Linking Industrial Point Sources to PFAS Contamination in Wells: Michigan Case Study

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Abstract

In recent years, concern has grown over widespread environmental contamination of per- and polyfluoroalkyl substances (PFAS) in the environment and especially in drinking water. These manmade chemicals were first synthesized in the mid-20th century and contain thousands of individual species, some of which are still used today. A wide variety of industries use or have historically used PFAS in their products and processes. PFAS chemicals are recalcitrant by design and thus do not break down in the environment. Federal and state agencies have begun testing of drinking water and other media for PFAS in order to understand the extent of contamination and determine any necessary regulatory actions. Testing is expensive, however, and it would be beneficial to be able to prioritize wells for testing based on their likely vulnerability. This study presents a methodology for identifying locations of industrial sites likely to use PFAS, and uses Michigan's robust well testing data alongside industry and environmental factors to identify any spatial relationships that can be used to help prioritize testing of wells.

Introduction

Per- and polyfluorinated alkyl substances, or PFAS, are contaminants of emerging concern due to their widespread occurrence and recalcitrance in the environment, their tendency to bioaccumulate, and the wide variety of health effects associated with exposure at low concentrations (Lindstrom et al., 2011b). PFAS are known to enter the environment from both industrial and non-industrial waste dumping and effluent, airborne deposition via fumes, firefighting foam use, and disposal of PFAS-containing products in landfills where leaching can occur (Liu et al., 2017; Wang et al., 2014). They have been found extensively in both surface and groundwater drinking water sources at concentrations ranging from non-detectable to hundreds of parts per trillion (ppt) (Boone et al., 2019). Substantial difficulty exists in predicting where high levels of contamination will occur and the causal sources. Identifying PFAS contamination in groundwater is complicated by the wide variety of PFAS compounds in use, the lack of transparency and the variability in their use across multiple products, manufacturing processes, and industries. Although they are used in a diverse spectrum of consumer products and industrial processes, PFAS compounds are often only used in small amounts. Finally, lacking government regulation of PFAS chemicals results in limited reporting of use of these persistent chemicals and thus also results in difficulty funding and targeting monitoring PFAS contamination in the environment (KEMI, 2015).

Studies have shown that exposure to PFAS is associated with a variety of adverse health outcomes in humans, including hypertension, low birth weight, liver problems, thyroid problems, decreased vaccine response in children, and higher incidence of several kinds of cancer (Agency for Toxic Substances and Disease Registry (ATSDR), 2018; Grandjean et al., 2012; Granum et al., 2013; Kim et al., 2018; Nian et al., 2019; X. W. Zeng et al., 2019; Z. Zeng et al., 2019). Although concerns about PFAS chemicals have existed for over a decade, they have only recently become subject to regulation and these regulations are limited in the scope of the types of PFAS they address. In 2009, PFOS was listed as a persistent organic pollutant under the Stockholm Convention, and PFOA was added to the list in 2019 (Stockholm Convention, 2019, 2008). In 2019, USEPA issued interim recommendations for groundwater contaminated

with PFOA and PFOS and is in early stage of risk assessment for both compounds in biosolids (EPA, Dec 2019).

Lack of information on industrial PFAS use results in the inability of groundwater managers and regulators to identify potential contamination sites efficiently or at a landscape scale for testing and monitoring. A few prior studies have looked at contamination sources of PFAS on various scales. Using data from USEPA's Third Unregulated Contaminant Monitoring Rule (UCMR 3), Hu et al (2016) looked for watershed-level correlation between PFAS contamination and the presence of wastewater treatment plants, military fire training areas, and fluoropolymer facilities that participated in the USEPA's stewardship program to phase out PFAS production (Hu et al., 2016). Associations were found between all types of sites and elevated PFAS concentrations in water. Guelfo et al (2018) created risk ranking for groundwater in the Providence, RI area based on industrial presence and aquifer vulnerability, listing Department of Defense sites, fluoropolymer manufacturers, landfills, and airports among the types of industrial sites contributing the most PFAS risk to water (Guelfo et al., 2018).

In this study we examined whether existing data about the location of industries known to use PFAS compounds can be used to predict future sites for monitoring. This differs from previous studies by incorporating all industry types found in the literature to use PFAS rather than just PFAS manufacturers, and incorporating extensive well testing data to spatially compare PFAS detections to possible point sources. Michigan was chosen as a study case as it began extensive statewide testing of drinking water wells for PFAS in 2018, and continued in 2019 with quarterly testing of wells that had tested at concentrations above 10 ppt the previous year (State of Michigan, 2018). Few states have completed such widespread testing of drinking water sources. Nationwide testing under UCMR 3 has higher detection limits and covers around 5000 drinking water sources nationwide as opposed to over 1000 in Michigan from state-funded testing alone (USEPA, 2012). The goal of this study is to identify characteristics of wells, including proximity to industrial sites, type of bedrock, and well depth, that significantly impact the likelihood of PFAS contamination in order to help focus testing efforts on wells that share these characteristics in other areas.

Methods

1.1 Michigan well and industry data

Well testing data in Michigan was gathered from the Department of Environment, Great

Lakes, and Energy's (EGLE) report on PFAS testing using a Freedom of Information Act (FOIA) request submitted in February of 2020. This data identified wells based on their Water Supply Serial Number (WSSN) and did not include locations. This well test data was merged with the spatially explicit well data from Michigan's GIS portal that also identified wells using WSSNs and could thus be merged with the well testing data (Michigan Dept. of Environment, Great Lakes, 2019). The PFAS testing data from Michigan included 1126 sites, of which 1087 were included in this analysis after removal of surface water sources.

The majority of wells were sampled between April and December of 2018, with one well sampled in September 2017 and seven wells sampled from January through September of 2019. Well water was tested using EPA's Method 537 Revised 1.1, which tests for 18 PFAS compounds. Concentrations were reported as a sum totals of PFOA and PFOS, and of all PFAS found; this study used the concentrations that included all 18 compounds. Among the well sites included in this study, 107 tested positive for PFAS with a range of total PFAS concentrations from 2 ppt to 1828 ppt and an average concentration of 28.794 ppt, whereas 974 sites did not contain PFAS at detectable levels. Notably, the site with a concentration of 1828 ppt was an outlier, as the next highest concentration was 72 ppt.

Industries that use or are sources of PFAS were identified in a literature search. Prior studies on point sources as well as studies analyzing products that contain PFAS led to the inclusion of the industry groups listed in Table 1 with their relevant NAICS codes and studies that indicated their PFAS use. Briefly, PFAS have a wide variety of industrial uses including nonstick coatings, metal finishing, fabric finishing, plastics manufacturing, lubricants, and pesticides (Buck et al., 2011; Glüge et al., 2020; Guelfo et al. 2018; Herzke et al. 2012; Kissa, 2001; Nascimento et al., 2018). Studies have also indicated PFAS use in medical devices, electronics, cleaning products, and photographic film processes (Clara et al., 2008; Gosetti et al., 2018; Lin et al., 2009; Xie et al., 2013). Studies indicating PFAS contamination at military sites and landfills are also well documented (Guelfo et al., 2018; Hu et al., 2016; Liu et al., 2017; Oliaei et al., 2013). Industrial sites for Michigan were spatially located using the NAICS codes listed in Tables 1 and 2, and the USEPA's Enforcement and Compliance History Online (ECHO) facility search (USEPA, 2018). Sites include both current and historic facilities as of 8 June, 2020. Industrial and well locations were mapped using a geographic information system (GIS), ESRI ArcMap version 10.7 (Redlands, CA, USA).

1.2 Linking industrial sites to PFAS contamination

Previous studies have shown that a 0.5 km radius buffer around a groundwater source and potential contaminants is a sufficiently sized area to study significant correlation between land use and contamination. Johnson and Belitz (2009) found that a 500m circle, though a highly simplified model, was sufficient to show correlation between volatile organic compound (VOC) contamination of groundwater and urban land use (Johnson and Belitz, 2009). Lindstrom et al. (2011a) found that 82% of wells within 0.5 km of fields that received contaminated biosolids tested positive for PFAS (Lindstrom et al., 2011a). Finally, impervious surfaces are known to cause rapid runoff that carries heavy loads of pollutants into waterways, which can ultimately impact groundwater sources (Frazer, 2005). Use of PFAS at airports and other places where AFFF foams may be used in paved areas could contribute to pollution from such runoff (Awad et al, 2011). One study has linked landfill, military, and PFAS-producing facilities to drinking water contamination on a hydrologic unit basis using UCMR3 testing data, but did not include industries that use PFAS or link specific sites to contamination (Hu et al. 2016). Based on these studies, we hypothesized that location within the 0.1 km, 0.5 km, and 1 km buffer zones of PFAS-related industrial sites, Coldwater Shale bedrock type, well-drained soils, and impervious land cover would show a significant positive correlation with PFAS detection in wells, whereas well depth would show a significant

negative correlation with PFAS detections. Coldwater Shale bedrock type was chosen because it was the most common bedrock type at contaminated well sites (35%). Based on previous literature and the prevalence of industries within the buffer zones (as indicated in Table 2), we also hypothesized specifically that the number and percentage of current and historic electroplating facilities, airports, and landfills within the buffer zones would be positively correlated with PFAS detections in wells (Awad et al., 2011; Hepburn et al., 2019; Milley et al., 2018). To test for effects of industry presence on PFAS well contamination in Michigan, we used a maximum likelihood method, Probit analysis and Moran's I as described below.

1.2.1 Probit Models

Using a Probit function, the spatial siting of industries and the bedrock characteristics were used to predict the probability of a positive well test for PFAS. To test for a relationship between industry presence on PFAS detections in wells, buffer zones of 0.1 km, 0.5 km, and 1 km were constructed around each well tested for PFAS. In this study, industry presence within these buffer zones was analyzed as an effect on detection and non-detection in wells while controlling for other factors such as bedrock type, land cover, and well depth. Wells were given a value of 0 if PFAS were not detected (ND), and given a value of 1 if any level of PFAS were detected. A value of 1 was given for wells located within the Coldwater Shale bedrock formation, and a value of 0 given for wells within all other bedrock formation types. Coldwater Shale was selected as it is the most common bedrock formation for wells with detected levels of PFAS. Well depth was based on the numerical value of the well's depth, in feet, and well depth squared is the squared value of the depth of each well in feet squared. A value of 1 was given for wells with a "developed, low intensity" land cover classification (NLCD code 22), and a value of 0 given for wells with any other land cover classification. A value of 1 was given for wells with "well drained", "excessively drained", or "somewhat excessively drained" soil drainage classifications. A value of 0 was given for wells with any other soil drainage classification. A few specific industries were chosen as separate variables because they were present at the highest numbers within the well buffers (see Table 2): landfill, electroplating, metal foil manufacture, and airport sites. All models were estimated as either percent, meaning the total percentage of sites within the given radius (0.5 km or 1 km) of a well that belong to the specific industry, or raw number, which is the number of industry sites classified as the specific industry within the given radius of a well. The percentage of "all others", which includes all the industry sites included in the analysis apart from the specified industries just outlined, was also estimated in each model. Lastly, a variable was assigned for the total number of quarters within the past three years that industry sites in a given radius had failed to comply with EPA environmental standards, with a maximum value of 12. A summary of the variables used in the Probit models is shown in Table 1.

Four Probit models were considered to test the effect of land use and type on the probability of detection, such as the number and types of industries within a certain buffer of the well, bedrock, land cover, soil type, well depth, and well depth squared on the probability of PFAS detection in a well using the equation as follows

$$0,1 = f[\#sites, depth, percentindustry, percentmajor facility, \#violations] \quad (1)$$

where the dependent variable is assigned values of 0 or 1. The four models separate the amount/percent and type of industries found within each buffer from the data found within other buffers. In this study, the detection of PFAS pollution at well sites is the dependent variable where a value of 1 indicates any detected level of PFAS pollution, otherwise the variable takes a value of 0. The limit of detection was 2 or 4 ppt depending on the analyte (AECOM 2018). Several independent variables were included in the Probit regression model: number of industrial sites (#sites) within each spatial buffer of the wells (0.1 km, 0.5 km, 1 km), well depth in feet (depth), the percentage of sites within 1 km of specific industry types thought to be more likely to contribute to contamination (airports, landfills, and electroplaters) (percentindustry), and the total number of quarters in which an industrial site had USEPA violations in the past three years^[1] (#violations). The Probit regression model in Equation 1 was analyzed using IBM SPSS Statistics 26, with statistically significant variables having a p -value no greater than 0.10. Results are in sections 2.3.1 and 2.3.2 (IBM Corp., 2019).

1.2.2 Cluster analysis

A negative Local Moran's I Index was performed to identify any spatial correlation between detections of PFAS in wells in relation to each other, separate from industry presence. This analysis indicates spatial outliers, where the data at one location is independent of the data at nearby locations (Anselin, 1995). In this study, spatial outliers indicate that the level of PFAS pollution at one well is independent of the level of PFAS pollution at nearby wells. A positive Local Moran's I Index value indicates spatial clustering, where the data at one location is similar to the data at nearby locations. In this study, spatial clusters indicate that the level of PFAS pollution at one well significantly resembles the levels at nearby wells. Spatial clustering defined by high-high (high values near other high values) and low-low (low values near other high values) clusters. High-high PFAS clusters indicate potential hotspots of PFAS pollution, whereas low-low clusters indicate areas of low pollution levels. Spatial outliers are represented by low-high (low value at one location near high values at nearby locations) and high-low outliers (high value at one location near low values at nearby locations). Local Moran's I results are significant in this study if the p -value is no greater than 0.05. The Index values I_i range from -1 to +1 and are determined using the following equations:

$$I_i = \frac{x_i - \bar{X}}{S_i^2} \sum_{j=1, j \neq i}^n w_{i,j} (x_j - \bar{X}) \quad (2)$$

where x_i is the level of PFAS pollution at well site i , $\bar{X}$ is the mean PFAS level across all wells, and w_{ij} is the spatial weight between well sites i and j , and

$$S_i^2 = \frac{\sum_{j=1, j \neq i}^n (x_j - \bar{X})^2}{n-1} \quad (3)$$

where n is the total number of well sites with detected levels of PFAS. An index value of -1 indicates negative correlation between positive PFAS detections or an outlier, a value of 0 indicates no correlation, and a value of +1 indicates a positive correlation or cluster.

Results And Discussion

2.1 Industrial and well sites

Using the NAICS codes listed in Table 3, 2696 relevant industry sites were identified in Michigan. Figure 1 shows the number of industrial sites present in each county. Among the well sites included in this study, 107 tested positive for PFAS with an average concentration of 28.794 ppt, whereas 974 sites did not contain PFAS at detectable levels. No identified industrial sites fell within 0.1 km of a well that tested positive for PFAS, and only two industrial sites fell within 0.1 km of any well identified in this study. A total of eight industrial sites were located within 0.5 km of wells that tested positive for PFAS, three of which were landfills. Other industries within half a kilometer of PFAS wells were metal coating, metal foil, plastics manufacturing, paper mill, and oil and grease manufacturing facilities. Table 3 summarizes the industry sites found within each radius. Twenty-six industrial sites fall within 1 km of wells that tested positive for PFAS; of these sites, eight were landfills, six were metal foil manufacturing, three were airport sites, two were paper mills, two were plastics manufacturing, one electroplating, one metal coating, one commercial printing, one oil and grease manufacturing, and one miscellaneous textiles facility. The totals for 1 km are inclusive of the sites found within 0.5 km.

2.2 Linking industrial sites to PFAS contamination

We hypothesized that the presence of the selected industries would have significant positive correlations with detections of PFAS in wells, as would the depth of the well, and that greater significance would be attributed to facilities belonging to industries already shown to contribute to PFAS contamination (i.e., airports, landfills, electroplaters) and to major facilities. In addition to likelihood of contamination decreasing with well depth, we also hypothesized that as the depth of the well increased, the likelihood of contamination would further decrease, indicated by well depth squared. Because of the lack of industrial sites within 0.1 km of wells, no Probit model was estimated for that buffer, but the model was estimated using both the number of each industry in the buffers as well as the percentage each industry in the buffers for 0.5 km and 1.0 km. It should be noted that in the case of the 0.5 km buffers, electroplating was replaced with metal foil due to limited values for metal foil in the 0.5 km buffers. Omnibus Tests determined that our four Probit models were all significant at greater than 99% confidence. The model results are shown in Tables 4 and 5.

2.2.1 Probit models, 1 km

The 1.0 km buffer models both showed that well depth, well depth squared, and well-drained soil were significantly likely to increase the probability of PFAS detection at greater than 99% confidence level (Table 4). In both cases, well depth positively affected the likelihood of detection, while well depth squared had a negative effect. This indicates that the likelihood of PFAS detection increases with deeper wells, but that the rate of increase declines as depth increases. The positive effect of greater well depth on the probability of PFAS detection runs counter to our hypothesized relationship. One explanation for this could be that wells in vulnerable areas are dug deeper as a preventative measure, but further study is necessary to confirm this. There was a positive relationship between percentage of electroplaters, along with percentage of all other industry and the likelihood of PFAS detection in the 1.0 km model, which corroborates our hypothesis.

The relatively high number of metal foil sites (compared to the other industries included in this study) within 1 km of positive-detect wells is surprising. Although this industry appeared on a list of PFAS-using industries created by the Minnesota Pollution Control Agency (MPCA), it has not yet become an industry of focus across literature examining PFAS pollution. Additional research on metal foil practices could help confirm if this industry contributes significantly to PFAS contamination in wells near facilities.

2.2.2 Probit models, 0.5 km

For both Probit models estimated at the buffer size of 0.5 km, well depth, well depth squared, and well-drained soil showed significant effects on likelihood to detect positive PFAS levels (Table 5). Similar the 1.0 km models, well depth and well-drained soil had a positive effect while well depth squared had a negative effect on the probability of detection. It appears based on these analyses that more permeable soil near a well is more important than presence of specific industry types when predicting PFAS contamination in wells. Despite the higher numerical value of metal foil sites within the 0.5 km buffers compared to other sites, the number and percent metal foil sites did not prove significant in this model. This is likely due to the number of metal foil sites still being low, despite being higher than other industry types. Percent and number of landfill sites were also not significant at this level, along with bedrock type, land cover, and quarters with violations.

2.2.3 Cluster analysis

A local Moran's I test examining clusters and outliers of high versus low concentrations of PFAS within wells that tested positive showed eight low-high outliers, three high-high clusters, and one high-low outlier all concentrated in southwest Michigan. A map of clusters and outliers can be seen in Figure 2. It is interesting to note that the high-high clusters and both types of outliers are predominantly located in Kalamazoo and Allegan counties, both of which have relatively high numbers of industry sites compared to other counties. About half of the industrial sites in these counties were landfills, which corresponds to

the percentage of landfills compared to total industrial sites in the state as a whole. In these two counties, however, only one of the landfills (1.6% of total landfills) was flagged as actively operating, whereas more than 4% of the state's total landfills were labeled active. A higher percentage of inactive landfills may indicate older landfills. Because landfills were not required to be lined until the Resource Conservation and Recovery Act of 1976, older landfills are more likely to cause leaching of contaminants into groundwater (Congress, 1976). Thus, a higher proportion of inactive landfills in Kalamazoo and Allegan counties may explain the clustering found in these counties from the Moran's I test. The presence of outliers, or wells with high concentrations having a significant number of wells with low concentrations nearby and vice versa, could be explained by landfill leachate permeating wells to different degrees. Further study of wells in this region, the groundwater flow near these wells, and their proximity to landfills could help explain the outliers found in this study. In addition to the clusters and outliers in southwest Michigan, ten low-low clusters were identified throughout the state. All other clusters and outliers determined using Local Moran's I have p -values exceeding the 0.05 threshold, and are not statistically significant to include in the study. Table 7 identifies the location of statistically significant Moran's I results, the Index values, and Z-scores.

Conclusions

Overall, this study underscores the difficulty of tying industrial presence to PFAS contamination in drinking water wells as a means of targeting groundwater testing at a landscape scale. Spatially, the Moran's I results pointed to landfills as a possible smoking gun for PFAS detection, due to a high portion of clusters coinciding with counties that host a high number of inactive landfills, but the more detail, site-specific probit model did not show significance between landfill presence and positive PFAS detections in wells. Future research could investigate the relationship between age of landfills and detections of PFAS, as this data set did not include landfill age. Although multiple variables were investigated in our predictive probit models to predict PFAS well contamination, the only factors that proved significant in all models were well depth, well depth squared, and soil type. The positive correlation found between well depth and PFAS detection contradicted our hypothesis, and a possible explanation for this is wells being dug deeper in vulnerable areas. The recalcitrance of PFAS prevents them from breaking down, and this allows them to eventually reach greater depths. It was found in one of the 1.0 km models that the higher the percentage of electroplating facilities within 1 km of the well compared to total industries studied, the more likely a well was contaminated with PFAS. Based on these results, it can be stated that wells constructed in well-drained soil and with electroplating facilities within a kilometer are likelier to be contaminated with PFAS. This information can be useful for identifying wells to prioritize for testing, but additional predictive parameters would be helpful for more accurate targeting. Future research in this area should consider additional parameters that may lead to trends in PFAS well contamination, such as slope, elevation, and groundwater flow direction surrounding wells.

Declarations

Ethical Approval and Consent to Participate

Not applicable.

Consent for Publication

Not Applicable

Availability of Data and Materials

The datasets used and/or analyzed during the current study are available from the corresponding author on reasonable request.

Competing Interests

The authors have no relevant financial or non-financial interests to disclose.

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Author Contributions

KS, RF, RK, MS, and TB contributed to the study conception and design. Material preparation, data collection and analysis were performed by KS, RF and TB. The first draft of the manuscript was written by KS and all authors commented on previous versions of the manuscript. KS, RF, RK, MS, and TB read and approved the final manuscript.

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References

AECOM, 2018. 2018 PFAS Sampling of Public Water Supplies in Michigan, Appendix A: Quality Assurance and Quality Control Summary Analytical.

Agency for Toxic Substances and Disease Registry (ATSDR), 2018. Toxicological Profile for Per- and Polyfluoroalkyl Substances: Draft for public comment.

Anselin, L., 1995. Local Indicators of Spatial Association—LISA. *Geogr. Anal.*

<https://doi.org/10.1111/j.1538-4632.1995.tb00338.x>

Awad, E., Zhang, X., Bhavsar, S.P., Petro, S., Crozier, P.W., Reiner, E.J., Fletcher, R., Tittlemier, S.A., Braekevelt, E., 2011. Long-Term Environmental Fate of Perfluorinated Compounds after Accidental Release at Toronto Airport. *Environ. Sci. Technol.* 45, 8081–8089. <https://doi.org/10.1021/es2001985>

Boone, J.S., Vigo, C., Boone, T., Byrne, C., Ferrario, J., Benson, R., Donohue, J., Simmons, J.E., Kolpin, D.W., Furlong, E.T., Glassmeyer, S.T., 2019. Per- and polyfluoroalkyl substances in source and treated drinking waters of the United States. *Sci. Total Environ.* 653, 359–369. <https://doi.org/10.1016/j.scitotenv.2018.10.245>

Buck, R.C., Franklin, J., Berger, U., Conder, J.M., Cousins, I.T., Voogt, P. de, Jensen, A.A., Kannan, K., Mabury, S.A., van Leeuwen, S.P.J., 2011. Perfluoroalkyl and polyfluoroalkyl substances in the environment: Terminology, classification, and origins. *Integr. Environ. Assess. Manag.* 7, 513–541. <https://doi.org/10.1002/ieam.258>

Clara, M., Scheffknecht, C., Scharf, S., Weiss, S., Gans, O., 2008. Emissions of perfluorinated alkylated substances (PFAS) from point sources - Identification of relevant branches. *Water Sci. Technol.* 58, 59–66. <https://doi.org/10.2166/wst.2008.641>

Congress, U. S. (1976). Resource Conservation and Recovery Act. *Public Law, 94*, 580.

EPA, 2018. Enforcement and Compliance History Online. <https://doi.org/https://echo.epa.gov/>

Frazer, L., 2005. Paving paradise: The peril of impervious surfaces. *Environ. Health Perspect.* <https://doi.org/10.1289/ehp.113-a456>

Glüge, J., Scheringer, M., Cousins, I.T., DeWitt, J.C., Goldenman, G., Herzke, D., Lohmann, R., Ng, C.A., Trier, X., Wang, Z., 2020. An overview of the uses of per- and polyfluoroalkyl substances (PFAS). *Environ. Sci. Process. Impacts.* <https://doi.org/10.1039/d0em00291g>

Gosetti, F., Bolfi, B., Robotti, E., Manfredi, M., Binotti, M., Ferrero, F., Bona, G., Marengo, E., 2018. Study of endocrine disrupting compound release from different medical devices through an on-line SPE UHPLC-MS/MS method. *Anal. Chim. Acta* 1042, 141–154. <https://doi.org/10.1016/J.ACA.2018.07.028>

Grandjean, P., Andersen, E.W., Budtz-Jørgensen, E., Nielsen, F., Mølbak, K.R., Weihe, P., Heilmann, C., 2012. Serum vaccine antibody concentrations in children exposed to perfluorinated compounds. *JAMA - J. Am. Med. Assoc.* 307, 391–397. <https://doi.org/10.1001/jama.2011.2034>

Granum, B., Haug, L.S., Namork, E., Stølevik, S.B., Thomsen, C., Aaberge, I.S., Van Loveren, H., Løvik, M., Nygaard, U.C., 2013. Pre-natal exposure to perfluoroalkyl substances may be associated with altered vaccine antibody levels and immune-related health outcomes in early childhood. *J. Immunotoxicol.* 10, 373–379. <https://doi.org/10.3109/1547691X.2012.755580>

Guelfo, J.L., Marlow, T., Klein, D.M., Savitz, D.A., Frickel, S., Crimi, M., Suuberg, E.M., 2018. Evaluation and management strategies for per-and polyfluoroalkyl substances (PFASs) in drinking water aquifers: Perspectives from impacted U.S. northeast communities. *Environ. Health Perspect.*
<https://doi.org/10.1289/EHP2727>

Hepburn, E., Madden, C., Szabo, D., Coggan, T.L., Clarke, B., Currell, M., 2019. Contamination of groundwater with per- and polyfluoroalkyl substances (PFAS) from legacy landfills in an urban redevelopment precinct. *Environ. Pollut.* 248, 101–113. <https://doi.org/10.1016/j.envpol.2019.02.018>

Herzke, D., Olsson, E., Posner, S., 2012. Perfluoroalkyl and polyfluoroalkyl substances (PFASs) in consumer products in Norway - A pilot study. *Chemosphere* 88, 980–987.
<https://doi.org/10.1016/j.chemosphere.2012.03.035>

Houtz, E.F., Higgins, C.P., Field, J.A., Sedlak, D.L., 2013. Persistence of perfluoroalkyl acid precursors in AFFF-impacted groundwater and soil. *Environ. Sci. Technol.* 47, 8187–8195.
<https://doi.org/10.1021/es4018877>

Hu, X.C., Andrews, D.Q., Lindstrom, A.B., Bruton, T.A., Schaider, L.A., Grandjean, P., Lohmann, R., Carignan, C.C., Blum, A., Balan, S.A., Higgins, C.P., Sunderland, E.M., 2016. Detection of Poly- and Perfluoroalkyl Substances (PFASs) in U.S. Drinking Water Linked to Industrial Sites, Military Fire Training Areas, and Wastewater Treatment Plants. *Environ. Sci. Technol. Lett.* 3, 344–350.
<https://doi.org/10.1021/acs.estlett.6b00260>

IBM Corp., 2019. IBM SPSS Statistics for Windows, Version 26.0. 2019.

Johnson, T.D., Belitz, K., 2009. Assigning land use to supply wells for the statistical characterization of regional groundwater quality: Correlating urban land use and VOC occurrence. *J. Hydrol.* 370, 100–108.
<https://doi.org/10.1016/j.jhydrol.2009.02.056>

KEMI, 2015. Occurrence and use of highly fluorinated substances and alternatives. Report from a government assignment 27–51.

Kim, M.J., Moon, S., Oh, B.-C., Jung, D., Ji, K., Choi, K., Park, Y.J., 2018. Association between perfluoroalkyl substances exposure and thyroid function in adults: A meta-analysis. *PLoS One* 13, e0197244.
<https://doi.org/10.1371/journal.pone.0197244>

Kissa, E., 2001. Fluorinated Surfactants and Repellants, Second Edi. ed. Taylor & Francis, New York.

Kotthoff, M., Müller, J., Jürling, H., Schlummer, M., Fiedler, D., 2015. Perfluoroalkyl and polyfluoroalkyl substances in consumer products. *Environ. Sci. Pollut. Res.* 22, 14546–14559.
<https://doi.org/10.1007/s11356-015-4202-7>

Lin, A.Y.C., Panchangam, S.C., Lo, C.C., 2009. The impact of semiconductor, electronics and optoelectronic industries on downstream perfluorinated chemical contamination in Taiwanese rivers. *Environ. Pollut.*

157, 1365–1372. <https://doi.org/10.1016/j.envpol.2008.11.033>

Lindstrom, A.B., Strynar, M.J., Delinsky, A.D., Nakayama, S.F., McMillan, L., Libelo, E.L., Neill, M., Thomas, L., 2011a. Application of WWTP biosolids and resulting perfluorinated compound contamination of surface and well water in Decatur, Alabama, USA. *Environ. Sci. Technol.* 45, 8015–8021. <https://doi.org/10.1021/es1039425>

Lindstrom, A.B., Strynar, M.J., Libelo, E.L., 2011b. Polyfluorinated compounds: Past, present, and future. *Environ. Sci. Technol.* 45, 7954–7961. <https://doi.org/10.1021/es2011622>

Liu, Z., Lu, Y., Wang, P., Wang, T., Liu, S., Johnson, A.C., Sweetman, A.J., Baninla, Y., 2017. Pollution pathways and release estimation of perfluorooctane sulfonate (PFOS) and perfluorooctanoic acid (PFOA) in central and eastern China. *Sci. Total Environ.* <https://doi.org/10.1016/j.scitotenv.2016.12.085>

Lo, J., Gilljam, fstedt, Leonel, J., Cousins, I.T., Benskin, J.P., 2015. Is Ongoing Sulfluramid Use in South America a Significant Source of Perfluorooctanesulfonate (PFOS)? Production Inventories, Environmental Fate, and Local Occurrence. <https://doi.org/10.1021/acs.est.5b04544>

Michigan Dept. of Environment, Great Lakes, and E., 2019. GIS Open Data, State of Michigan [WWW Document]. URL <https://gis-michigan.opendata.arcgis.com/datasets/egle::water-wells-southwestmichigan>

Milley, S.A., Koch, I., Fortin, P., Archer, J., Reynolds, D., Weber, K.P., 2018. Estimating the number of airports potentially contaminated with perfluoroalkyl and polyfluoroalkyl substances from aqueous film forming foam: A Canadian example. *J. Environ. Manage.* 222, 122–131. <https://doi.org/10.1016/j.jenvman.2018.05.028>

Nascimento, R.A., Nunoo, D.B.O., Bizkarguenaga, E., Schultes, L., Zabaleta, I., Benskin, J.P., Spanó, S., Leonel, J., 2018. Sulfluramid use in Brazilian agriculture: A source of per- and polyfluoroalkyl substances (PFASs) to the environment. *Environ. Pollut.* 242, 1436–1443. <https://doi.org/10.1016/j.envpol.2018.07.122>

Nian, M., Li, Q.Q., Bloom, M., Qian, Z. (Min), Syberg, K.M., Vaughn, M.G., Wang, S.Q., Wei, Q., Zeeshan, M., Gurram, N., Chu, C., Wang, J., Tian, Y.P., Hu, L.W., Liu, K.K., Yang, B.Y., Liu, R.Q., Feng, D., Zeng, X.W., Dong, G.H., 2019. Liver function biomarkers disorder is associated with exposure to perfluoroalkyl acids in adults: Isomers of C8 Health Project in China. *Environ. Res.* 172, 81–88. <https://doi.org/10.1016/j.envres.2019.02.013>

Oliaei, F., Kriens, D., Weber, R., Watson, A., 2013. PFOS and PFC releases and associated pollution from a PFC production plant in Minnesota (USA). *Environ. Sci. Pollut. Res.* <https://doi.org/10.1007/s11356012-1275-4>

Qualtrics, 2019. Qualtrics.

Schaider, L.A., Balan, S.A., Blum, A., Andrews, D.Q., Strynar, M.J., Dickinson, M.E., Lunderberg, D.M., Lang, J.R., Peaslee, G.F., 2017. Fluorinated Compounds in U.S. Fast Food Packaging. *Environ. Sci. Technol. Lett.* 4, 105–111. <https://doi.org/10.1021/acs.estlett.6b00435>

State of Michigan, 2018. PFAS Response - Statewide Testing Initiative [WWW Document]. URL https://www.michigan.gov/pfasresponse/0,9038,7-365-86510_87918-464299-,00.html (accessed 8.10.20).

Stockholm Convention, 2019. SC-9 / 12: Listing of perfluorooctanoic acid (PFOA), its salts and PFOA-related compounds 2–4.

Stockholm Convention, 2008. Listing of perfluorooctane sulfonic acid (PFOS), its salts and perfluorooctane sulfonyl fluoride (PFOSF) in Annex B of the Stockholm Convention on Persistent Organic Pollutants. *Unep/Pops/Cop.4/17* 15–18.

USEPA, 2012. The Third Unregulated Contaminant Monitoring Rule.

Wang, Z., Cousins, I.T., Scheringer, M., Buck, R.C., Hungerbühler, K., 2014. Global emission inventories for C4–C14 perfluoroalkyl carboxylic acid (PFCA) homologues from 1951 to 2030, Part I: production and emissions from quantifiable sources. *Environ. Int.* 70, 62–75. <https://doi.org/10.1016/J.ENVINT.2014.04.013>

Xie, S., Wang, T., Liu, S., Jones, K.C., Sweetman, A.J., Lu, Y., 2013. Industrial source identification and emission estimation of perfluorooctane sulfonate in China. *Environ. Int.* 52, 1–8. <https://doi.org/10.1016/J.ENVINT.2012.11.004>

Zeng, X.W., Bloom, M.S., Dharmage, S.C., Lodge, C.J., Chen, D., Li, S., Guo, Y., Roponen, M., Jalava, P., Hirvonen, M.R., Ma, H., Hao, Y.T., Chen, W., Yang, M., Chu, C., Li, Q.Q., Hu, L.W., Liu, K.K., Yang, B.Y., Liu, S., Fu, C., Dong, G.H., 2019. Prenatal exposure to perfluoroalkyl substances is associated with lower hand, foot and mouth disease viruses antibody response in infancy: Findings from the Guangzhou Birth Cohort Study. *Sci. Total Environ.* 663, 60–67. <https://doi.org/10.1016/j.scitotenv.2019.01.325>

Zeng, Z., Song, B., Xiao, R., Zeng, G., Gong, J., Chen, M., Xu, P., Zhang, P., Shen, M., Yi, H., 2019. Assessing the human health risks of perfluorooctane sulfonate by in vivo and in vitro studies. *Environ. Int.* 126, 598–610. <https://doi.org/10.1016/J.ENVINT.2019.03.002>

Tables

Table 1: Industry groups included in this study, the associated NAICS codes for specific industries within those groups, and the sources that led to the inclusion of those industries.

Industry Type	NAICS Codes	Source(s)
Textiles and Carpets	313210, 313310, 313320, 314110, 314999, 316110	Glüge et al. 2020; Guelfo et al. 2018; Herzke et al. 2012; Kotthoff et al. 2015;
Chemical Manufacturing	325199, 325320, 325622, 325612, 424690	Glüge et al. 2020; Guelfo et al. 2018; Hu et al. 2016; Nascimento et al. 2018
Paper Products	322121, 322219, 322220, 323111, 323120,	Glüge et al. 2020; Guelfo et al. 2018; Herzke et al. 2012; Schaider et al. 2017
Plastics/ Rubber Manufacturing	325211, 326112, 326113,	Glüge et al. 2020; Guelfo et al. 2018; Schaider et al. 2017
Grease, Oil, and Lubricant-related industries	324110, 324191, 333318, 336310	Glüge et al. 2020; Guelfo et al. 2018;
AFFF Use	488119, 928110	Glüge et al. 2020; Guelfo et al. 2018; Hu et al. 2016; Milley et al. 2018
Electroplating/Metal Finishing	332813, 332999	Glüge et al. 2020; Guelfo et al. 2018;
Dry Cleaners	812310, 812320	Clara et al. 2008; Glüge et al. 2020
Paint and Coating Manufacturing	325510	Glüge et al. 2020; Herzke et al. 2012;
Medical Instruments/Apparatuses	333249	Glüge et al. 2020; Gosetti et al. 2018
Photographic Film/Chemicals	325992	Glüge et al. 2020; Xie et al. 2013
Landfills	562212	Guelfo et al. 2018; Hepburn et al. 2019; Hu et al. 2016; Liu et al. 2017; Oliaei et al. 2013
Electronics	3344 (all)	Glüge et al. 2020; Lin et al. 2009

Table 2: Summary statistics of variables used in Probit regression analysis – 0.5 km and 1 km buffers (N= 1081).

Variable	Variable Description	Model(s)	Expected Effect on Dependent Variable (+/-)	Mean/(sd)	Min.	Max.
Detect	0 if non-detect (ND), 1 if detect (D)	All	n/a	0.099	0	1
Bedrock	0 if other bedrock type, 1 if "Coldwater Shale" formation	All	+	0.31 (0.46)	0	1
Welldepth	Well depth (ft.)	All	-	173.90 (107.76)	21	815
Welldepthsq	Well depth squared	All	-	4184.86 (60263.90)	576	664225
Landcover	0 if other land classification, 1 if "developed, low intensity" (NLCD code 22)	All	+	0.23 (0.42)	0	1
Soildrainage	0 if other drainage classification, 1 if "well drained", "excessively drained", or "somewhat excessively drained"	All	+	0.74 (0.44)	0	1
Percentlandfill	Percent of sites as landfills within 0.5 km of wells	1	+	0.01 (0.09)	0	1
Percentmetalfoil	Percent of sites as metal foil within 0.5 km of wells	1	+	0.01 (0.07)	0	1
Landfill	Number of sites as landfills within 0.5 km of	2	+	0.01 (0.16)	0	3

	wells					
Metalfoil	Number of sites as metal foil within 0.5 km of wells	2	+	0.01 (0.07)	0	1
FacQtrsWit	Total number of quarters within last 3 years where all sites within 0.5 km buffer have failed to comply with EPA environmental standards	1 and 2	+	0.05 (0.66)	0	12
Percentairport	Percent of sites as airports within 1 km of wells	3	+	0.01 (0.07)	0	1
Percentlandfill	Percent of sites as landfills within 1 km of wells	3	+	0.03 (0.17)	0	1
Percentelectroplating	Percent of sites as electroplating within 1 km of wells	3	+	0.01 (0.12)	0	1
Percentallothers	Percent of sites as all other industry types within 1 km of wells	3	+	0.93 (0.25)	0	1
Airport	Number of sites as airports within 1 km of wells	4	+	0.01 (0.11)	0	2
Landfill	Number of sites as landfills within 1 km of wells	4	+	0.07 (0.53)	0	10
Electroplating	Number of sites as electroplating within 1 km of wells	4	+	0.02 (0.16)	0	2

Allothers	Number of sites as all other industry types	4	+	0.06 (0.30)	0	3
	within 1 km of wells					
FacQtrsWit	Total number of quarters within last 3 years where all sites within 1 km buffer have failed to comply with EPA environmental standards	3 and 4	+	0.08 (0.88)	0	12

Model 1: 0.5km buffer, percent of industry sites

Model 2: 0.5km buffer, number of industry sites

Model 3: 1km buffer, percent of industry sites

Model 4: 1km buffer, number of industry site

Table 3: Michigan Industries within defined buffer sizes (0.1,0.5,1.0 km) of drinking water wells with detected PFAS concentrations. For total number of identified industrial sites in Michigan, n = 2,696ⁿ.

NAICS Code	Industry Classification	Number of facilities within each size buffer zone around contaminated wells		
		0.1 km	0.5 km	1.0 km
313320	Fabric Coating Mills (n = 9)	0	0	0
313310	Textile and Fabric Finishing Mills (n = 4)	0	0	0
336310	Motor Vehicle Gasoline Engine and Engine Parts Manufacturing (n = 48)	0	0	0
324110	Petroleum Refineries (n = 11)	0	0	0
325320	Pesticide and Other Agricultural Chemical Manufacture (n = 10)	0	0	0
325992	Photographic Film, Paper, Plate, and Chemical Manufacture (n = 7)	0	0	0
326112	Plastics Packaging Film and Sheet (including Laminated) Manufacture (n = 9)	0	0	0
325211	Plastics Material and Resin Manufacture (n = 116)	0	1	2
316110	Leather and Hide Tanning and Finishing (n = 7)	0	0	0
322220	Paper Bag and Coated and Treated Paper Manufacture (n = 14)	0	0	0
332813	Electroplating, Plating, Polishing, Anodizing, and Coloring (n = 317)	0	1	2
314110	Carpet and Rug Mills (n = 2)	0	0	0
812310, 812320	Dry Cleaning and Laundry Services (n=133)			6
325199	All Other Basic Organic Chemical Manufacture (n = 70)	0	0	0
325510	Paint and Coating Manufacture (n = 104)	0	0	0
323111	Commercial Printing (except Screen and Books) (n = 102)	0	0	1
313210	Broadwoven Fabric Mills, Manmade Fibers and Silk (n = 4)	0	0	1

424690	Chemicals and Allied Products, Not Elsewhere Classified (n = 14)	0	0	0
488119	Airports (n = 69)	0	0	3
322121	Paper Mills (n = 33)	0	1	2
332999	Metal Foil and Leaf Manufacture (n = 205)	0	1	6
324191	Lubricating Oils and Greases (n = 46)	0	1	1
562212	Solid Waste Landfill (n = 1373)	0	3	8
928110	Department of Defense Sites (n = 7)	0	0	0
323120	Support Activities for Printing (n = 13)	0	0	0
322219	Sanitary Food Containers, Except Folding (n = 3)	0	0	0
325611	Perfumes, Cosmetics, and other Toilet Preparations (n = 46)	0	0	0
333249	Surgical and Medical Instruments and Apparatus (n = 45)	0	0	0
325612	Specialty Cleaning, Polishing, and Sanitation Preparations (n = 27)	0	0	0
333318	Service Industry Machinery, Not Elsewhere Classified (n = 16)	0	0	0
334413	Semiconductor and Related Device Manufacture (n = 20)	0	0	0
326113	Unsupported Plastics Film and Sheet (n = 19)	0	0	0
314999	Waterproof Outerwear (n = 8)	0	0	0
Total (n = 2696)		0	8	26

ⁿn = number of industries of each code in Michigan. Some sites classified as multiple industries on list and are counted totals for each industry, but once in grand total.

Table 4: Probit regression analysis results for 1 km buffers (n = 1081).

Variable	Model 1: 1 km buffer, %sites	B	Sig.	Model 2: 1 km buffer, #sites	B	Sig.
	Wald Chi-Square			Wald ChiSquare		
(Intercept)	0.027	0.062	0.869	17.743	0.791	0.000***
Bedrock	0.077	0.033	0.782	0.009	0.011	0.924
Welldepth	27.196	0.007	0.000***	27.190	0.007	0.000***
Welldepthsq	19.258	-9.900*10 ⁻⁶	0.000***	19.148	-9.853*10 ⁻⁶	0.000***
Landcover	1.291	0.155	0.256	1.221	0.150	0.269
Soildrainage	7.758	-0.384	0.005***	7.771	-0.382	0.005***
Percentairport/ Airport	0.036	-0.129	0.850	2.669	-0.603	0.102
Percentlandfill/ Landfill	1.626	0.552	0.202	0.109	0.035	0.741
Percentelectroplating/ Electroplating	2.987	2.150	0.084*	1.104	0.549	0.293
Percentallothers/ Allothers	4.924	0.735	0.026**	0.893	-0.172	0.345
FacQtrsWit	0.127	-0.021	0.722	0.101	-0.018	0.751
	Wald Chi-Square	Degrees of Freedom	Sig.	Wald Chi-Square	Degrees of Freedom	Sig.
Omnibus test	44.864	10	0.000	40.760	10	0.000
Observations	1081					

*, **, *** indicates statistical significance at $p \leq 0.10$, $p \leq 0.05$, and $p \leq 0.01$ respectively

Table 5: Probit regression analysis results for 0.5 km buffers (n = 1081).

Variable	Model 3: 0.5 km buffer, % sites	B	Sig.	Model 4: 0.5 km buffer, #sites	B	Sig.
	Wald Chi- Square			Wald Chi- Square		
(Intercept)	18.050	0.793	0.000***	17.973	0.792	0.000***
Bedrock	0.104	0.038	0.747	0.100	0.037	0.752
Welldepth	26.119	0.007	0.000***	26.106	0.007	0.000***
Welldepthsq	18.241	-9.589*10 ⁻⁶	0.000***	18.239	-9.596*10 ⁻⁶	0.000***
Landcover	1.054	0.139	0.305	1.062	0.139	0.303
Soildrainage	7.437	-0.371	0.006***	7.508	-0.373	0.006***
Percentlandfill/ Landfill	0.421	-0.310	0.516	0.151	-0.107	0.697
Percentmetalfoil/ Metalfoil	0.524	-0.456	0.469	0.520	-0.454	0.471
FacQtrsWit	0.937	-0.062	0.333	0.932	-0.062	0.334
	Wald Chi- Square	Degrees of Freedom	Sig.	Wald Chi- Square	Degrees of Freedom	Sig.
Omnibus test	37.040	8	0.000	36.783	8	0.000
Observations	1081					

*, **, *** indicates statistical significance at $p \leq 0.10$, $p \leq 0.05$, and $p \leq 0.01$ respectively

Table 6: Significant clusters and outliers identified using Local Moran's I for PFAS clusters in Michigan (detected wells only).

Well Location	City/Town	Cluster/Outlier Type	Moran's I index value	Z-score	p-value ¹
Plainwell		High-high	0.000036	1.390	0.03
Kalamazoo		High-high	0.00014	2.813	0.012
Plainwell		High-high	0.000105	1.684	0.05
Parchment		High-low	-0.0009	1.842	0.048
Houghton		Low-low	0.000002	0.272	0.004
Sherman Township		Low-low	0.000002	0.314	0.02
Richmond Township		Low-low	0.023118	0.140	0.01
Richmond Township		Low-low	0.023118	0.174	0.01
Marquette		Low-low	0.00003	0.190	0.01
KL Sawyer AFB		Low-low	0.000004	0.407	0.002
Escanaba		Low-low	0.000004	0.321	0.018
Gladstone		Low-low	0.000004	0.363	0.006
Kawkawlin		Low-low	0.000008	0.263	0.016
Northport		Low-low	0.000003	0.227	0.046
Parkwood Village		Low-low	0.000006	0.706	0.028
Climax		Low-high	-0.000058	-2.904	0.032
Delton		Low-high	-0.000034	-2.438	0.046
Gun Plain Township		Low-high	-0.000124	-2.904	0.032
Lockport Township		Low-high	-0.00003	-1.890	0.044
Otsego		Low-high	-0.000061	-1.594	0.042
Portage		Low-high	-0.000058	-3.485	0.012
Kalamazoo		Low-high	-0.000049	-2.159	0.048
Kalamazoo		Low-high	-0.000146	-3.898	0.032

¹Threshold for significance was defined at p-values of 0.05 or below.

Figures

Figure 1

(a) Total groundwater wells per county, indicated by the numerical value on each county, and percentage of wells with PFAS in each county as shown by color scale. (b) Number of PFAS industry sites in Michigan by county.

Figure 2

Local Moran's I for PFAS clusters in Michigan (detected wells only).