

Spatial Prediction of Concentrations of Per- and Polyfluoroalkyl Substances (PFAS) in European Soils

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Cite This: <https://doi.org/10.1021/acs.estlett.3c00633>



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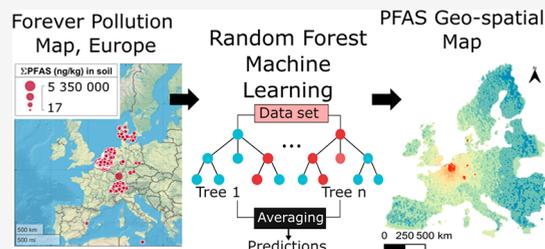
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ABSTRACT: Currently, little is known about the spatial distribution of per- and polyfluoroalkyl substances (PFAS) in soils. In this study, machine learning was applied to a data set from the Map of Forever Pollution in Europe (MFPE) containing 6697 scattered PFAS soil concentration measurements to comprehensively predict PFAS concentrations in European soils. The model is based on a regression analysis between the PFAS soil concentrations and the distance to presumptive point sources indicated by the MFPE. Generally, decreasing PFAS concentrations were observed with increasing distances to the nearest point sources. Subsequently, on the basis of the regression analysis, a map was generated showing the PFAS concentrations in European soils by interpolating the model predictions at 10 000 random spots where no measurements were available. The map revealed that a significant portion of European soils is potentially contaminated with PFAS at concentrations of >5000 ng/kg. At this concentration, the mobilization of PFAS can lead to seepage water concentrations of 2–5 ng/L surpassing current and proposed drinking water guidelines in Europe. This illustrates the need for lower PFAS soil threshold values. Overall, the produced map provides, for the first time, comprehensive information about European PFAS soil contamination and serves as a basis for assessing its environmental risks.

KEYWORDS: per- and polyfluoroalkyl substances (PFAS), soil contamination, Map of Forever Pollution in Europe (MFPE), spatial analysis, machine learning



1. INTRODUCTION

Per- and polyfluoroalkyl substances (PFAS) make up a group of >5000 anthropogenically produced compounds with unique physicochemical surface properties that have been widely used since the 1950s in products such as surfactants in textiles, packaging material, or aqueous film-forming foams (AFFF).^{1–4} The two major PFAS classes are perfluoroalkyl carboxylates (PFCA) and perfluoroalkanesulfonates (PFSA), whereby perfluorooctanesulfonate (PFOS) and perfluorooctanoate (PFOA) have been most frequently analyzed.^{5,6} PFAS are composed of a fluorinated carbon chain and a functional hydrophilic group with a strong carbon–fluorine bond, making them hardly (bio)degradable, highly persistent, and possibly bioaccumulative.^{7–10} Consequently, PFAS are now widely distributed at a wide range of concentrations in the environment, including soil, surface water, and groundwater as well as biota.^{1,11–13} The ubiquitous environmental presence of PFAS is recognized as a threat to humans, already at low concentrations (nanograms per liter range), due to adverse health effects associated with PFAS (see ref 14 and references therein for a comprehensive review). As a result, global concerns about environmental PFAS contamination have become more serious over the past two decades, which has in turn led to stricter safety regulations.^{15–17} For instance, in 2018, the European Food Safety Authority (EFSA) decreased

the tolerable daily intake from 1500 and 150 ng/kg of body weight for PFOA and PFOS, to 1.8 and 0.8 ng/kg of body weight, respectively. Moreover, in the 2020 revision, the threshold value was decreased even further to 0.63 ng/kg of body weight for the sum of four PFAS, including PFOA, PFOS, perfluorononanoic acid (PFNA), and perfluorohexanesulfonic acid (PFHxS).¹⁸ In addition, in March 2023, the U.S. Environmental Protection Agency (EPA) introduced, for the first time, a nationwide threshold value of 4 ng/L in drinking water for PFOA and PFOS.¹⁹

Despite the efforts to decrease threshold values for PFAS concentrations, PFAS contamination remains a significant environmental and public health concern due to the persistent nature of PFAS and their widespread use combined with their toxicity at low concentrations.^{14,20} A crucial step toward reducing the emission of PFAS to the environment involves the identification of potential PFAS sources and the determination of how they release PFAS into the environment.

Received: September 5, 2023

Revised: October 25, 2023

Accepted: October 26, 2023

This provides then the basis for implementing appropriate actions regarding future monitoring strategies and remedial measures.^{21–23} A prerequisite to this step is to compile extensive *in situ* measurements of PFAS concentrations with respect to suspected point sources. The recently published Map of Forever Pollution in Europe (MFPE)²⁴ provides, for the first time, a large data set of ~6600 measurements of PFAS concentrations in soils in Europe along with the locations of the suspected PFAS point sources, including firefighting incidents/activities, industrial sites, military sites, wastewater management sites, airports, and PFAS manufacturing sites.²⁴ However, the MFPE remains on a descriptive level, particularly with the lack of information about PFAS soil concentrations at the locations where no measurements are available. Moreover, the MFPE does not explore the relationship between different point sources and PFAS soil concentrations.

To bridge this research gap, the overall goal of this study is to predict PFAS concentrations in soils throughout Europe using a machine learning approach. The model predicts the PFAS soil concentration on the basis of the relationship between PFAS concentration and distance to nearest point sources. The predictions are then interpolated to generate a map of soil contamination by PFAS in Europe. This generated map across Europe can be used to assess the extent of soil contamination by PFAS, facilitating the implementation of targeted remedial strategies for minimizing environmental exposure to these harmful chemicals.

2. METHODS

2.1. Data Source. In this study, geo-scattered measurements of PFAS soil concentrations were retrieved from the MFPE.²⁴ This map was created by *Le Monde* and its 17 partners from the Forever Pollution Project and published on February 23, 2023.²⁴ The map partially illustrates the extent of Europe's contamination by PFAS and provides geo-spatial information about contaminated sites. In total, it shows 19 500 locations where PFAS concentrations are measured, of which 6697 data points indicate total PFAS concentrations (\sum PFAS) in soil ranging from 17 to 5 350 783 ng/kg. \sum PFAS denotes the sum of all detectable PFAS, including PFOS, PFOA, PFNA, PFBS (perfluorobutanesulfonic acid), PFHxS, and PFHxA (perfluorohexanoic acid), with a detection limit of 10 ng/kg in soil. The geo-coordinates of the 6697 soil sampling locations, which were used as the population for this study, are shown in Figure S1. In addition, the map contains geographic coordinates of the so-called presumptive contamination sites, which are point sources with current or past activities associated with possible release/leakage of PFAS into soils. This includes 12 271 firefighting incidents/training activities, 1025 airports (including commercial civilian airports and air bases), 3204 industrial areas, 735 military sites, 4789 waste management sites, and 20 PFAS manufacturing sites (Figure S2).

2.2. Model Development. To predict PFAS soil concentrations in Europe, the measured \sum PFAS soil concentrations, reported by the MFPE, were first analyzed with respect to linear distances from the nearest presumptive point sources. Subsequently, a regression analysis was performed using a machine learning algorithm to model the spatial relation between the PFAS concentrations and the distance to the presumptive point sources. The machine learning model was aimed at capturing the nonlinearity of trends as well as the general patterns, which in turn enabled us

to predict PFAS concentrations at the locations where no measurements were available. The algorithm was implemented in R programming language with the Random Forest package.²⁵ Random Forest was chosen due to its robustness, in comparison to alternative algorithms, when dealing with substantial statistical noise and outliers like those present in the source data used in this study.^{25,26} It also can sufficiently prevent overfitting, a common source of error in predictive machine learning, by randomly selecting subsets of features and observations to build individual decision trees.²⁵ The model treated the \sum PFAS soil concentrations as the target, and each data set of calculated distances as an independent and spatially continuous predictor (six predictors in total).

The model was constructed by partitioning the total data set into an 80% training set and a 20% testing set. To optimize its performance, the model hyperparameters were tuned by using a grid search approach with a fixed random seed. The model was run through 10-fold cross validation, and the prediction results from the 10 cross validations were averaged. The model performance was evaluated using various statistical parameters, including the coefficient of determination (R^2), the root-mean-squared error (RMSE), and the standard deviation (SD). It was found that the model predictions deviate from the exceptionally high PFAS concentrations, and therefore, high values (>500 000 ng/kg) were excluded from the input data set. This, however, had minor effects on the model performance and reliability, as more than 90% of the measurements were still included in the input data.

The developed model was then used to predict the \sum PFAS soil concentrations at 10 000 random locations in Europe. Subsequently, to generate a spatially continuous map of \sum PFAS soil concentration over Europe, an inverse distance-weighted (IDW) interpolation in QGIS²⁷ was implemented on top of the model predictions at 10 000 random locations. The use of IDW ensures that the gradual changes in \sum PFAS across the entire region are captured.²⁷ It also maintains the map's smoothness and continuity, thereby improving its visual clarity and interpretability. Although IDW may introduce some inaccuracies, the trade-off was minimized through trial and error and by choosing a pixel resolution of 0.1 that allows small-scale features to be contained, while minimizing the noise.

Finally, the \sum PFAS seepage water concentration from the predicted \sum PFAS soil concentrations was estimated using the following equation:²⁸

$$C_w = \frac{C_{\text{soil}}}{\phi_w / \rho + K_d} \quad (1)$$

where C_w is the \sum PFAS seepage water concentration (nanograms per liter), C_{soil} is the predicted \sum PFAS soil concentration (nanograms per kilogram), ϕ_w is the water content (dimensionless), ρ is the soil bulk density (kilograms per liter), and K_d is the soil–water distribution coefficient (liters per kilogram). The average K_d value for PFAS can vary depending on the specific PFAS and environmental conditions. In the current study, K_d was set in the range of 1000–2000, which corresponds to the intermediate range of K_d values reported for PFOA and PFOS.²⁹ The water content of soils in Europe is in the range of 0.4–0.8, with an average of 0.6–0.7.³⁰ The soil bulk density typically falls between 1.0 and 1.8 kg/L, with an average of ~1.4 kg/L.³¹

In our model, we used only the correlation between PFAS soil concentrations and the distance to presumptive sources to predict Σ PFAS soil concentrations across Europe and did not consider other parameters. This choice was driven by the understanding that the concentration–distance relationship is not necessarily categorized on the basis of the geographical zones/regions. Consequently, this approach minimizes the potential bias originating from the non-uniform distribution of the soil samples (population characteristics), which are predominantly located in Belgium, Denmark, and Switzerland (Figure S1). In contrast, a correlation between the unevenly distributed PFAS soil concentration measurements and parameters such as annual precipitation or soil physical properties leads to an over-representation of the parameter ranges in the predominantly sampled locations in Belgium, Denmark, and Switzerland [the condition of samples (Figures S6 and S9)]. This can be attributed to the substantial regional/zonal variations of these variables. For instance, the soil physical properties in Belgium, Denmark, and Switzerland do not represent the full spectrum of the European soil physical properties (Figure S6). Therefore, a correlation between PFAS measurements and soil physical properties as well as other parameters such as annual precipitation in these regions cannot be related or applied to other parts of Europe.

3. RESULTS AND DISCUSSION

3.1. PFAS Spreading from Point Sources. Scatter plots of the Σ PFAS soil concentration versus the determined distance to nearest point sources are presented in Figure 1A–F. The plots show a decreasing Σ PFAS concentration with increasing distance to the firefighting incident sites, industrial sites, and waste management sites (Figure 1A–C). However, the PFAS spreading patterns for these sources are distinct. While it appears that the firefighting incidents, waste management sites, and industrial sites release the PFAS into soils only within a radius of 10–20 km (Figure 1A–C), the PFAS manufacturing sites seem to spread the PFAS farther in soils (Figure 1D). Although the reasons for such spreading patterns are currently unknown, they might be related to the total volume of the released PFAS as well as the time period during which the PFAS were emitted. For the military sites and airports, there are comparably fewer PFAS measurements at short distances due to restricted access as mentioned by the MFPE²⁴ (Figure 1E,F). This could possibly introduce bias when assessing the contribution of military activities and airports to PFAS soil contamination in Europe. In this case, as panels E and F of Figure 1 make evident, no clear trend can be detected.

3.2. Regression Analysis. An R^2 value of 0.82 and an RMSE of 0.46 were obtained when utilizing the entire data set for the prediction of PFAS soil concentrations (Table S1). Notably, there was a discernible difference of 0.22 between the R^2 values derived from the training and test data. This discrepancy can indicate the presence of an overfitting pattern during the training process. In such instances, regression coefficients tend to capture random fluctuations in the data rather than fitting to global patterns. In this case, overfitting appears to be moderate, as evidenced by the close and comparable values of RMSE between the training and test data, 0.53 and 0.97, respectively.

To further evaluate the performance of the model, the predicted and measured PFAS soil concentrations were cross-plotted (Figure S4A). For both the training and test data sets,

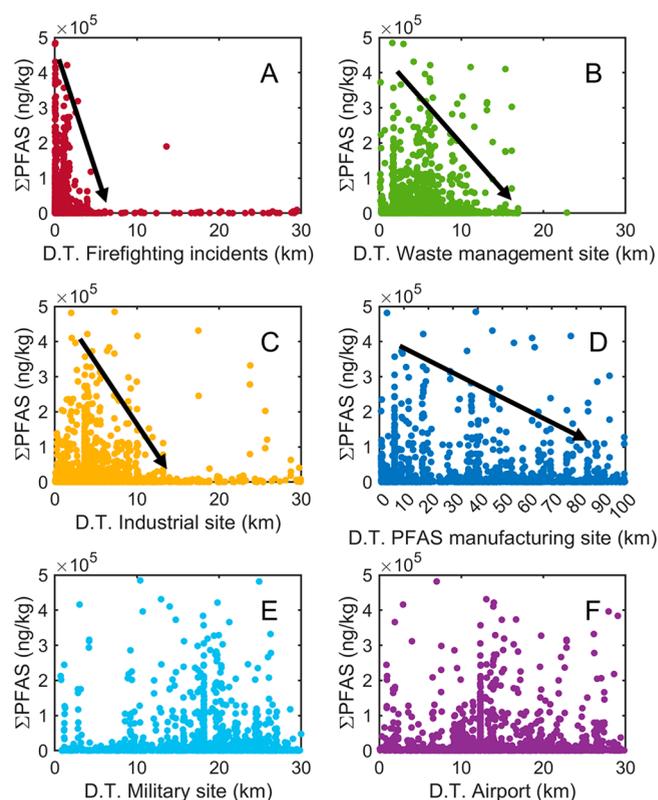


Figure 1. Total PFAS concentrations (Σ PFAS) in soil with respect to the distance from the nearest presumptive point sources (D.T., distance to), including (A) firefighting incidents, (B) waste management sites, (C) industrial sites, (D) PFAS manufacturing sites, (E) military sites, and (F) airports. The black arrow indicates the general trend that can be visually detected.

most of the data points cluster around the bisector line, indicating a decent model performance. Moreover, the relative deviation (RD) for the test data set (Figure S4B) shows a symmetrical distribution around zero, meaning that the model is not biased toward overprediction or underprediction. Overall, the metrics demonstrate that the PFAS soil concentrations can be predicted with satisfactory accuracy using a concentration–distance relation, thus providing the prerequisite for generating the geo-spatial map of PFAS soil concentrations across Europe. In addition, the performance of our model is superior (with an R^2 of 60% to unseen data) compared to that of previous predictive models (with R^2 values in the range of 35–55%) that rely on regression analysis between PFAS soil concentrations and their sample coordinates (latitude and longitude).³² It should be noted that adding geo-coordinates to our model had a minimal impact on the model's performance; thus, they were not included.

3.3. Map of PFAS in Soil. The generated geo-spatial map of the Σ PFAS soil concentrations in Europe is shown in Figure 2. The map shows that the soils in central and western Europe are potentially contaminated with PFAS at concentrations exceeding 30 000 ng/kg. The cause of such relatively high PFAS soil concentrations in these zones is likely to be related to the high density of PFAS point sources in these areas. Furthermore, the map suggests that large areas of central Europe and the southern part of Scandinavia are potentially contaminated with PFAS at concentrations of >5000 ng/kg

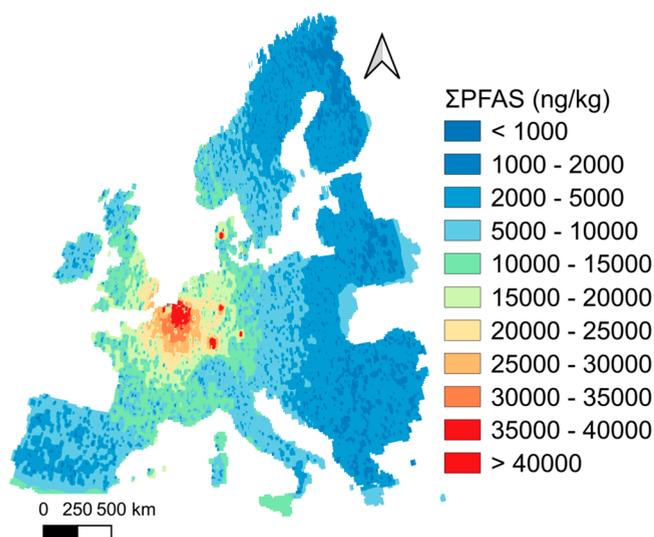


Figure 2. Geo-spatial map of predicted PFAS [ΣPFAS (nanograms per kilogram)] concentrations in European soils. The different colors represent different PFAS soil concentrations.

(Figure 2). Moreover, the soil in the eastern part of Europe and, as expected, in remote areas, particularly in the northern part of Scandinavia is less contaminated with PFAS with concentrations of <2000 ng/kg (Figure 2). The lower ΣPFAS concentrations in these areas can be attributed to fewer PFAS emission activities.

Using eq 1, it can be estimated that a PFAS soil concentration of 5000 ng/kg may result in ΣPFAS concentrations ranging from 2 to 5 ng/L in the soil seepage water. This range exceeds the current Danish threshold values for drinking water (2 ng/L) for the sum of PFOA, PFOS, PFNA, and PFHxS and is in the prohibited range of the U.S. EPA guideline values for PFOA and PFOS.^{19,33} Concerningly, the soils of large areas of Europe exceed a PFAS soil concentration of 5000 ng/kg (Figure S10), which poses a threat to freshwater resources used by drinking water supplies with potentially adverse health effects for humans. In the context of threshold values, it is worth noting that currently there is no dedicated European legislation on soil quality with regard to PFAS contamination, unlike for drinking water. Only recently, in Norway, an updated report by the national EPA has proposed to decrease the threshold value for PFOS from 100 000 to 2000 ng/kg of soil and setting a new threshold value for PFOA as low as 11 000 ng/kg of soil.³⁴ The results of this study highlight the necessity of reviewing the current legislation and imposing new threshold values for PFAS soil concentrations in Europe to protect freshwater resources from PFAS contamination and limit human exposure to such toxic chemicals.

In doing so, it is evident that more PFAS soil measurements are urgently needed to refine the estimation of PFAS soil concentrations in Europe and worldwide. Our recommendation is to adopt a sampling strategy similar to the Land Use and Cover Area frame Statistical survey (LUCAS) Europe, which will ensure that the sample data fulfill essential conditions such as representativeness on a spatial scale.³¹ With the availability of additional PFAS soil measurements, our model can be improved further by incorporating climate variables and environmental/landscape covariates such as soil properties and population density. It is also possible to account for the

synergic effects of multiple sources using a plume model instead of the distance to individual point sources. Moreover, the variations in PFAS soil concentration across different ecological zones and strata can be determined using geo-statistical methods.³⁵ It can be expected that the resulting model will be indispensable in devising effective mitigation strategies, the purpose of which is to curb the spread of PFAS and ultimately safeguard humans from excessive exposure to these harmful substances.

■ ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge at <https://pubs.acs.org/doi/10.1021/acs.estlett.3c00633>.

The Supporting Information contains detailed information about the applied machine learning approach in this study (PDF)

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Notes

The authors declare no competing financial interest.

■ ACKNOWLEDGMENTS

The authors thank the authors of the Map of Forever Pollution in Europe (MFPE), including Gary Dagorn, Raphaëlle Aubert, Stéphane Horel, Luc Martinon, and Thomas Steffen, for allowing us to use the data set of the MFPE for this study.

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