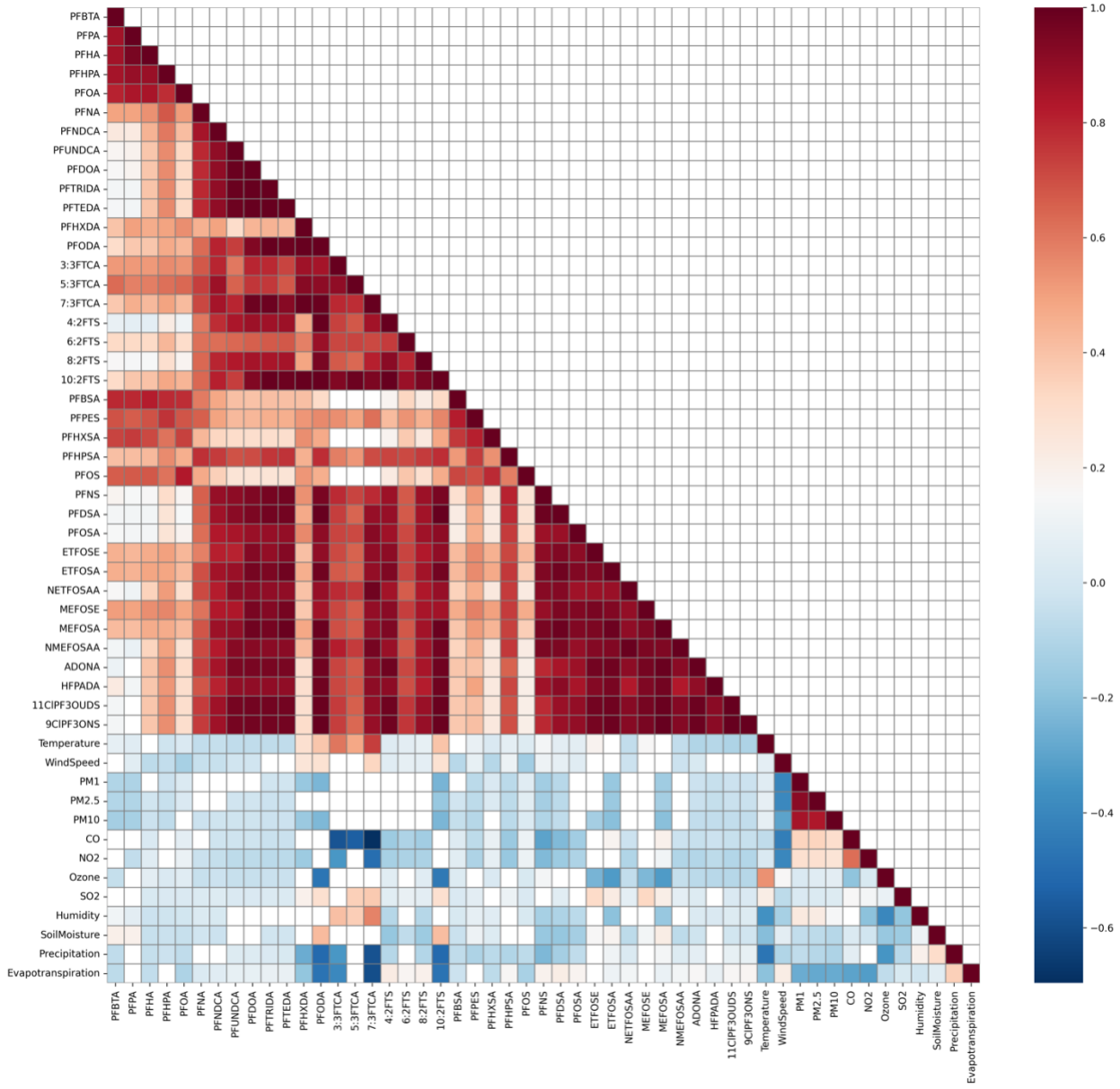

Figure 3.8. The Spearman's rank ($p<0.05$) correlation coefficient heatmap plot for PFAS and soil, air, and weather factors. The red color indicates a positive correlation, and the blue color represents the negative correlation between each of the two parameters, the darker the relationship between them is more robust.

As for the heatmap plot by Spearman's rho, short-chain PFCA have a moderate relationship with PFNA, 3:3 FTCA, 5:3 FTCA and perfluoroalkane sulfonamido substances(except NETFOSAA and NMEFOSAA), have moderate to strong correlations with short-chain PFSA and long-chain PFSA with less than eight carbons, short-chain PFCA except

PFBTA have a moderate relationship with 7:3 FTCA; long-chain PFCA have moderate to strong correlations with fluorotelomer carboxylic acids, moderate relationship with short-chain PFSA, long-chain PFCA (except PFOA) have moderate to very strong relationships with fluorotelomer sulfonic acids, long-chain PFCA (except PFOA, PFNA and PFHXDA) have strong to very strong relationships with long-chain PFSA with more than eight carbon (perfluorononane sulfonic acid (PFNS), perfluorodecane sulfonic acid (PFDSA) and perfluorooctanesulfonamide (PFOSA)), perfluoroalkane sulfonamido substances, ADONA, HFPA-DA, 11ClPF3OUDS and 9ClPF3ONS; fluorotelomer carboxylic acids have a strong relationship with fluorotelomer sulfonic acids and moderate relationship with perfluoropentane sulfonic acid (PFPES); fluorotelomer sulfonic acids have strong to very strong relationships with perfluoroalkane sulfonamido substances, ADONA, HFPA-DA, 11ClPF3OUDS and 9ClPF3ONS; perfluoroalkane sulfonamido substances have very strong correlations with ADONA, HFPA-DA, 11ClPF3OUDS and 9ClPF3ONS; last, ADONA and HFPA-DA have a very strong relationship with 11ClPF3OUDS and 9ClPF3OUDS. The correlations between PFAS and environmental variables are summarized in Table 3.2.

Table 3.2 The Spearman correlation coefficient between PFAS with air, soil, and weather

PFAS or PFAS group	**Correlation**	
PFBTA	Weak	Soil moisture
PFHXDA	Weak	Temperature, wind speed
	Negative weak	Precipitation
PFODA	Moderate	Soil moisture
	Weak	Temperature, wind speed, SO2

	Negative moderate	Ozone, precipitation, evapotranspiration
	Negative weak	PM2.5, PM10
3:3 FTCA	Moderate	Temperature, humidity
	Negative moderate	CO
	Negative weak	NO2, precipitation, evapotranspiration
5:3 FTCA	Moderate	Temperature
	Weak	SO2, humidity
	Negative moderate	CO
7:3 FTCA	Strong	Temperature
	Moderate	Humidity
	Weak	Wind speed, SO2
	Negative moderate	CO, NO2, precipitation, evapotranspiration
4:2 FTS	Weak	Evapotranspiration
10:2 FTS	Moderate	Soil moisture
	Weak	Temperature, wind speed, SO2
	Negative moderate	Ozone, precipitation, evapotranspiration
	Negative weak	PM2.5, PM10
PFNS	Negative weak	CO, NO2, precipitation
PFDSA	Weak	SO2

	Negative weak	CO
ETFOSE	Weak	SO2
	Negative weak	Ozone
ETFOSA	Weak	SO2
	Negative weak	Ozone, PM10
MEFOSE	Weak	SO2
	Negative weak	Ozone
MEFOSA	Weak	SO2, soil moisture
	Negative weak	Ozone

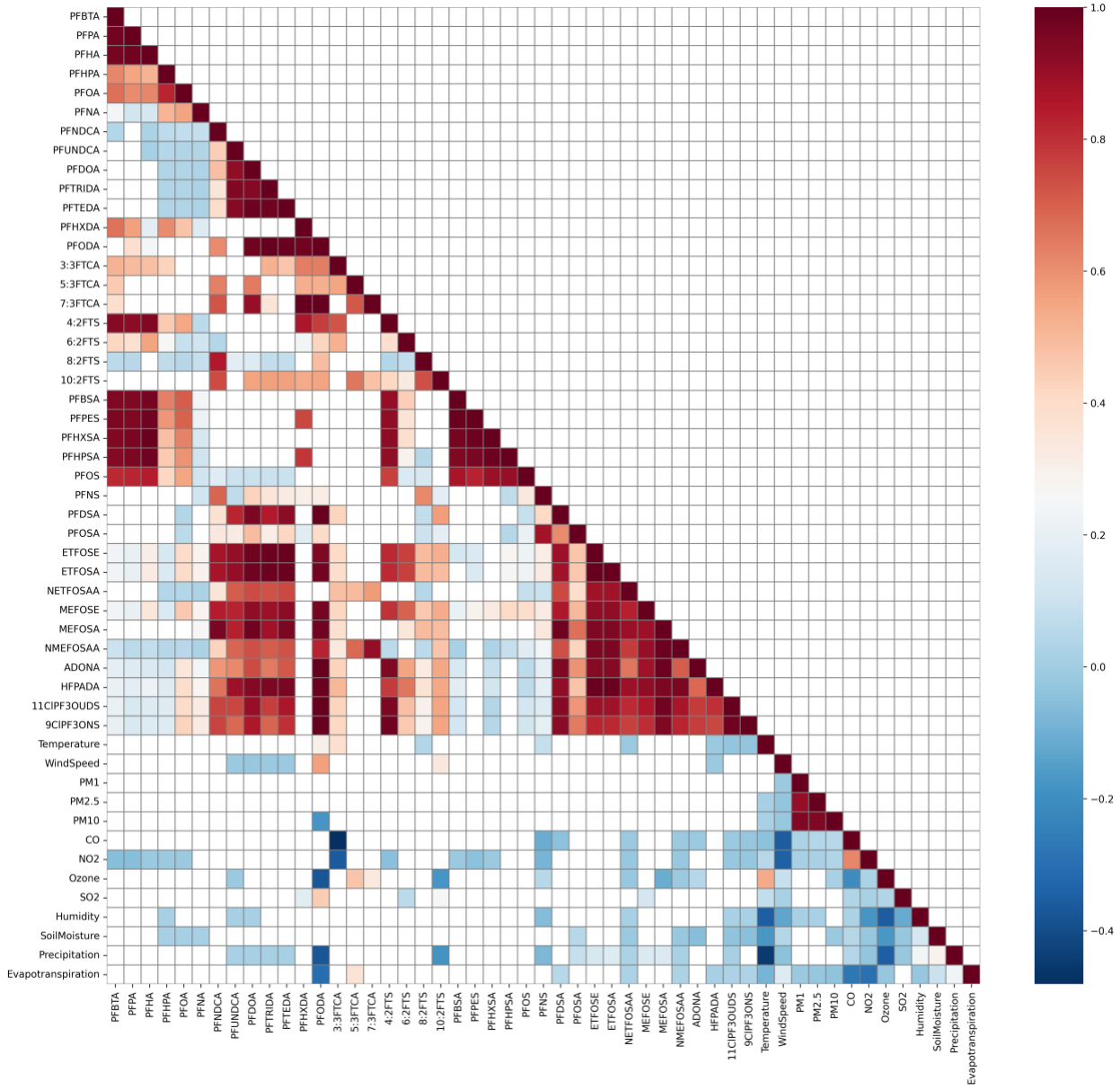

Figure 3.9. The Pearson's ($p<0.05$) correlation coefficient heatmap plot for PFAS and other factors

As the heatmap created by the Pearson correlation coefficient, short-chain PFCA have a moderate relationship with 3:3 fluorotelomer carboxylic acid (3:3 FTCA), short-chain PFCA (except PFHPA) have a moderate relationship with PFOA, a strong relationship with PFOS and a very strong relationship with 4:2 FTS, short-chain PFSA, PFHXSA, and perfluoroheptane sulfonic acid (PFHPSA), in other words, short-chain PFCA (except PFHPA) have very a strong

relationships with PFSA with four to seven carbons; except PFNA and PFHXDA, long-chain PFCA have strong to very strong with perfluoroalkane sulfonamido substances, ADONA, HFPA-DA, 11CLPF3OUDS and 9CLPF3ONS; fluorotelomer sulfonic acids have moderate to strong relationships with perfluoroalkane sulfonamido substances (except NETFOSAA, MEFOSA and NMEFOSAA); perfluoroalkane sulfonamido substances have strong to very strong relationships with ADONA, HFPA-DA, 11ClPF3OUDS and 9ClPF3ONS; ADONA and HFPA-DA have very strong relationships with 11ClPF3OUDS and 9ClPF3ONS. Also, the relationship of PFAS with environmental factors is shown in Table 3.3.

Table 3.3. The Pearson correlation coefficient between PFAS with air, soil, and weather

PFAS or PFAS group	**Correlation**	
PFODA	Moderate	Wind speed, SO2
	Negative weak	Ozone, precipitation, evapotranspiration
3:3 FTCA	Negative moderate	CO
	Negative weak	NO2
5:3 FTCA	Moderate	Ozone
	Weak	Evapotranspiration
7:3 FTCA	Weak	Ozone
10:2 FTS	Weak	SO2

Pearson correlation is used to measure the linear relationship, while the Spearman correlation is used to determine the monotonic correlation. Besides, since the purpose of this study emphasizes the relationship between PFAS and air, soil, and weather factors, the connection within PFAS is described in words and only takes the coefficient larger than 0.7 into

account. According to Table 3.2 and Table 3.3, most relationships within PFAS and the correlation between PFAS and air, soil, and weather elements are more complex than linear correlation, which involves nonlinear patterns. The figures and the tables show that the correlation between PFAS and the PFAS subclass is much stronger than the relationship between PFAS and other air pollutants, soil conditions, and weather factors. To sum up, from the Pearson correlation coefficient, the fluorotelomer carboxylic acids have a stronger correlation between air, weather, and soil-related data than other PFAS. As for the coefficient from Spearman, fluorotelomer carboxylic acids and perfluoroalkane sulfonamido substances both have a moderate correlation with SO2.

3.2 Comparative analysis of facility-specific distribution and variability of PFAS subclasses with emphasis on predominant compounds

To know the variability and diversity of the PFAS data used in this research, I chose the standard deviation mean bar and found out the data I analyzed has an extensive range of values. A logarithmic scale is utilized to approve the figure of the airport, chemicals, cleanup program, bulk fuel terminal/ refinery, chrome plating, MSW landfills, other landfills, military privatized sites, and wastewater treatment plants and help for assessment. The error bar in the plot represents the standard deviation of the mean of each PFAS species.

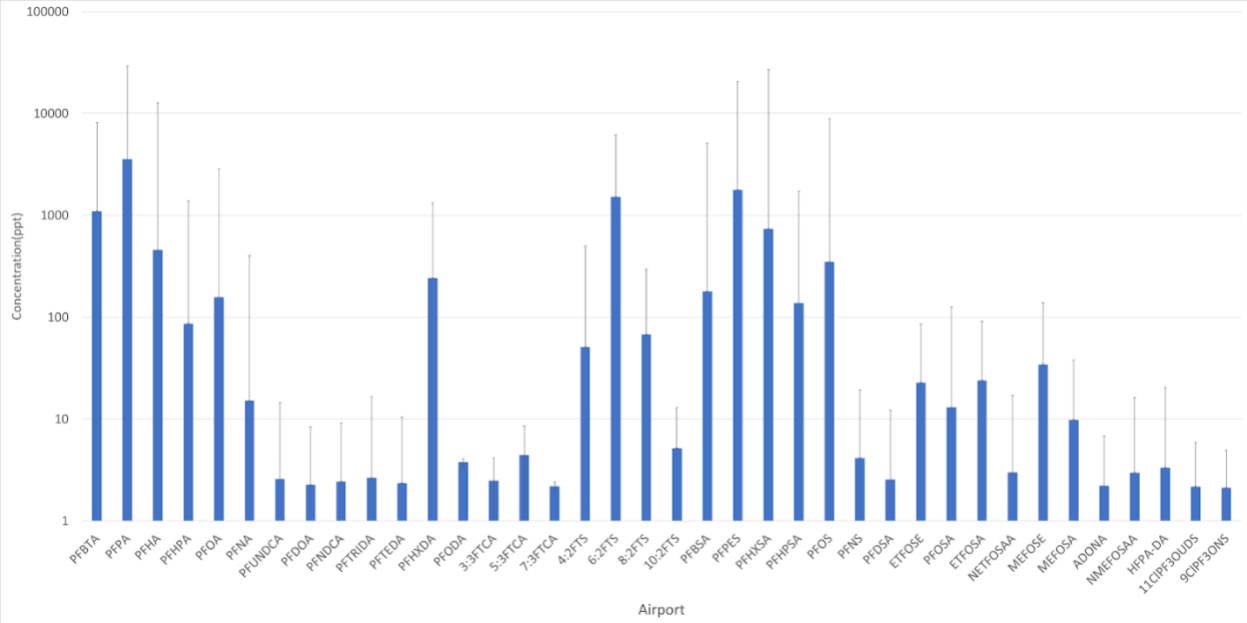

Figure 3.10. The mean and standard deviation bar chart of the airport

The highest concentration of the airport is PFPA, which is a short-chain PFCA, and it is clear that the total concentration of short-chain PFCA is much higher than the concentration of long-chain PFCA, at the same time, the highest PFSA is also a short-chain PFSA: PFPES.

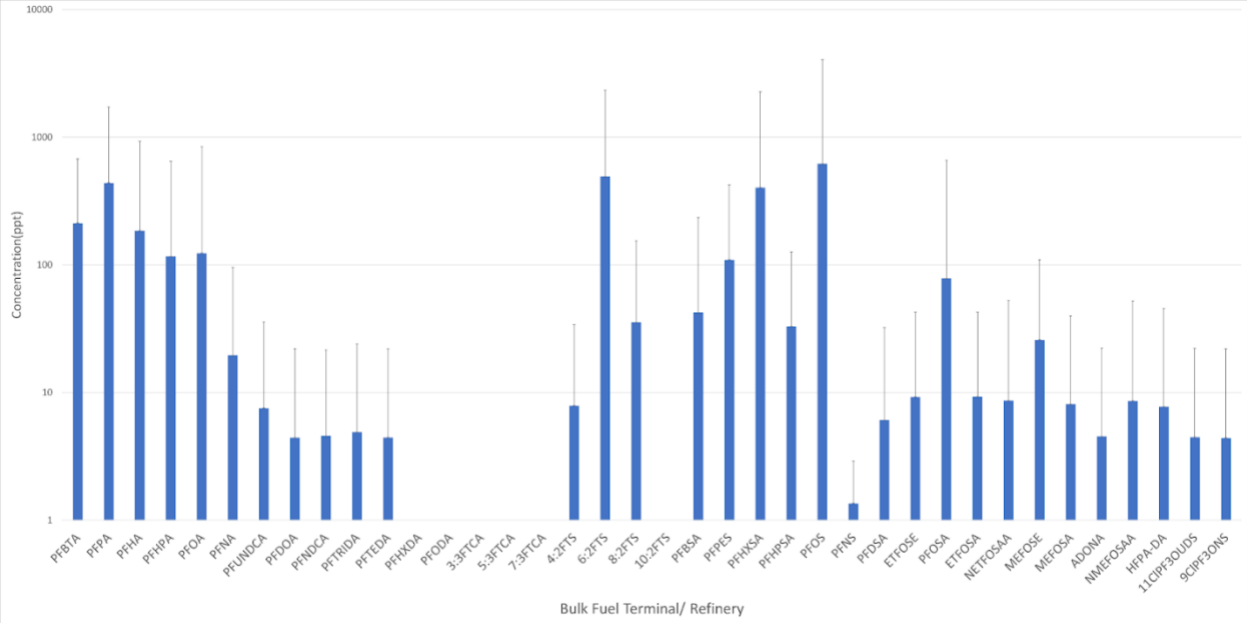

Figure 3.11. The mean and standard deviation bar chart of bulk fuel terminals/ refinery

The highest PFAS in bulk fuel terminal/ refinery is PFOS, which is also a long-chain PFSA. In this type of facility, the short-chain PFCA concentration is higher than the long-chain PFCA, and the long-chain PFSA concentration is higher than the short-chain PFSA.

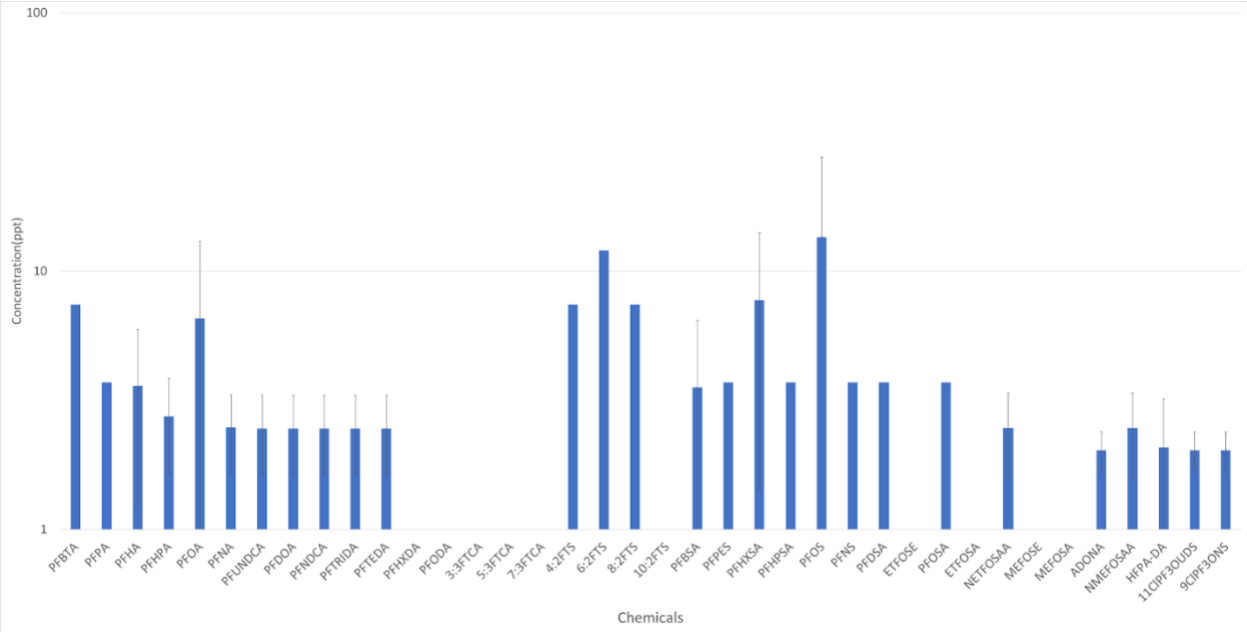

Figure 3.12. The mean and standard deviation bar chart of chemicals

For the chemicals, the concentration of every short-chain PFCA is higher than every long-chain PFCA; the highest concentration PFAS is PFOS, which is a long-chain PFSA.

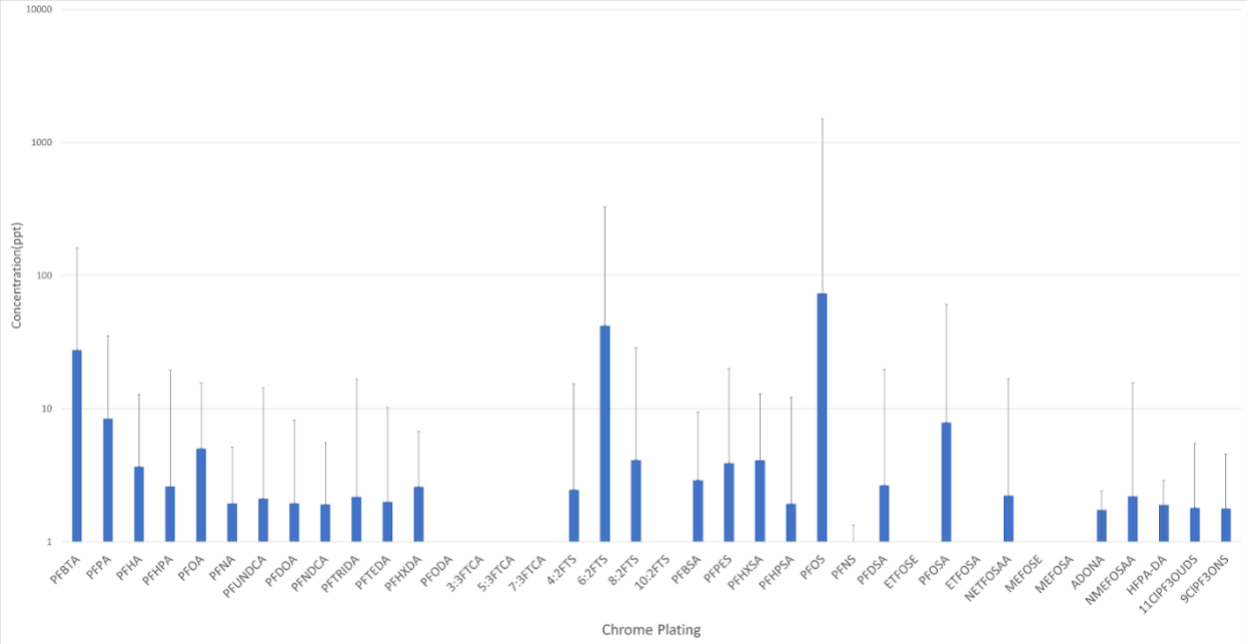

Figure 3.13. The mean and standard deviation bar chart of chrome plating

For the chrome plating facility, the highest PFAS is PFOS, which is a long-chain PFSA. Above all, the highest concentration of PFCA is a short-chain PFCA: PFBTA. Short-chain PFCA concentration is higher than long-chain PFCA, and the long-chain PFSA is higher than short-chain PFSA.

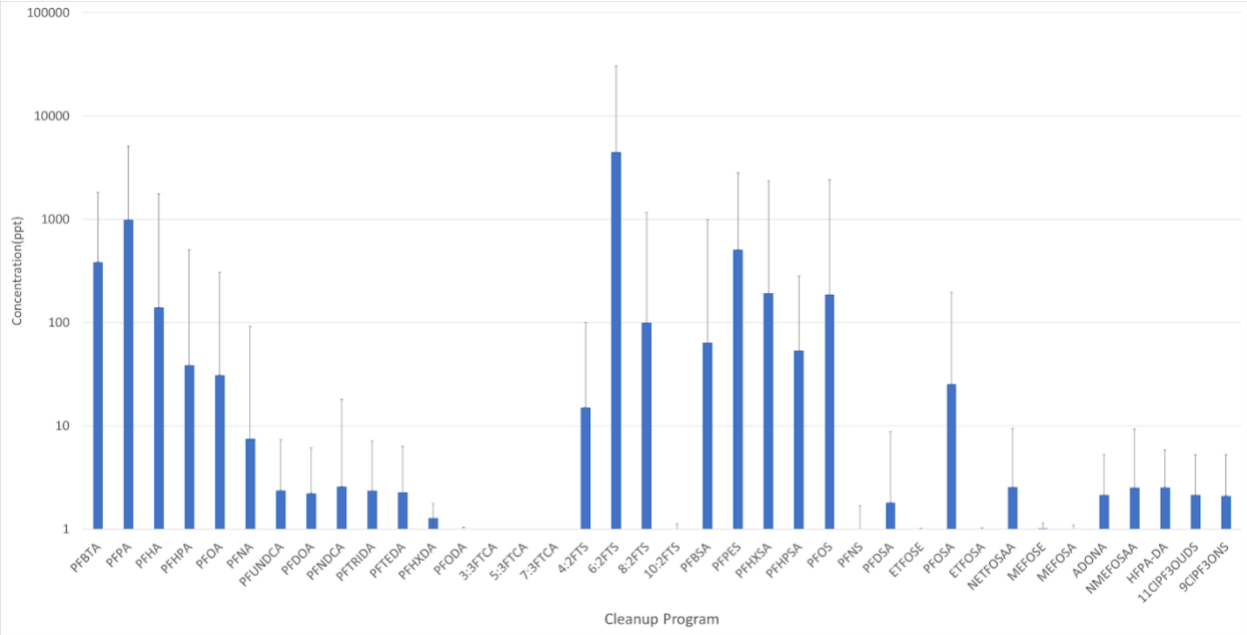

Figure 3.14. The mean and standard deviation bar chart of cleanup program sites

For the cleanup program, the highest concentration of PFAS is 6:2 FTS, short-chain PFCA is much higher than long-chain PFCA, and the highest PFCA is PFPA, a short-chain PFCA, the average of short-chain PFSA is higher than long-chain PFSA, the highest PFSA is PFPES, a short-chain PFSA.

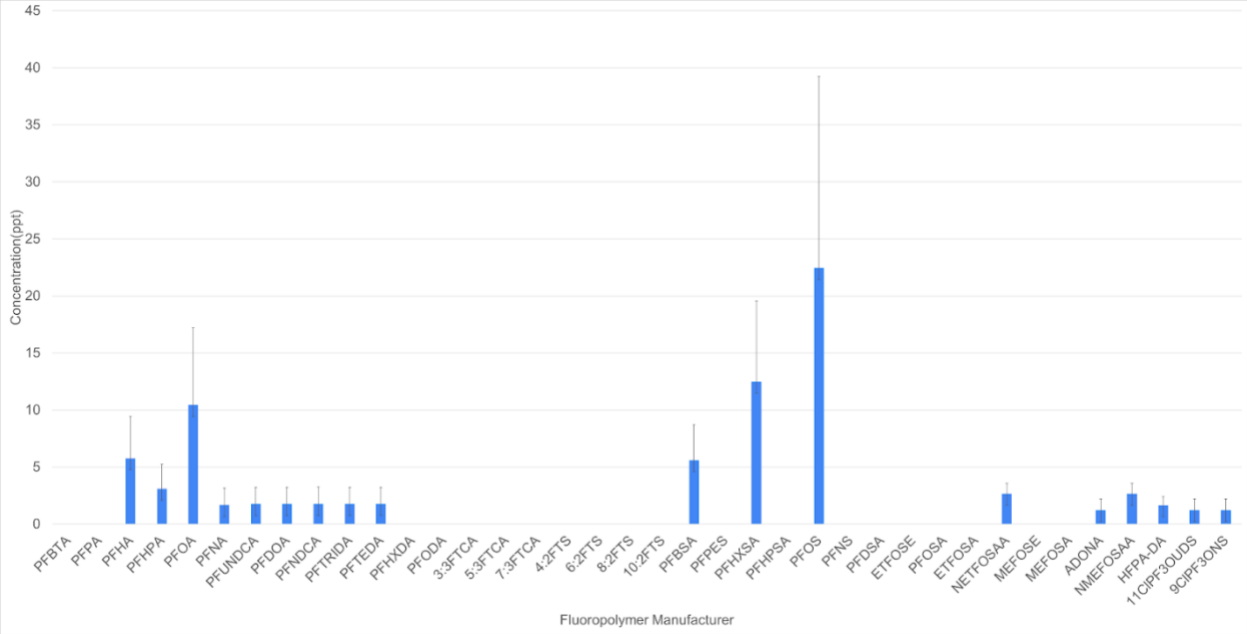

Figure 3.15. The mean and standard deviation bar chart of fluoropolymer manufacturer

For the fluoropolymer manufacturer, the highest concentration of PFAS is PFOS; the average of short-chain PFCA is more significant than that of long-chain PFCA, while the average of the long-chain PFSA is higher than that of the short-chain PFSA.

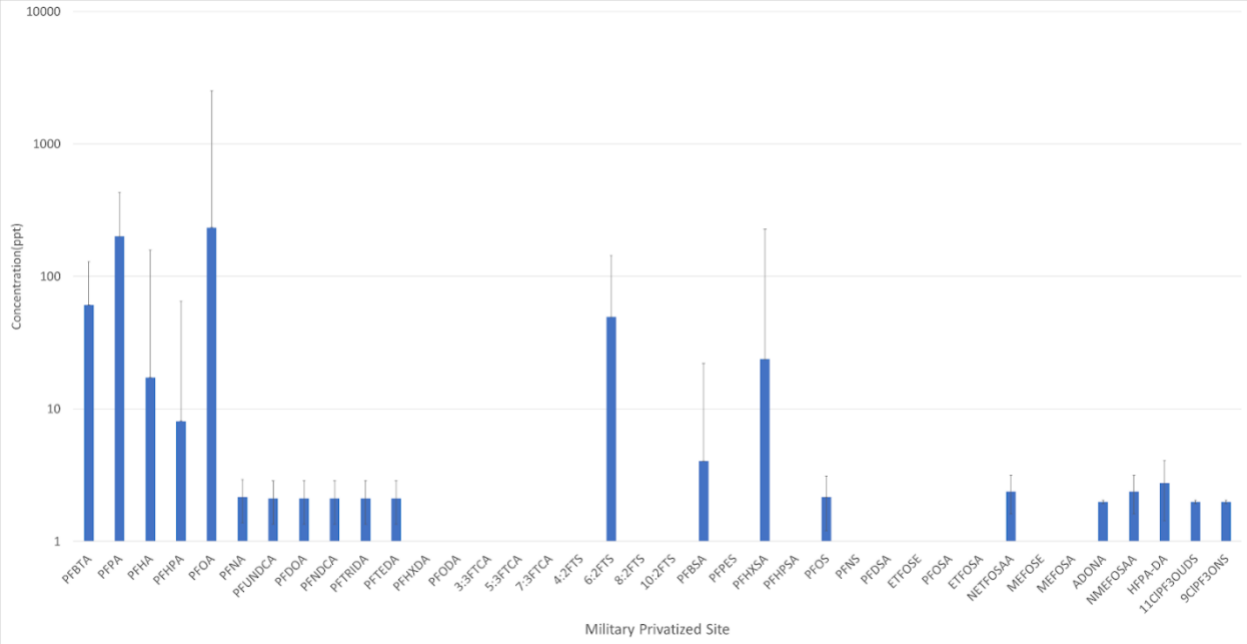

Figure 3.16. The mean and standard deviation bar chart of military cleanup sites

Both short-chain PFCA and short-chain PFSA concentrations are much higher for military cleanup sites than long-chain PFCA and long-chain PFSA. The highest PFAS: PFOA is a long-chain PFCA.

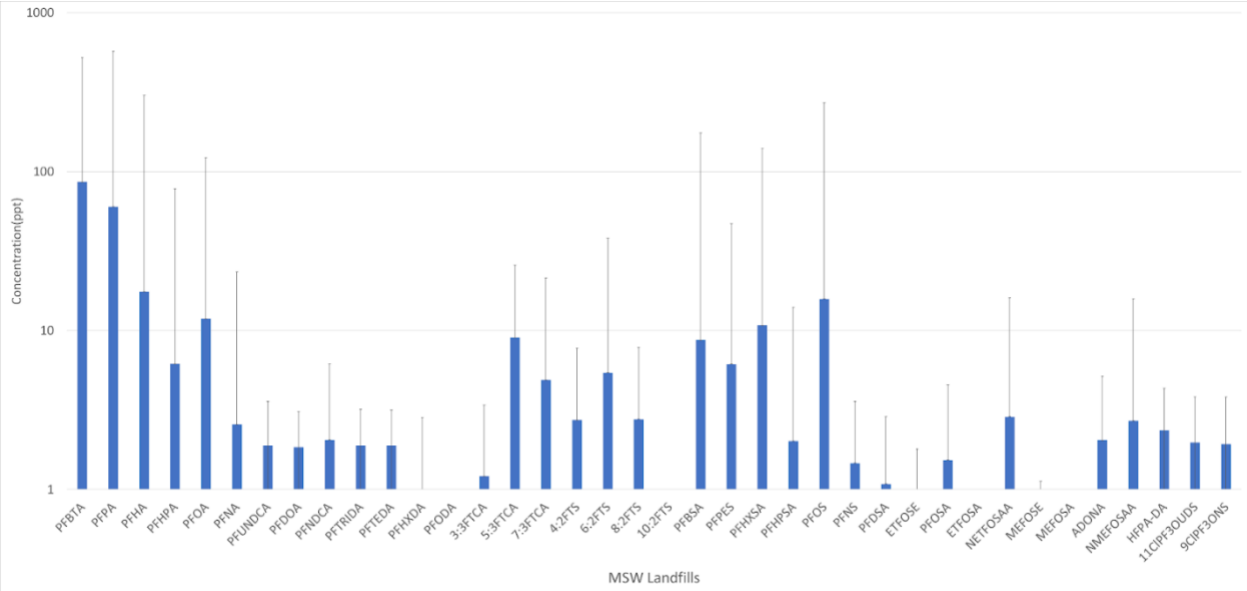

Figure 3.17. The mean and standard deviation bar chart of the MSW landfill

The MSW landfill has a higher short-chain PFCA than long-chain PFCA; the short-chain and long-chain PFSA are around similar levels. The highest concentration PFAS is a short-chain PFCA: PFBTA.

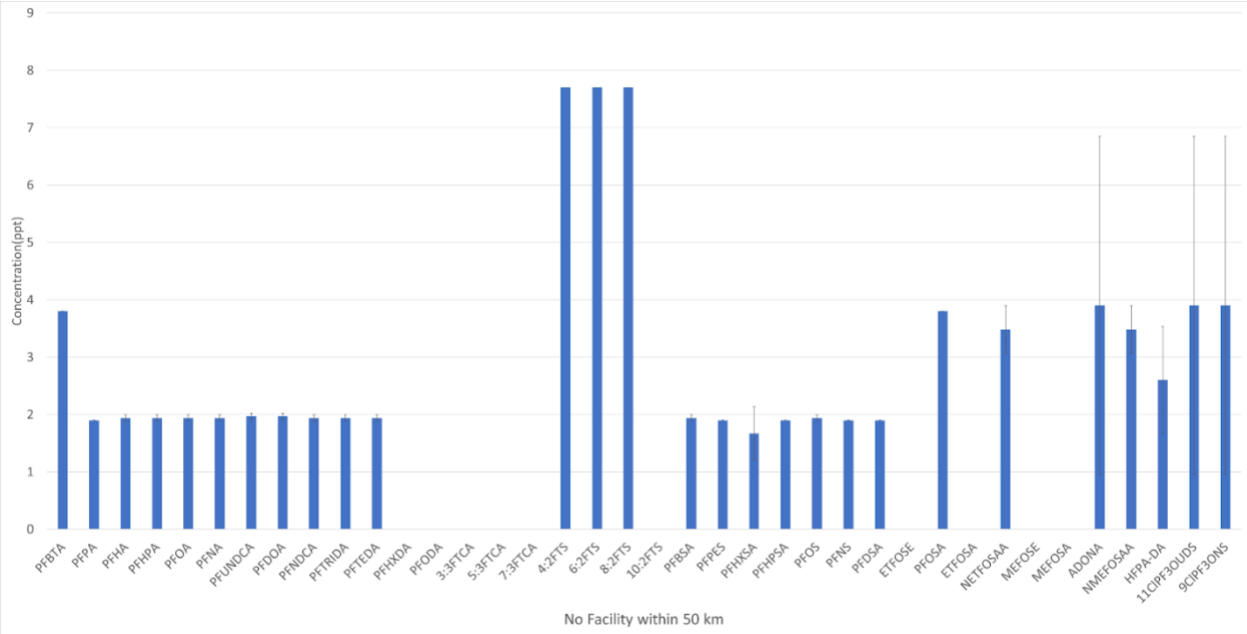

Figure 3.18. The mean and standard deviation bar chart of no facility within 50 km

As for the type of no facility within 50 km, Flurotelomer sulfonic acids account for the largest concentration. PFCA and PFSA are at the same level despite the long-chain and short-chain.

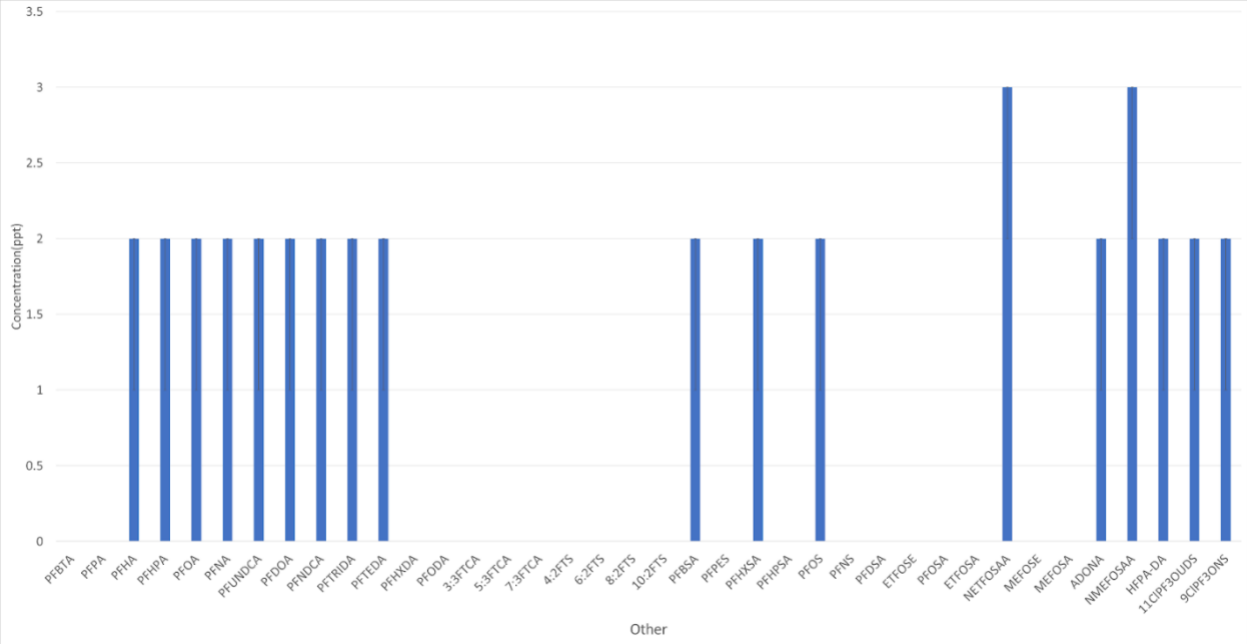

Figure 3.19. The mean and standard deviation bar chart of other facility type

The highest concentration of the other type of facilities PFAS are NETFOSAA and NMEFOSAA, they are both perfluoroalkane sulfonamido substances; PFSA and PFCA are at precisely the same level, but the total concentration of PFCA is higher than PFSA.

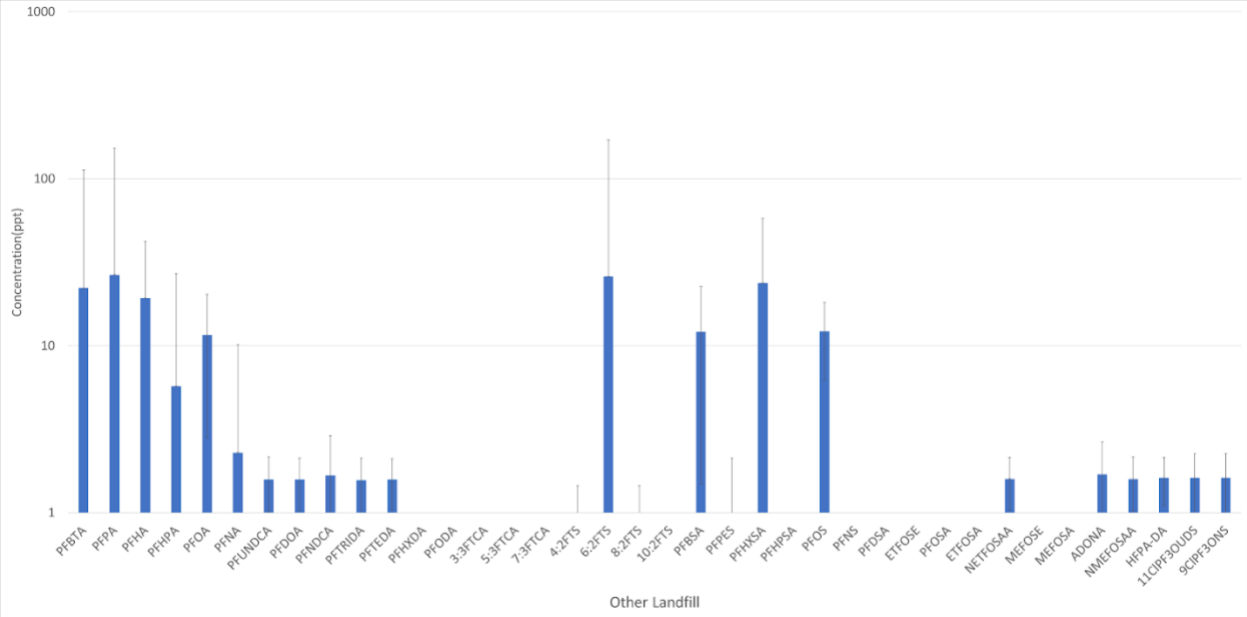

Figure 3.20. The mean and standard deviation bar chart of other landfill

For the other landfill, the short-chain PFCA concentration is much higher than the long-chain PFCA, while the long-chain PFSA concentration is higher than the short-chain PFSA. And the highest concentration PFAS is a short-chain PFCA: PFPA.

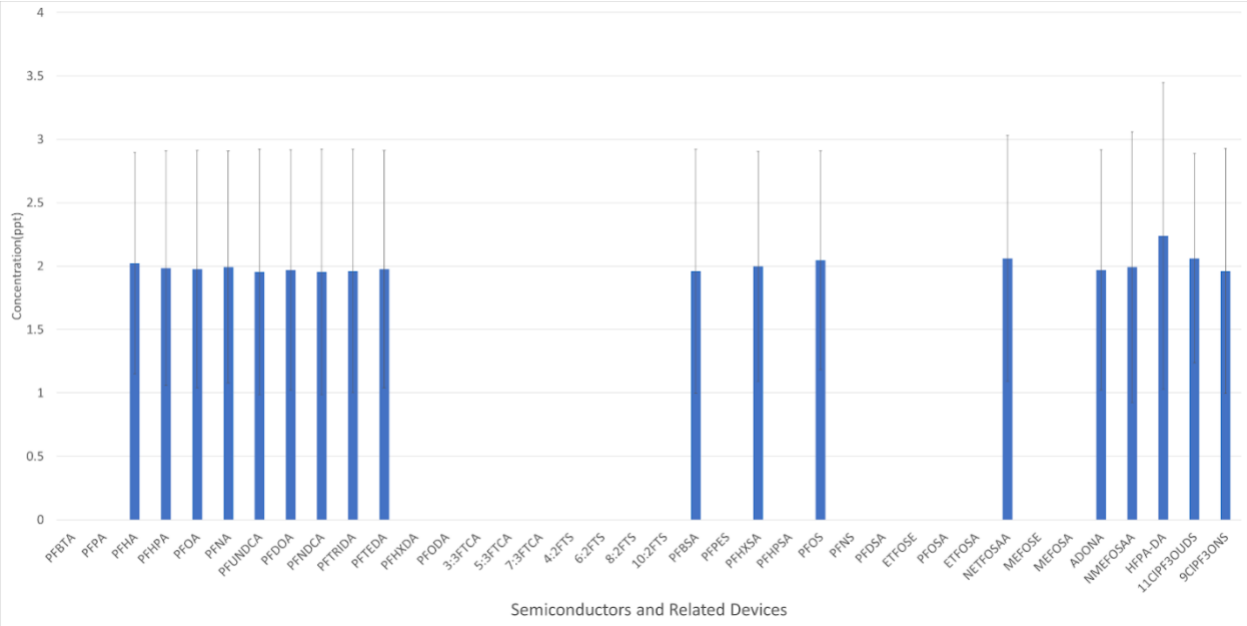

Figure 3.21. The mean and standard deviation bar chart of semiconductors and related devices

For the semiconductors and related devices, the distribution of each PFAS looks even, both around 2 ppt. The detected PFCA are much more than PFSA.

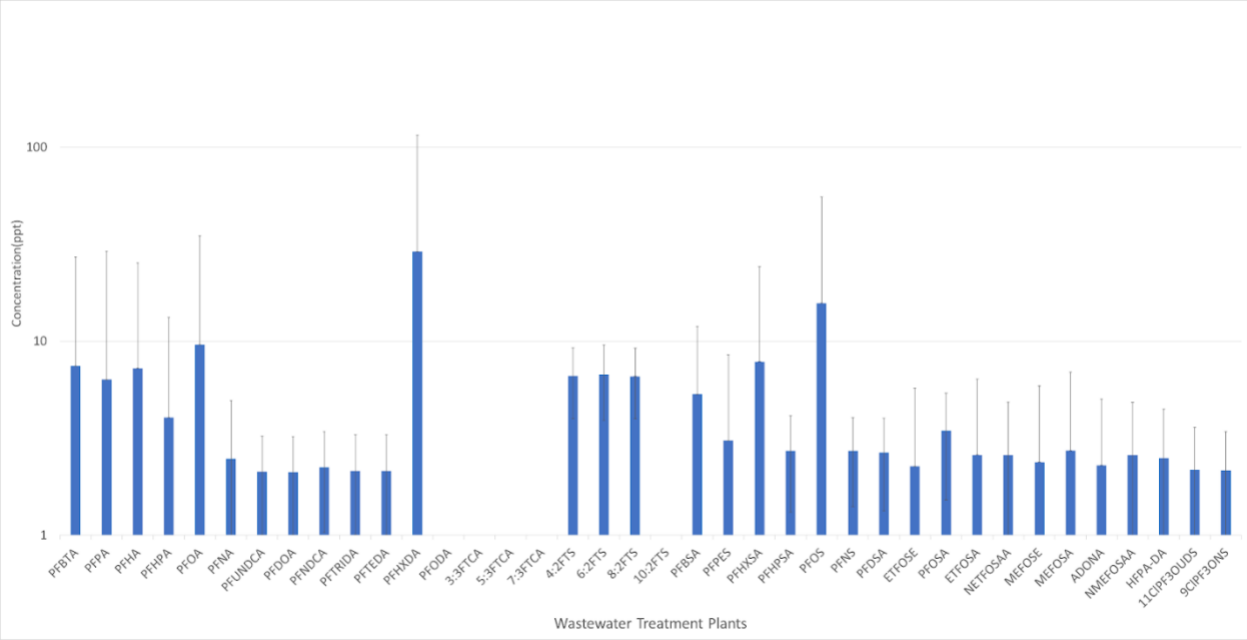

Figure 3.22. The mean and standard deviation bar chart of wastewater treatment plants

As for the wastewater treatment plants, the short-chain PFCA concentration is higher than the long-chain PFCA. However, the highest concentration is a long-chain PFCA: PFHXDA. The long-chain PFSA is higher than the short-chain PFSA.

In most cases, short-chain PFCA concentration is higher than long-chain PFCA, while the long-chain PFSA are higher than short-chain PFSA. Some research shows that more short-chain PFAS are discovered in the groundwater due to the less mobility of long-chain PFAS, which would be adsorbed by soil and sediment before entering the groundwater[64–66]. So, the phenomenon that long-chain PFCA concentration is lower than short-chain PFCA can be explained by the reason. However, with the situation within the PFSA, it may be because the number of short-chain PFSA is only two, while the amount of long-chain PFSA is 6.

To better illustrate the main PFAS in each facility, I list the three PFAS with the highest concentration of each industry from the above figures in Table 3.4. Because the concentration in the facility type industry is 0, the plot of the industry wasn't put on this part.

Table 3.4. The top three PFAS with the highest concentration in each facility

Facility type	Top 3 highest PFAS
No facility within 50 km	4:2FTS, 6:2FTS, 8:2FTS
Airport	PFPA, 6:2FTS, PFPES
Chemical	PFOS, 6:2FTS, PFBTA
Cleanup program	6:2FTS, PFPA, PFPES
Fluoropolymer manufacturers	PFOS, PFHXSA, PFOA
Bulk fuel terminal/refinery	PFOS, 6:2FTS, PFPA
Chrome plating	PFOS, 6:2FTS, PFBTA
MSW landfills	PFBTA, PFPA, PFHA
Other landfill	PFPA, 6:2FTS, PFHXSA
Military privatized site	PFOA, PFPA, PFBTA
Other	NETFOSAA, NMEFOSAA
Semiconductor and related device	HFPA-DA, 11CLPF3OUDS, NETFOSAA
Wastewater treatment plants	PFHXDA, PFOS, PFOA

As shown in Table 3.4, 6:2 FTS is the most widespread PFAS, present in over half (53.8%) of the facilities' top three lists. Besides, PFOS is the most frequently occurring highest PFAS above all the facility types.

3.3 Comparison with previous study data

To verify the credibility of the data used in this study, relevant research results were selected to compare with. According to Clara et al.'s[8] study in Australia, PFOS is the highest emission from the metal industry, and the average of the metal industry PFAS concentration is

60 ppt. These results show a high similarity between the highest PFAS. For the PFOS concentration, the data I used is higher than the previous study from Clara et al. The probable reasons for the disparity are Clara et al.'s study only focuses on 11 PFAS, and the PFAS concentration was tested in 2008, 15 years before now. From Bao et al.'s study[67] at a fluoropolymer industrial park in China, the top three highest PFAS are PFBSA, PFOA, and PFHXSA, which are also very similar to my results, which are PFOS, PFHXSA, and PFOA, also in my study, PFBSA is the 5th highest concentration. From Alnehem's study[68], who tested the hard chromium plating site in Iggesund's groundwater PFAS. The highest 3 PFAS from the research are PFOS, perfluorobutane sulfonic acid (PFBuS), and PFHXSA; the dominant subclass is PFOS, which is the same. However, in the datasets analyzed in my study, PFBuS was not detected in California. 6:2 FTS was not detected in Alnehem's research.

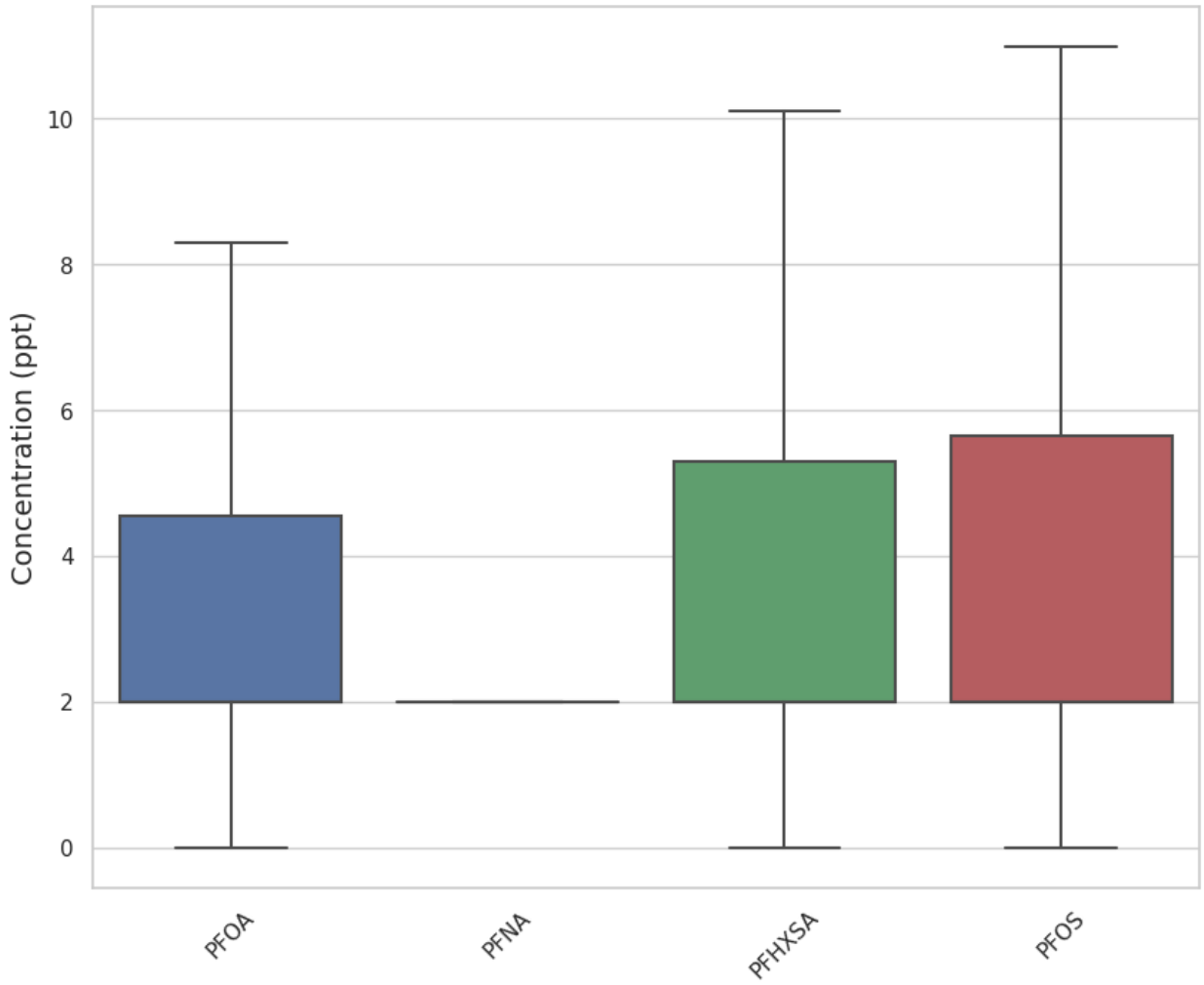

Figure3.23. The box plot of four PFAS: PFOA, PFNA, PFHXSA, and PFOS in the airport. Different colors show different PFAS.
PFOA:(minimum: 0, lower quartile:2, median:2, upper quartile:4.6, maximum: 8.5)
PFNA:(minimum: 0, lower quartile:2, median: 2, upper quartile: 2, maximum: 2)
PFHXSA:(minimum: 0, lower quartile:2, median: 2, upper quartile: 5.3, maximum:10.1)
PFOS:(minimum: 0, lower quartile:2, median: 2, upper quartile: 5.7, maximum:11)

Since the median of those four PFAS is the same, both equal to 2 ppt, comparing the position of the boxes and getting the sequence from highest to lowest: PFOS, PFHXSA, PFOA, and PFNA. The order in Carey et al.'s[27] research results showed the same succession.

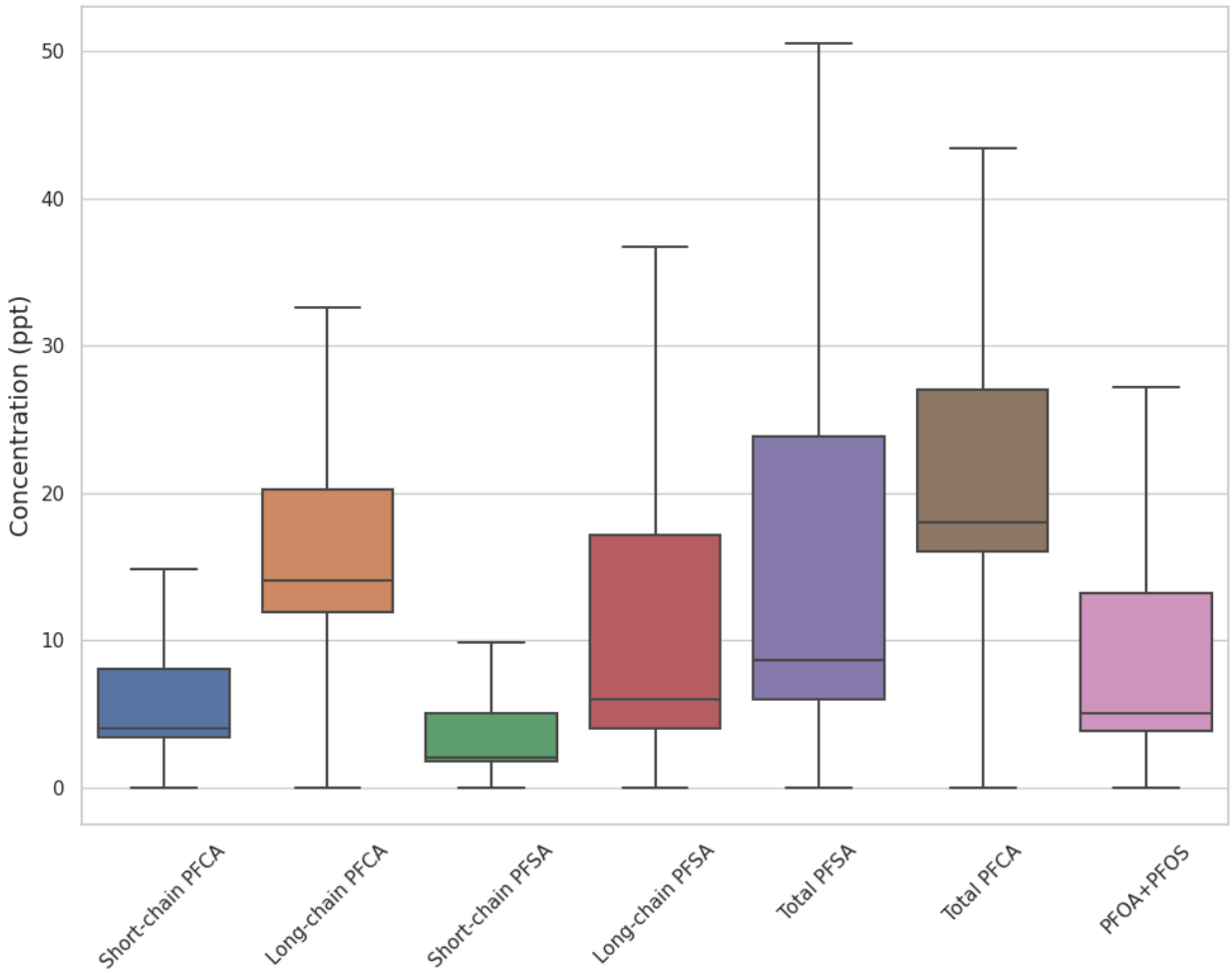

Figure 3.24. Box plots of MSW landfills
Short-chain PFCA:(minimum: 0, lower quartile: 3.4, median: 4, upper quartile: 8, maximum:14.8)
Long-chain PFCA:(minimum: 0, lower quartile: 11.9, median:14, upper quartile: 20.2, maximum:32.6)
Short-chain PFCA:(minimum: 0, lower quartile:1.8, median: 4, upper quartile: 8, maximum: 14.8)
Long-chain PFSA:(minimum: 0, lower quartile: 4, median:6, upper quartile: 17.1, maximum: 36.7)
Total PFSA:(minimum: 0, lower quartile:6, median:8.6, upper quartile:23.8, maximum:50.5)
Total PFCA:(minimum: 0, lower quartile: 16, median:18, upper quartile: 27, maximum: 43.4)
PFOA+PFOS:(minimum: 0, lower quartile:3.5, median: 5, upper quartile: 13.2, maximum: 27.2)

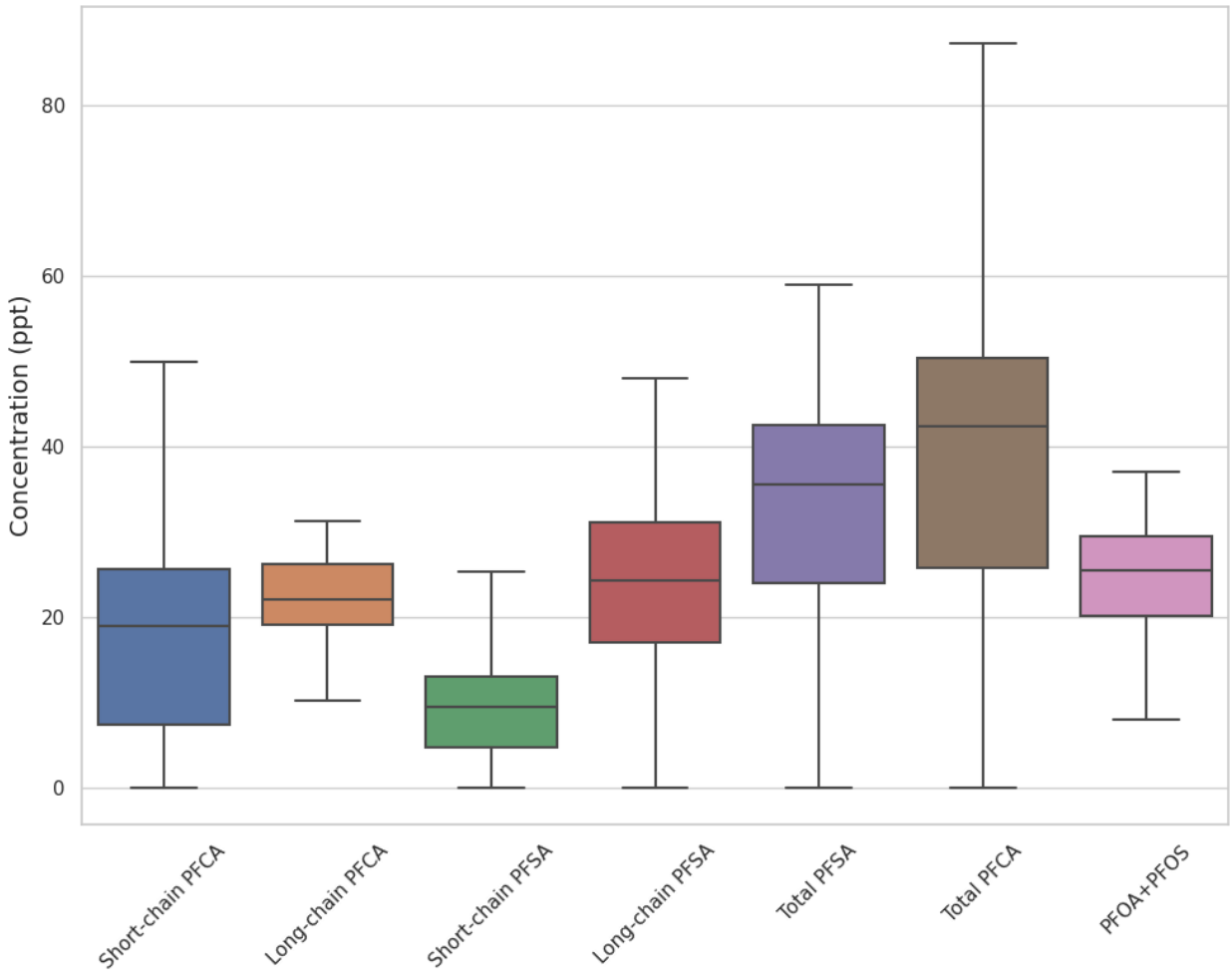

Figure 3.25. Box plots of other landfills
Short-chain PFCA:(minimum: 0, lower quartile: 7.3, median: 18.9, upper quartile: 25.6, maximum:49.9)
Long-chain PFCA:(minimum: 10.1, lower quartile: 19, median: 22, upper quartile: 26.2, maximum:31.2)
Short-chain PFCA:(minimum: 0, lower quartile:4.7, median: 9.4, upper quartile: 13, maximum: 25.3)
Long-chain PFSA:(minimum: 0, lower quartile: 16.9, median: 24.2, upper quartile: 31, maximum: 48)
Total PFSA:(minimum: 0, lower quartile: 24, median: 35.5, upper quartile: 43, maximum: 59)
Total PFCA:(minimum: 0, lower quartile: 25.8, median: 42.27, upper quartile: 50.4, maximum: 87.2)
PFOA+PFOS:(minimum: 0, lower quartile: 20.1, median: 25.4, upper quartile: 30, maximum: 37)

Comparing Zhang et al.'s study[69], which focuses on landfill PFAS in groundwater and stormwater, has found the concentration of PFCA, PFSA, and PFOA+PFOS. According to

Zhang et al.'s results, the order of magnitude of the median is short-chain PFCA, long-chain PFCA, PFOA+PFOS, long-chain PFSA, and short-chain PFSA, and the median of total PFCA is larger than PFSA. The sequence of concentration magnitude of the median is in the order of my result of MSW landfills: Long-chain PFCA, long-chain PFSA, PFOA+PFOS, short-chain PFCA, short-chain PFSA, the median of PFCA is larger than PFSA. The sequence of concentration magnitude of the median is in the order of my result of other landfills: PFOA+PFOS, long-chain PFSA, long-chain PFCA, short-chain PFCA, and short-chain PFSA. Although the sequence is different, and the concentration varies. This is because Zhang sampled different kinds of landfills and merged the data from all the sampling sites into one boxplot. Above all, from Zhang's and this study's results, there is one point in common: the concentration of the total PFCA is larger than PFSA.

3.4 Assessing the distribution of PFAS contamination in California groundwater systems

The area of the dots shown in Figure 3.26 demonstrates the level of the concentration, different colors represent different facility types.

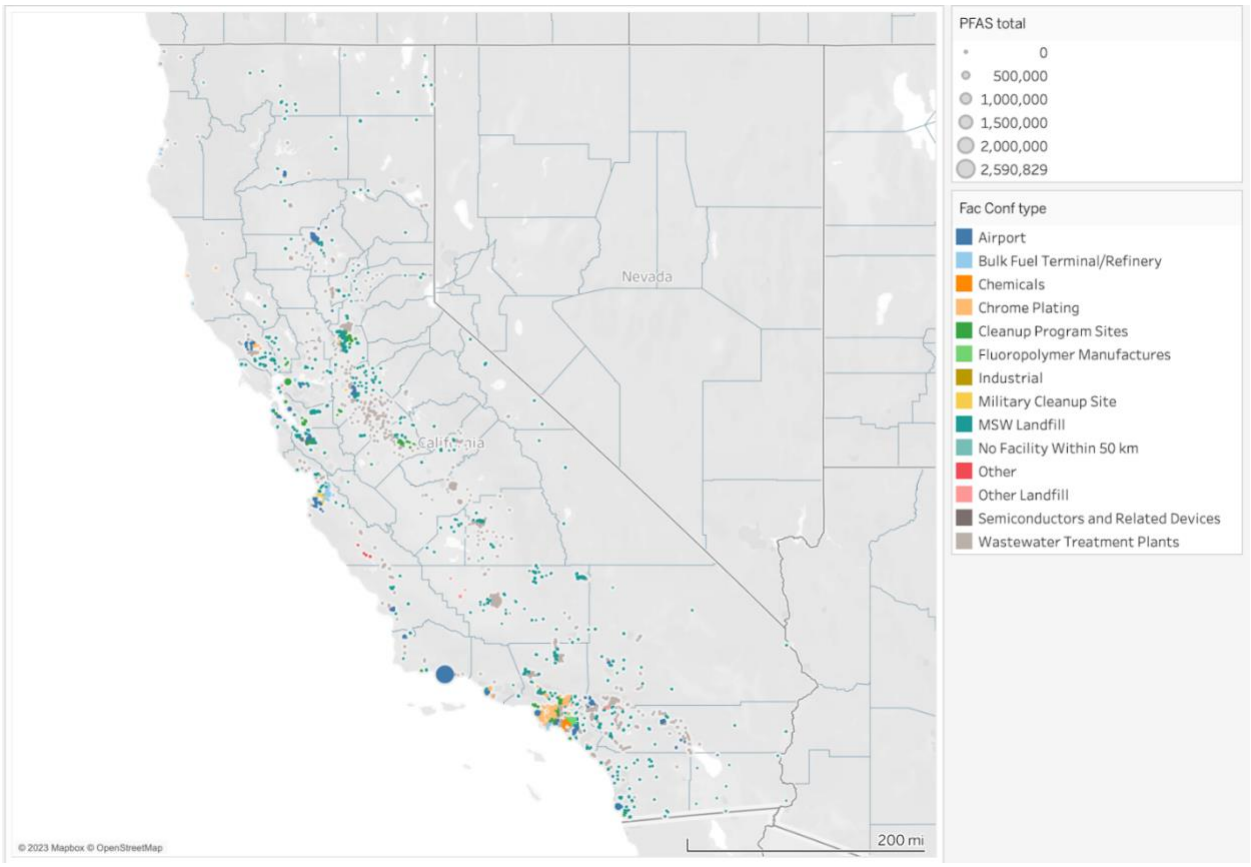

Figure 3.26. Total PFAS pollutant hotspots in California by different facilities

Figure 3.26 shows the distribution of every facility in this study. The most widely distributed facility types are cleanup program sites and MSW landfills. The graph also demonstrates that the highest concentration of PFAS contamination is from the airport in southern California, near Santa Barbara. The amount of facilities is the most in the Greater Los Angeles area. These hotspot maps include short-chain and long-chain PFCA and PFSA, as well as the most commonly studied 4 PFAS: PFOA, PFOA, PFHXSA, and PFNA[70].

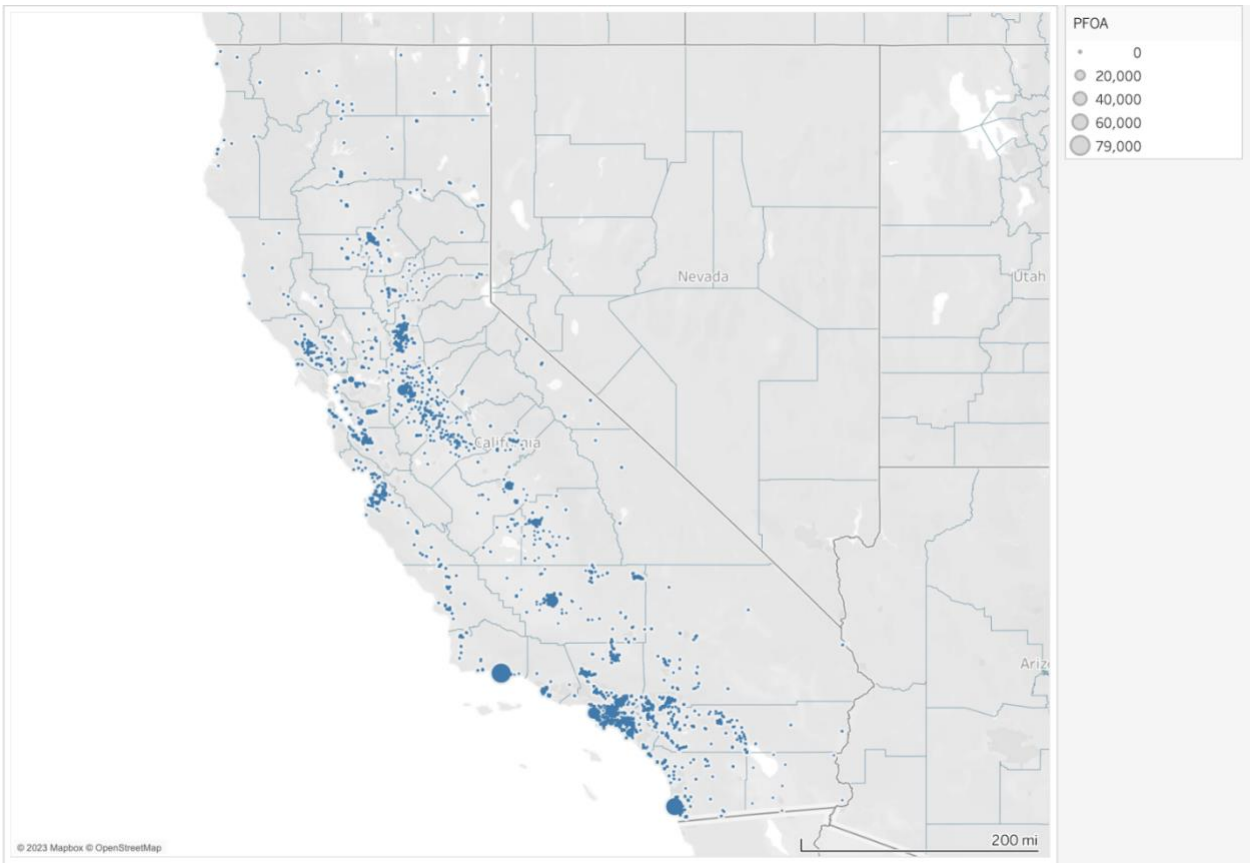

Figure 3.27. PFOA hotspots map of California

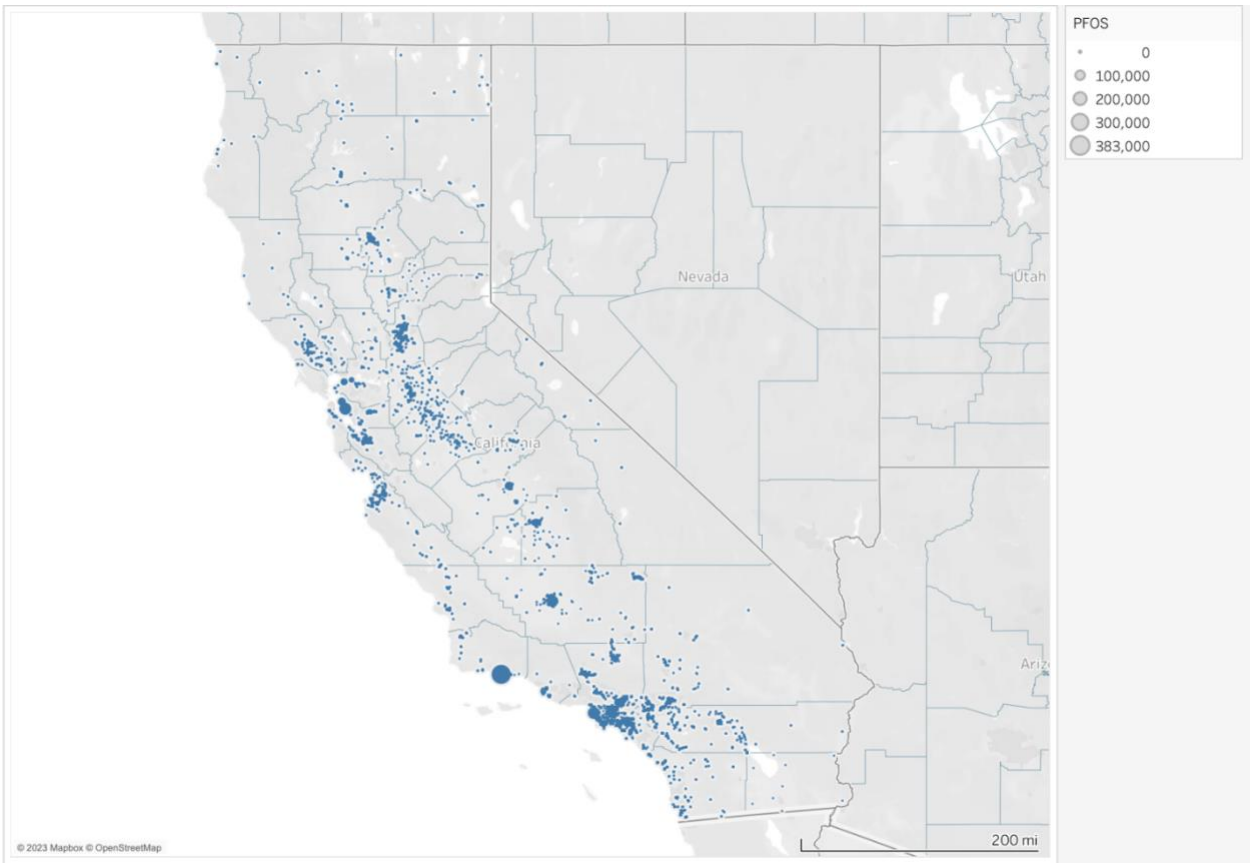

Figure 3.28. PFOS hotspots map of California

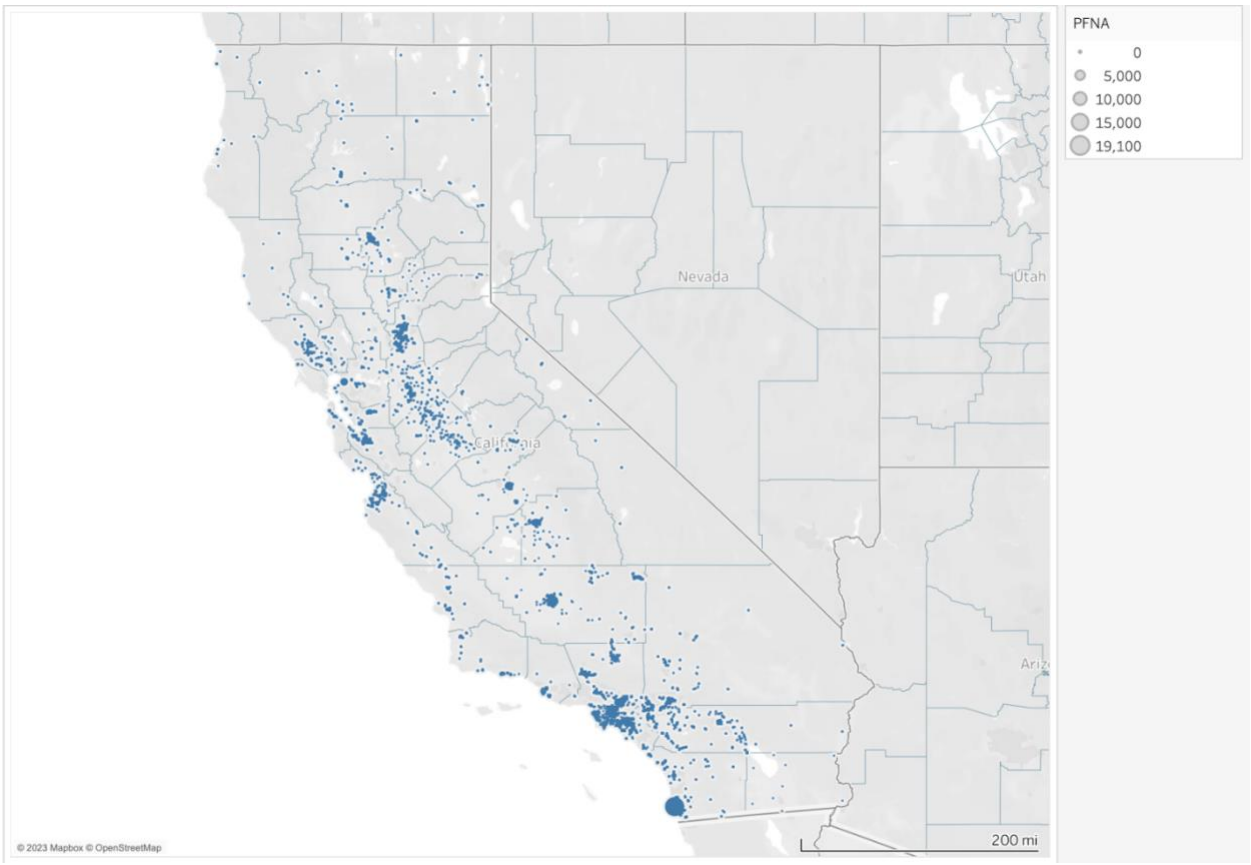

Figure 3.29. PFNA hotspots map of California

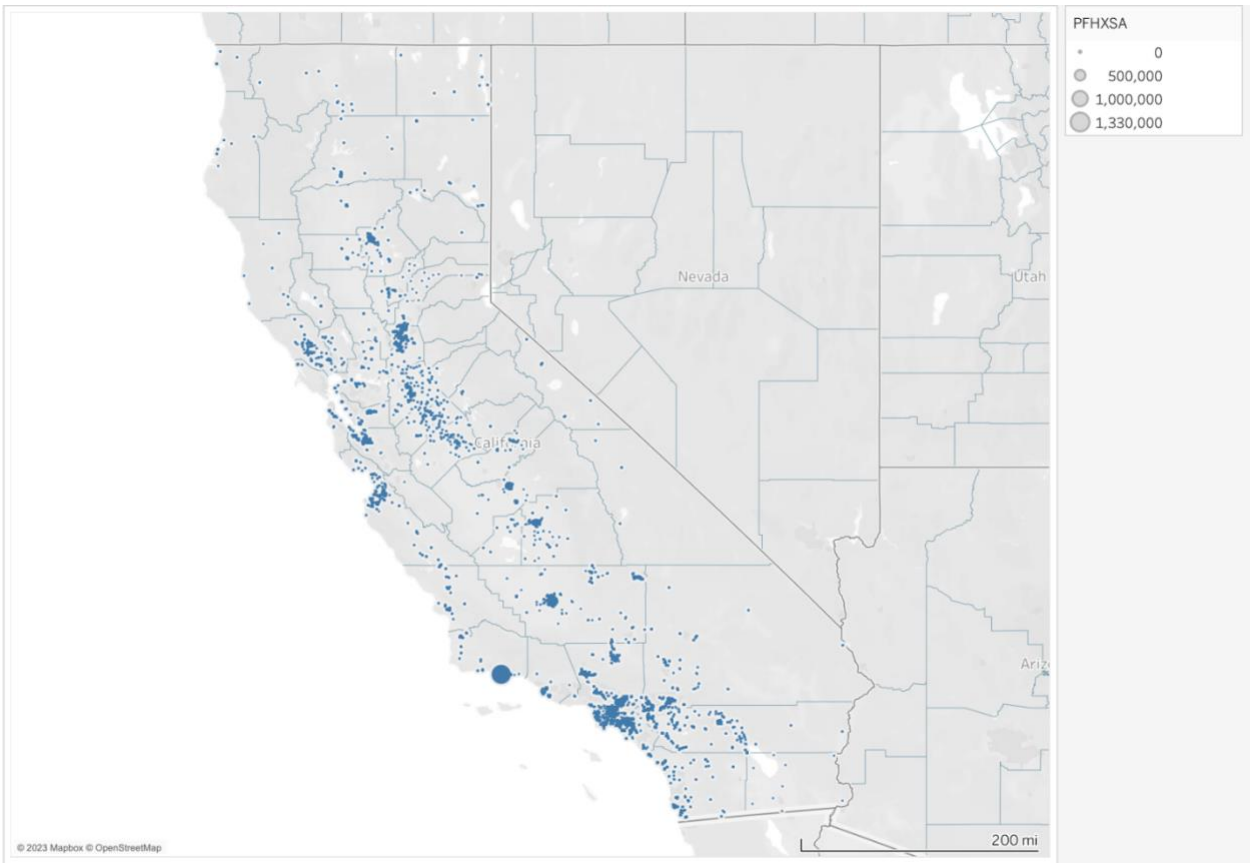

Figure 3.30. PFHXSA hotspots map of California

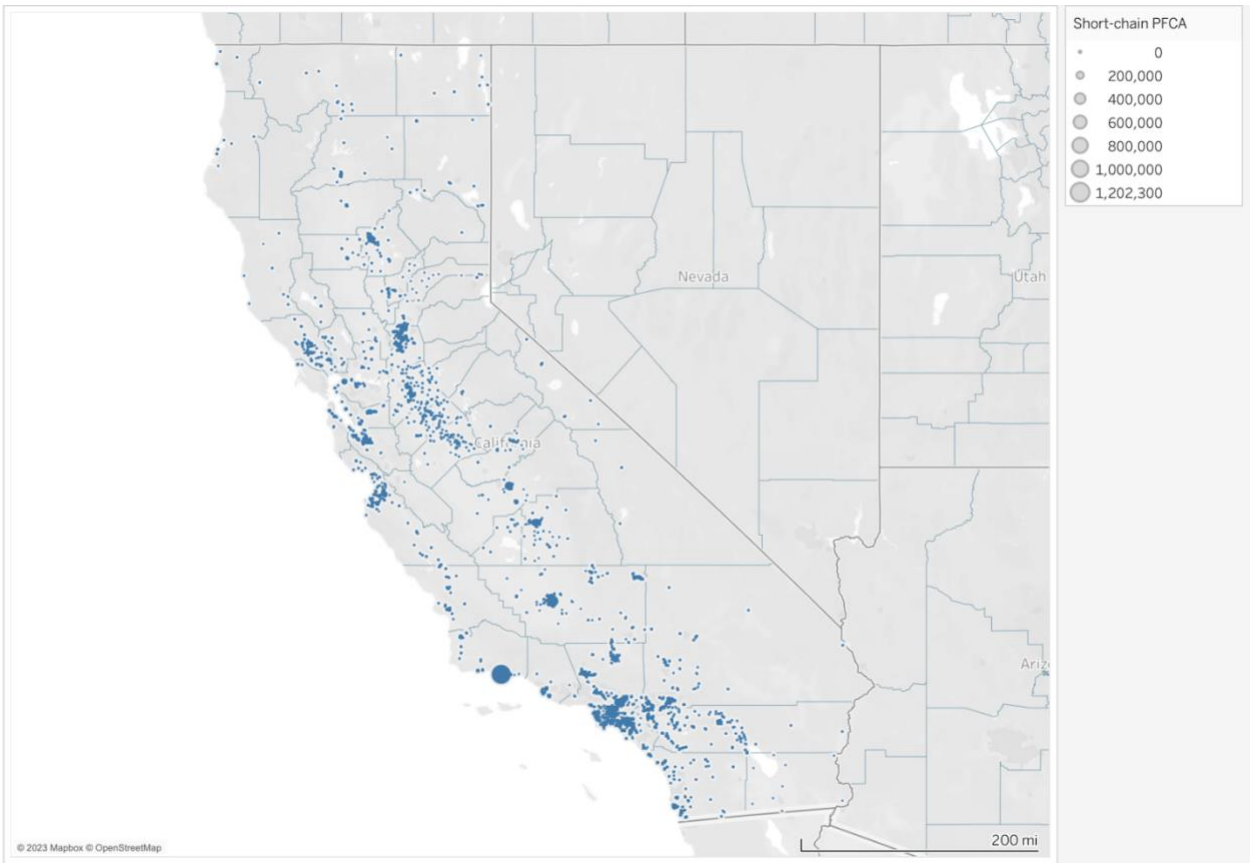

Figure 3.31. Short-chain PFCA hotspots map of California

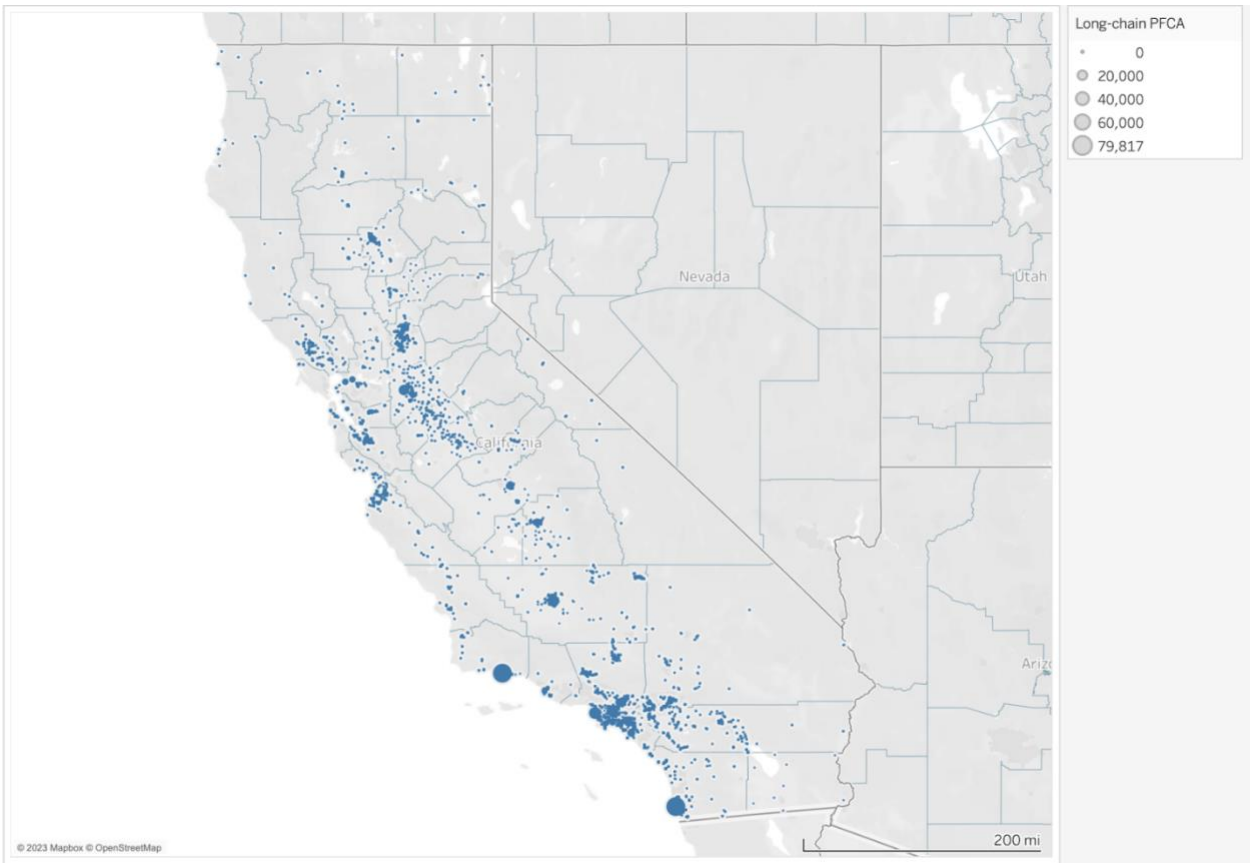

Figure 3.32. Long-chain PFCA hotspots map of California

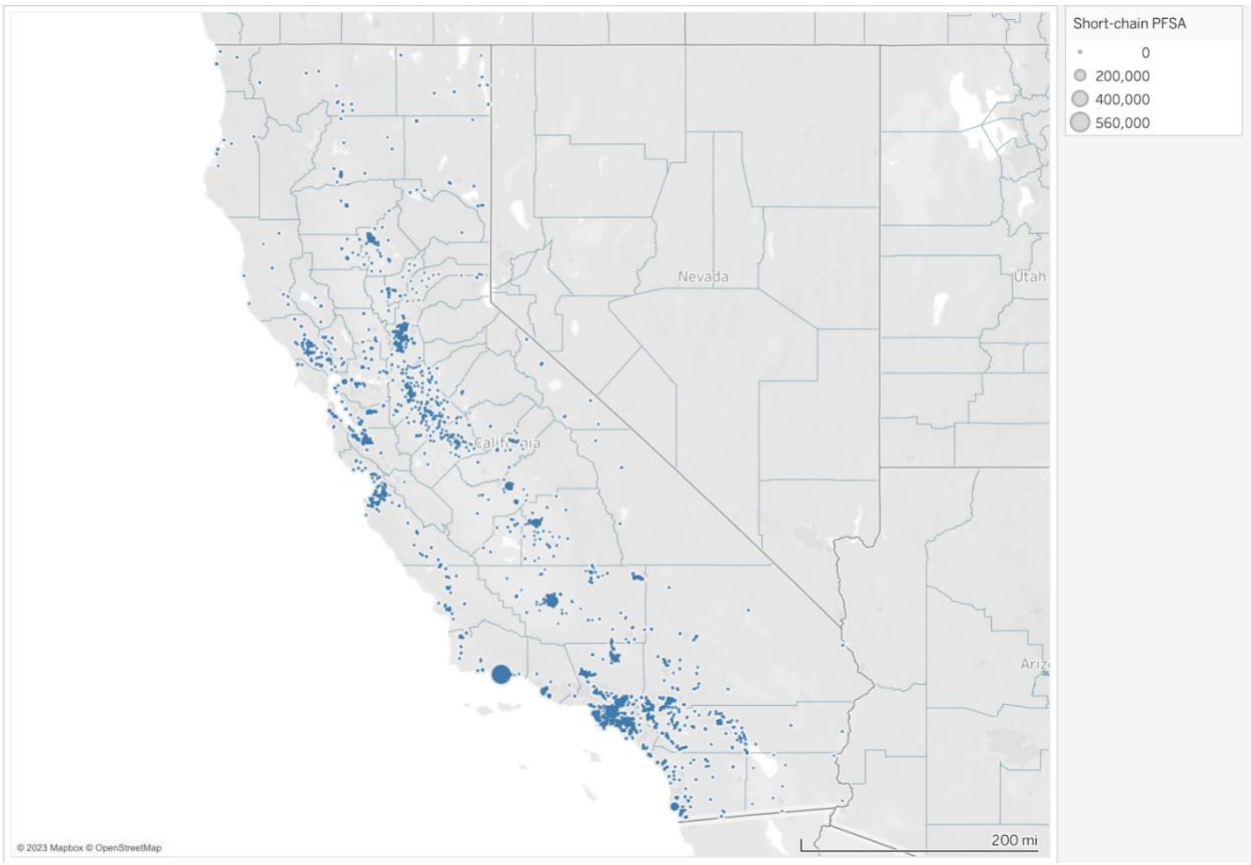

Figure 3.33. Short-chain PFSA hotspots map of California

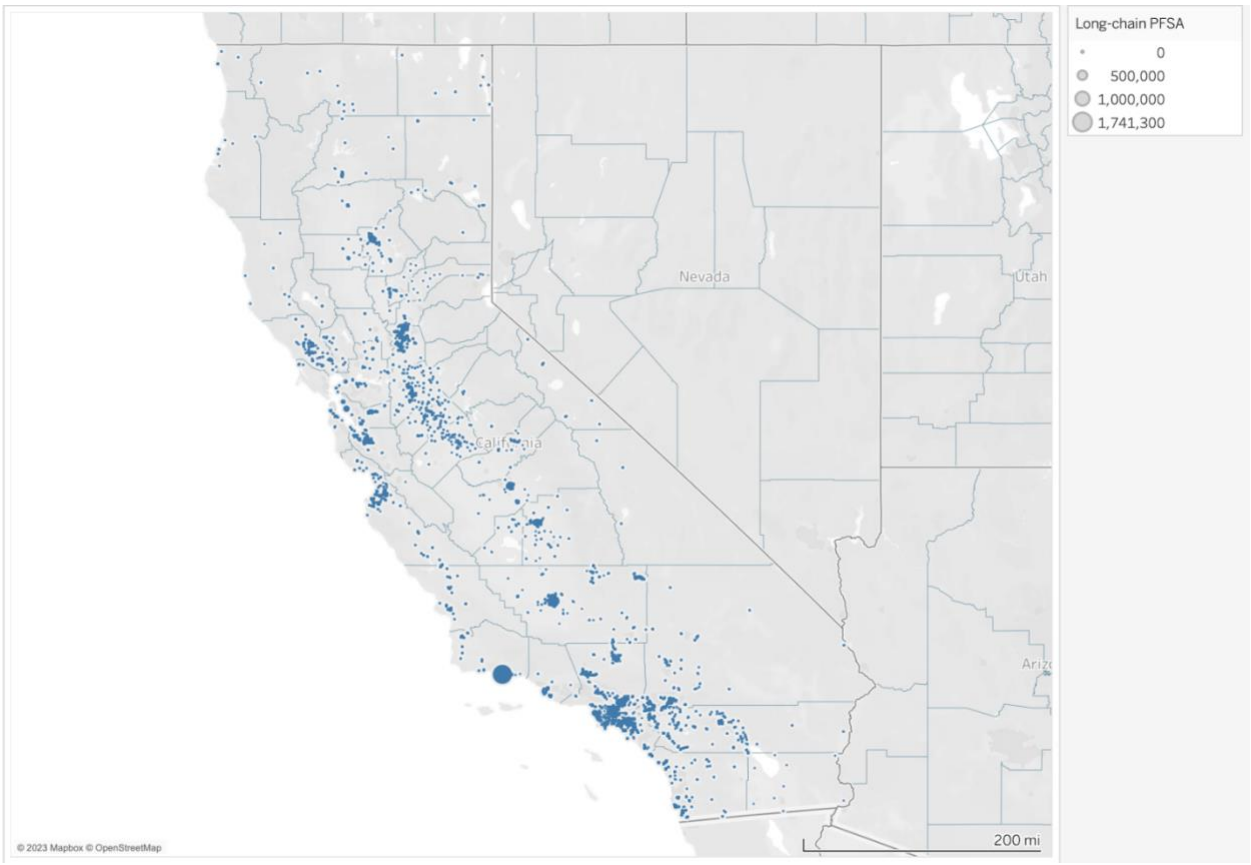

Figure 3.34. Long-chain PFSA hotspots map of California

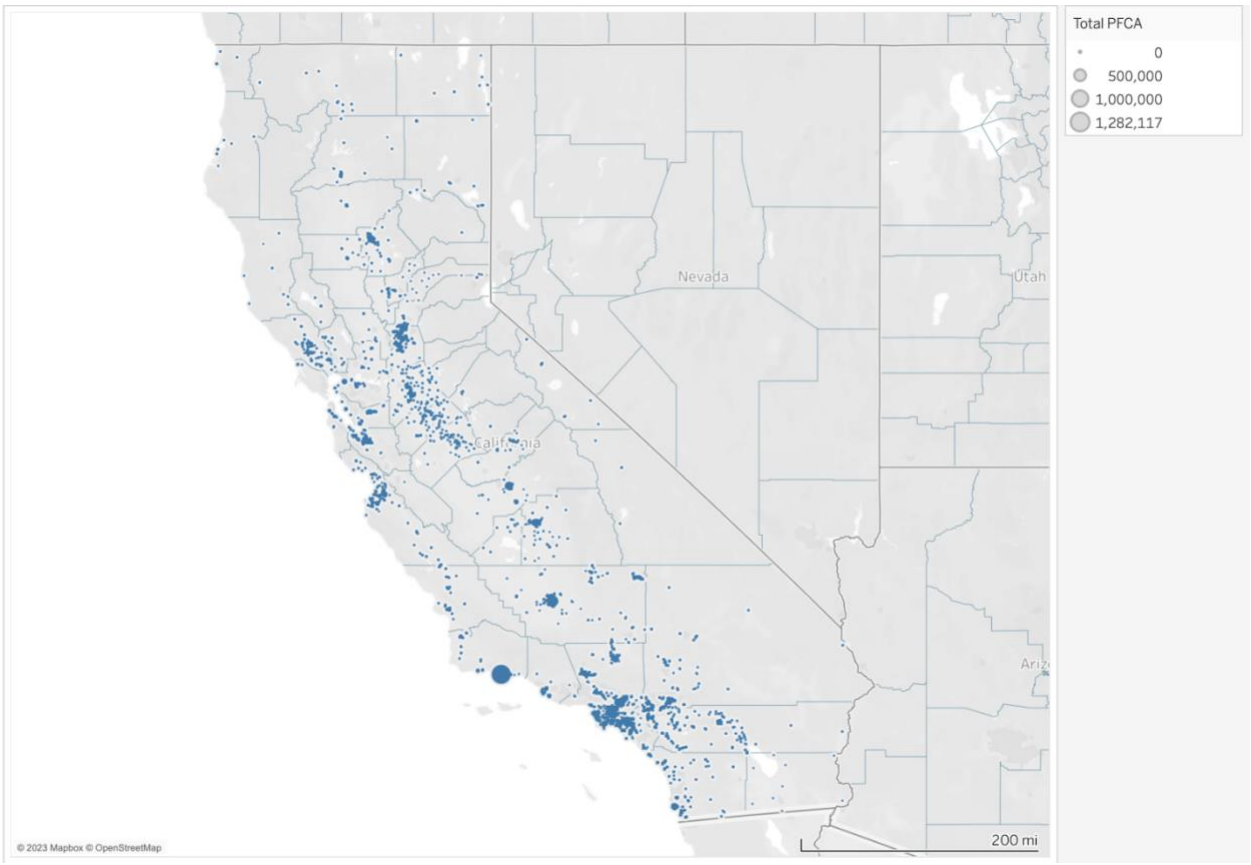

Figure 3.35. Total PFCA hotspots map of California

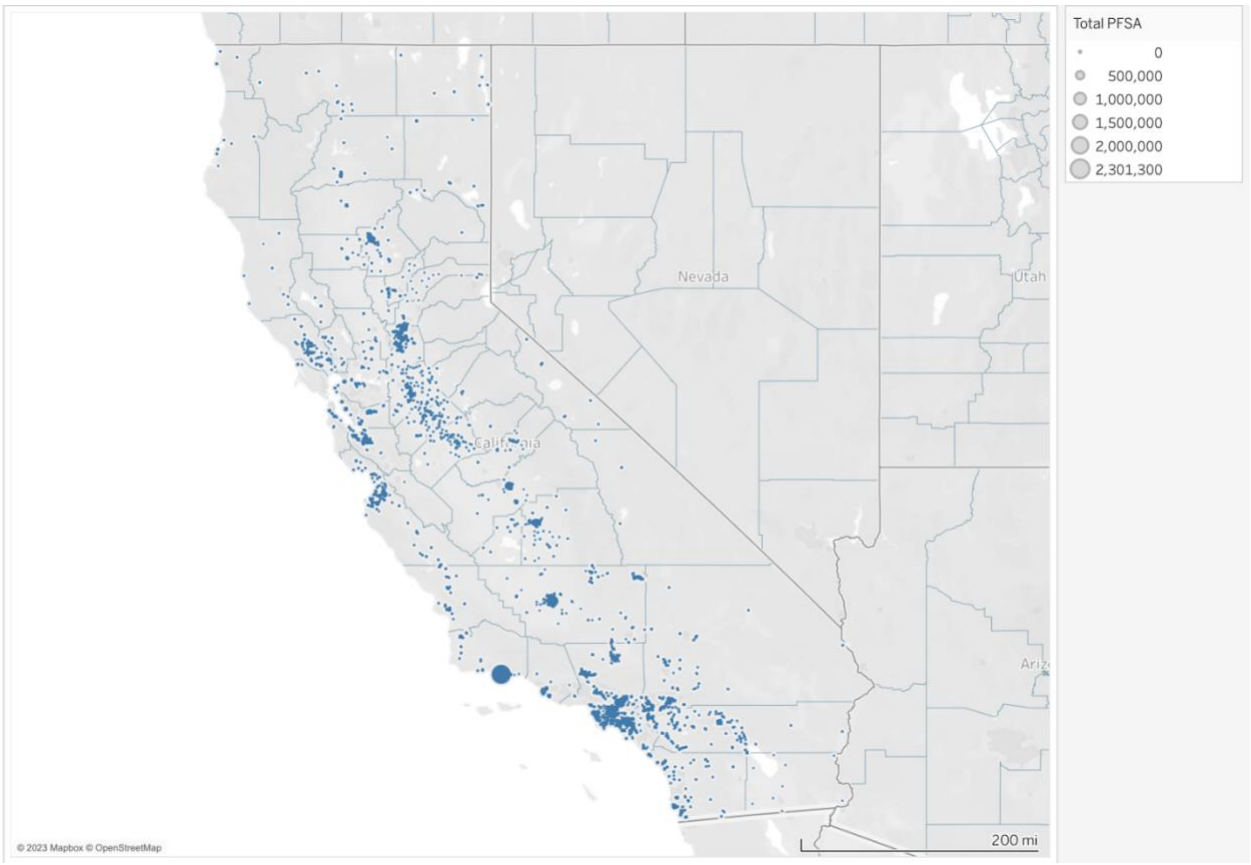

Figure 3.36. Total PFSA hotspots map of California

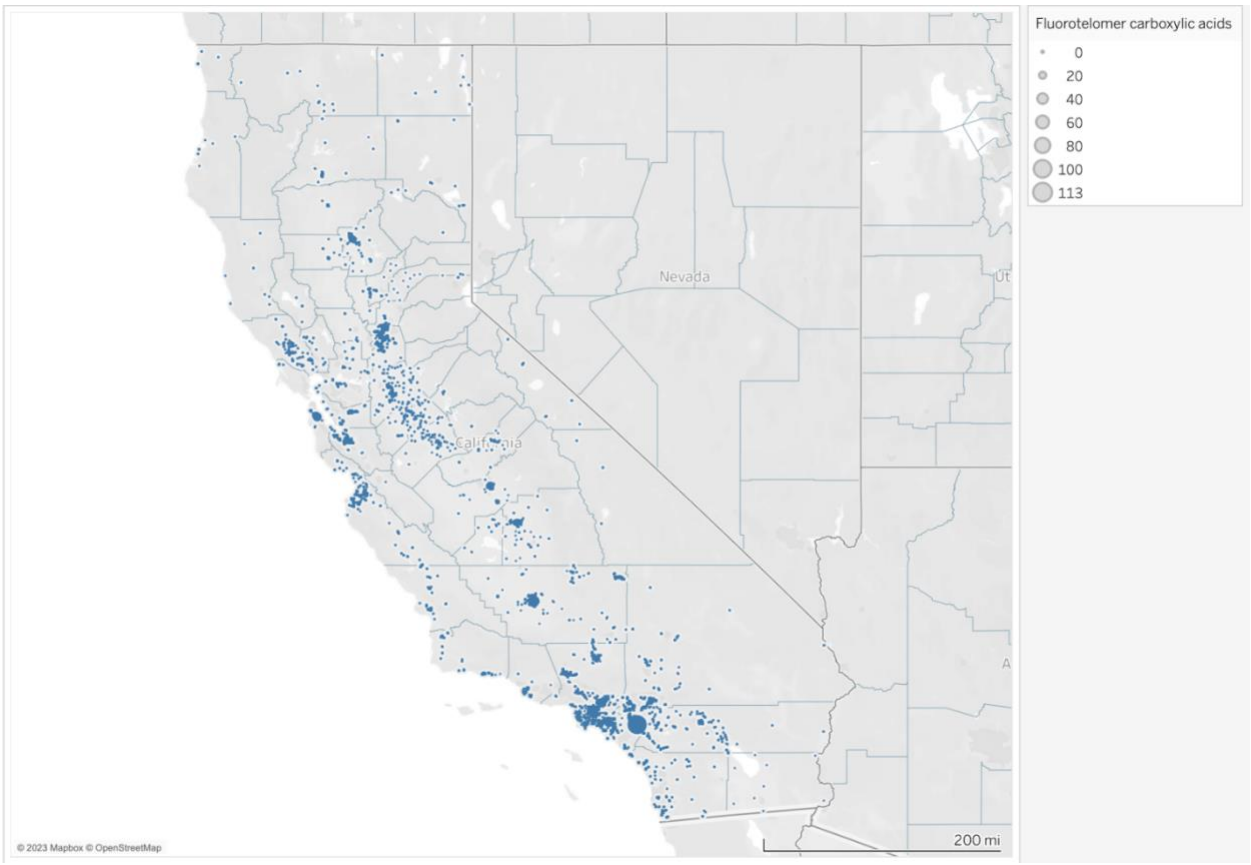

Figure 3.37. Fluorotelomer carboxylic acids hotspots map of California

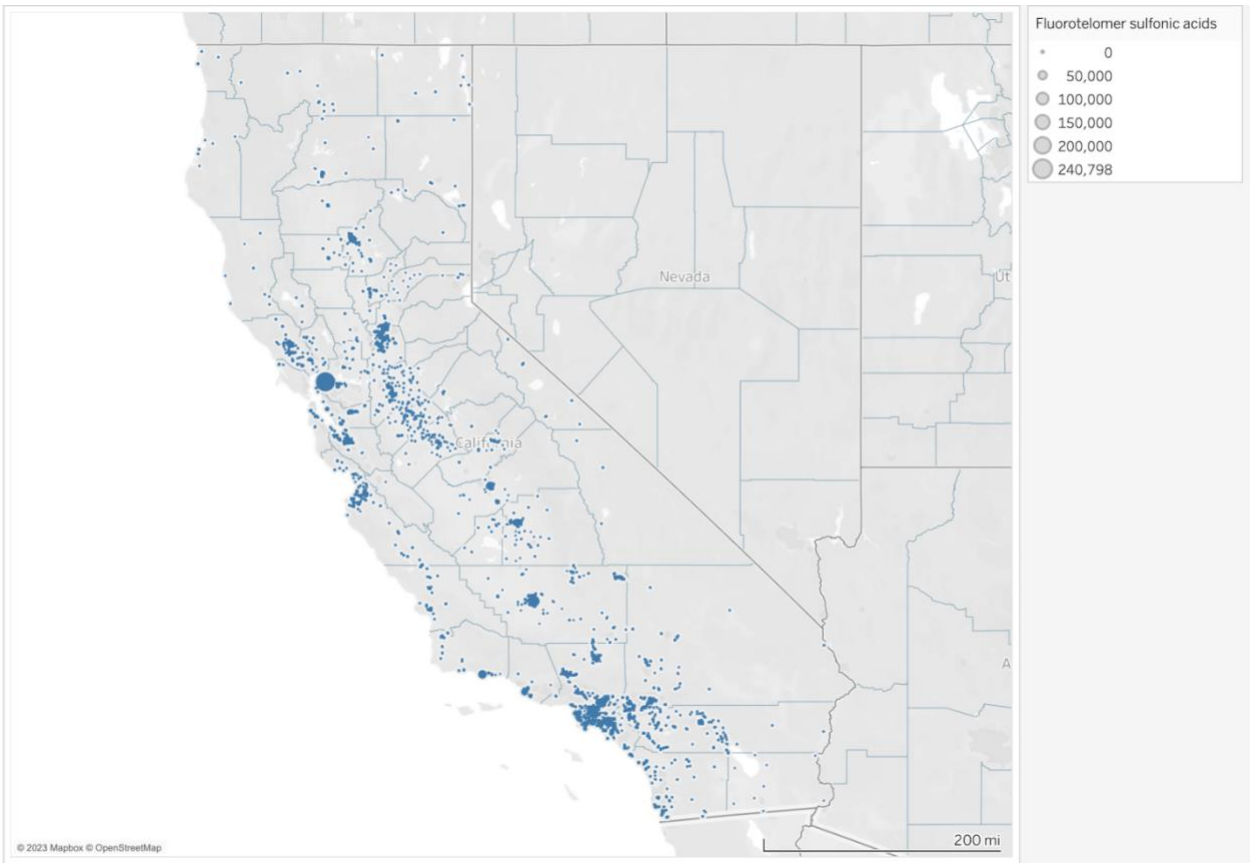

Figure 3.38. Fluorotelomer sulfonic acids hotspots map of California

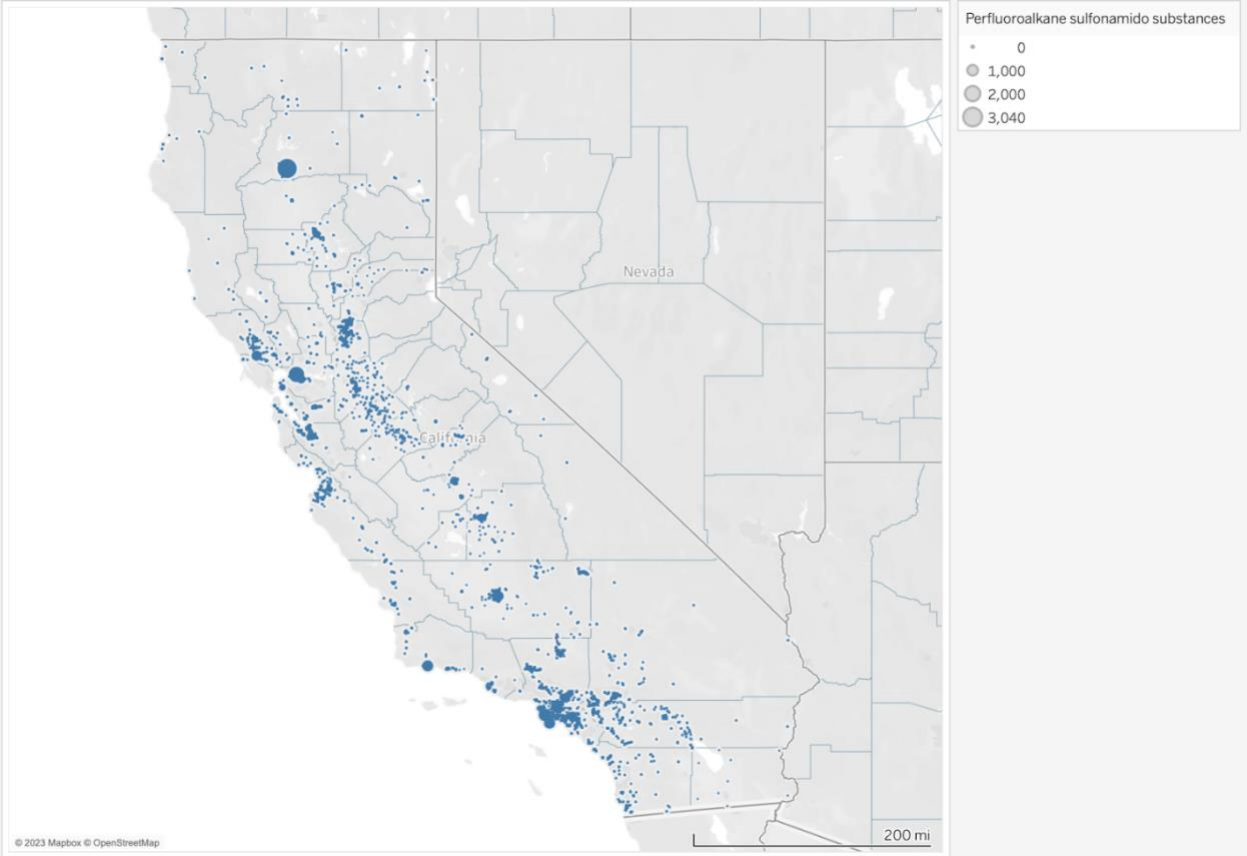

Figure 3.39. Perfluoroalkane sulfonamido substances hotspots map of California

All the hotspot maps show that the high-impact area of PFAS contamination is the Greater Los Angeles Area. Additionally, Sacramento, San Francisco, and Santa Barbara are also gathering places with high concentrations of PFAS. PFOA, PFNA, besides, long-chain PFCA also have a hotspot in San Diego County.

4. Conclusion and recommendations

This research adopts different statistical and spatial analysis approaches to predict and analyze the distribution and the source of PFAS in California groundwater. The total amount of PFAS studied in this research is 38 and has been categorized into 11 groups. The total quantity of facility types is 14. In summary, the significant PFAS of each facility was listed in the tables. Airports were identified as the most significant source of groundwater PFAS pollution affecting California, and cleanup program sites and MSW landfills were recognized as the most extensively spread contamination source of California groundwater PFAS. According to previous studies, firefighting training and emergency fire suppression are the most common paths that AFFF uses in airports[71]. As reported by the researchers who studied Australian surface water samples influenced by the airports, there are also high concentrations of 6:2 FTS and PFPA[72]. 6:2 FTS has also been proven to be directly related to AFFF use[72,73], so in my results, the PFPA, 6:2FTS, and PFPES occupy the top three highest PFAS in California groundwater. The usage of AFFF in airports influenced most California PFAS contamination. The issue of PFAS contamination not only occurs in California but is also a global environmental issue that needs to be resolved.

Meanwhile, over an extended period of the application of PFAS in various industrial and commercial uses, the contamination concentration remains a colossal base. Due to the straitened circumstances of the PFAS degradation, it will need to take effort to eliminate. Most importantly, groundwater is a prominent drinking water source[73]and intensified attention should be given to groundwater PFAS contamination. Identifying the type of PFAS of various facilities would also be advantageous for target PFAS removal. In future studies, groundwater contamination should be recommended to investigate to determine the PFAS composition of facilities in a broader

study area. Furthermore, with the increased attention to groundwater PFAS contamination, the PFAS in drinking water can be studied and removed at the source.

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