From: Sam i
To: Coffin Butte Landfill Appeals
Subject: Testimony revised/resubmitted with pdfs
Date: Friday, October 10, 2025 9:24:13 PM
Attachments: Testimony October 10, 2025.pdf
Landfill Gas-PFAS.pdf
Decontamination of landfill waste leads to increase in toxic chemicals, says study Waste The Guardian.pdf
PFAS hazard in surface water.pdf

Dear Benton County Commissioners,

I oppose Coffin Butte landfill's expansion.
Clean air and water are precious vital resources for all of us in Benton County.
A feeling of ease in our communities because our elected officials consistently stand up for our safety, health and welfare, is vitally important.
Promises have been broken over the years by the owners and operators of Coffin Butte landfill. And our county has failed in its oversight.
The landfill expansion if approved weighs on our common resources to a great extent over an extended period of time, affecting our grandchildren and their children.

It's Wet Here

This post-war landfill is in a very wet part of a wet valley.
Adair Village gets 51 inches of rain, on average, per year.
At Arlington, an alternative regional landfill with large capacity and accessible by rail, the average annual rainfall is less than 9 inches.

There is a strong link between wet conditions and leachate production in landfills.
Every droplet of water that splashes down on an open landfill cell will slowly trickle through the trash, transforming into a concentrated liquid waste known as leachate.

Moisture encourages bacterial decomposition, which is the primary process for methane generation in landfills.

Landfill Gas

Billowing tarps, tears and odors indicate the release of methane and landfill gasses into our air. These gasses also contain PFAS. "According to an EPA-funded study recently published in the peer-reviewed Environmental Science and Technology Letters, PFAS could be escaping landfills via gas at concentrations similar to — if not higher than — liquid leachate."
I have attached the research paper entitled *Landfill Gas: A Major Pathway for Neutral Per- and Polyfluoroalkyl Substance (PFAS) Release.*

In the paper, researchers noted that they have "detected "unexpectedly" high levels of PFAS in landfill gas, adding to a growing body of evidence on how "forever chemicals" leave waste sites."

PFAS Forever Chemicals

All over the world, PFAS in landfills are growing. In Europe and the UK research has been forward thinking and robust. I cite here a Guardian article from November 4, 2024 telling of a project seeking to remove PFAS forever chemicals from leachate that contaminates groundwater and surface water - and can cause health problems, including kidney and testicular cancer.

"Processes intended to decontaminate noxious liquid landfill waste before it enters rivers and sewers have been found to

increase the levels of some of the worst toxic chemicals, a study has shown.

Landfills are well known to be a main source of PFAS forever chemicals – or per- and polyfluoroalkyl substances – but the new study shows that the treatment plants designed to clean up the liquid waste can instead boost the levels of banned PFAS such as PFOA and PFOS, in some cases by as much as 1,335%."

https://www.theguardian.com/environment/2024/nov/04/decontamination-of-landfill-waste-leads-to-increase-in-toxic-chemicals-says-study

Currently, PFAS are ubiquitous in surface waters- and that means the Willamette River, into which untold numbers of gallons of PFAS containing landfill leachate from Coffin Butte have been released untreated after being transported to the Corvallis and Salem water treatment plants.

Solutions do not exist to "treat" PFAS forever chemicals. It behooves us to lessen the amount of toxic leachate in our region by not approving the expansion of Coffin Butte in this very wet part of the Willamette Valley.

Local and global concerns regarding the persistence of PFAS, how they move through the environment, and the potential for adverse health impacts of PFAS are increasing. Here in Benton County, we have the ability to make a decision in order to safeguard the health of our population and our natural resources.

A Big Liability

The Coffin Butte owners and operators have not been good stewards. They have not been good partners. There are many incidences of violations and mishandling of the confidence and trust placed in them.

Coffin Butte landfill is not a resource to us - rather it is a mountainous and growing liability and a source of real health and environmental concerns.

The landfill's expansion would further impinge upon our rights to our health and our finite natural resources of clean air and water. There are consequences of leachate that percolates into groundwater, or that is disposed of in the Willamette River. Consequences of PFAS that burp into the air along with landfill gasses. Those PFAS forever chemicals are percolating into our bodies and natural environments.

From Politico, October 2025- *A group of 24 European politicians whose blood was tested for toxic PFAS chemicals over the summer all had the substances in their bodies, the NGOs involved in the testing revealed Tuesday.*
"I tested positive for four substances, and three of them can harm unborn children, act as endocrine disruptors, cause liver damage, and are suspected of being carcinogenic," said Danish Environment Minister Magnus Heunicke in a written statement, describing his results as a "frightening reality."

PFAS in our environments are ubiquitous locally and globally. "Owing to their resistance to heat, water, and oil, over 14,000 per - and polyfluoroalkyl substances (PFAS) are extensively utilized in various industrial and consumer applications, such as in nonstick
cookware, firefighting foams, food containers, and anti-staining fabrics."

Please oppose this expansion.
Thank you for your diligence.

Susan Walenza
1415 NW Greenwood Place
Corvallis

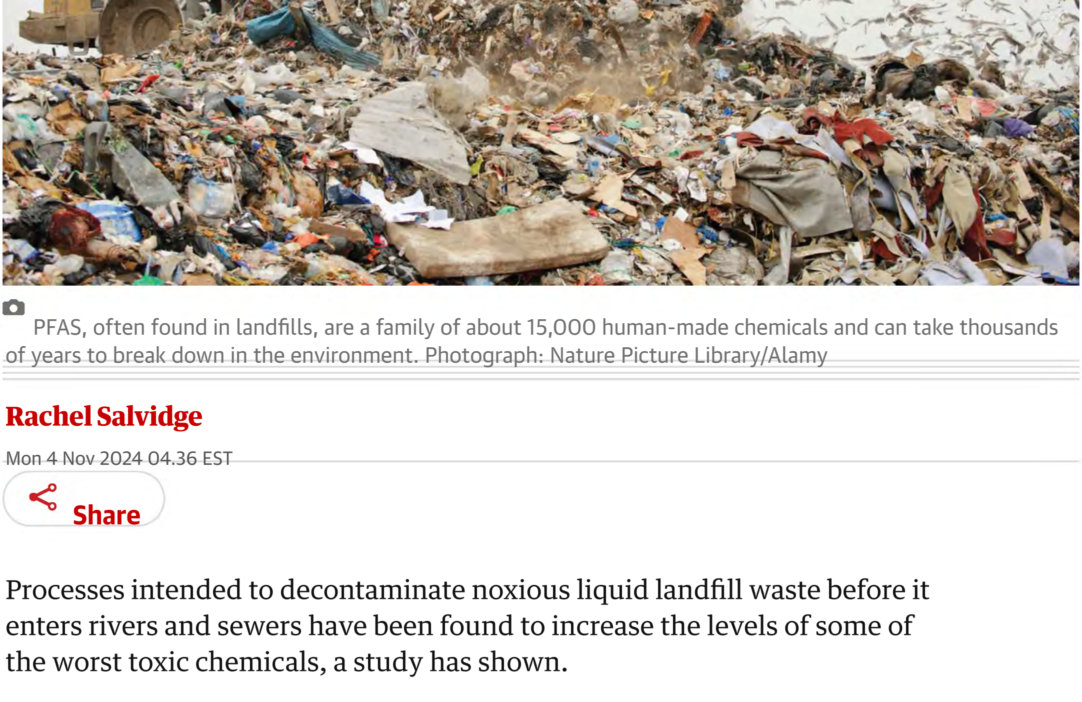
Susan

Waste

This article is more than **10 months old**

Decontamination of landfill waste leads to increase in toxic chemicals, says study

Exclusive: Researchers find treatment plants designed to clean up leachate liquid waste boost levels of banned PFAS



PFAS, often found in landfills, are a family of about 15,000 human-made chemicals and can take thousands of years to break down in the environment. Photograph: Nature Picture Library/Alamy

Rachel Salvidge

Mon 4 Nov 2024 04.36 EST

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Processes intended to decontaminate noxious liquid landfill waste before it enters rivers and sewers have been found to increase the levels of some of the worst toxic chemicals, a study has shown.

Landfills are well known to be a main source of PFAS forever chemicals – or per- and polyfluoroalkyl substances – but the new study shows that the treatment plants designed to clean up the liquid waste can instead boost the levels of banned PFAS such as PFOA and PFOS, in some cases by as much as 1,335%.

'Forever chemicals': what are PFAS and what risk do they pose?

Read more

PFAS are a family of about 15,000 human-made chemicals with nonstick properties that are used in a wide range of consumer products and industrial processes. They can take thousands of years to break down in the environment and the handful that have been studied in detail have been found to be toxic, with PFOA and PFOS linked to cancers and other diseases. PFAS pollution is widespread, having been found in the remotest parts of the world, and it is thought every US citizen has it in their blood.

Using data from an Environment Agency investigation into landfill liquid waste, which is known as leachate, Dr David Megson from Manchester Metropolitan University, who co-authored the study found "that instead of removing the banned chemicals PFOS and PFOA our treatment plants are actually creating them … likely being formed from the transformation of other PFAS within a chemical soup".

Megson is concerned that the understanding of what is going on in the UK at landfill sites is poor and that monitoring "only looks at a few specific PFAS, so we are only getting a tiny snapshot of what is actually out there and what impact it may be having".

The study looked at the leachate from 17 historical and operational landfills, just a fraction of the total across the country. Pippa Neill from the Ends Report, a co-author of the study, said that "with potentially hundreds of landfill operators legally allowed to discharge their treated leachate into the environment" there is an "urgent need" for more research so that PFAS can be disposed of properly.

There is also "an urgent need to ban all PFAS globally, whether through the existing Stockholm convention or a new global treaty on PFAS", according to Dr Sara Brosché, an adviser at the International Pollutants Elimination Network. "PFOS and PFOA were known by the producers to be toxic from the beginning of their use in consumer products, and they continue to poison the environment and our bodies many years after they have been regulated. A multitude of PFAS are now in use with little or no publicly disclosed information about where they are used or their health impacts."

In an attempt to halt contamination, the European Commission is considering a groundbreaking proposal to regulate thousands of PFAS as one class, something that is being fiercely contested by the PFAS industry. The UK has not followed the EU's lead, prompting dozens of the world's leading PFAS experts to write directly to UK ministers on Thursday, urging the government to "take a more ambitious approach and follow the science … Regulating all PFAS as one group is the only way to tackle PFAS pollution".

Dr Shubhi Sharma, a scientific researcher at the charity Chem Trust, said: "PFAS emissions from landfills can contaminate the surrounding groundwater and surface water and are linked to serious health risks, such as kidney and testicular cancer. The UK government must take immediate action to regulate this entire group of PFAS."

Dr Daniel Drage, an associate professor at the University of Birmingham, is also concerned that the same thing is happening in a range of treatment systems.

"It's paramount that we identify other treatment processes that remove PFAS from leachate prior to its release into the environment," he said. "This is a multibillion pound global public health issue and likely to go beyond government expenditure. I would suggest that industries that have profited substantially from the use of PFAS over the last half a century have a moral duty to protect future generations from the consequences of these uses."

A spokesperson for the Environment Agency confirmed it is "working closely with the landfill industry" and that it is "carrying out further investigations about PFAS within the landfill waste mass, treatment processes, and on the consequences of the treatment that leachate undergoes."

Climate breakdown is likely to exacerbate pollution from landfills, according to Prof Kate Spencer from Queen Mary University of London. Particularly "for historic landfills that are not lined these PFAS chemicals can enter surface and groundwaters with potential consequences for ecological and human health. This is likely to increase as the severity and frequency of flooding increases", she said.

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Letter

Landfill Gas: A Major Pathway for Neutral Per- and Polyfluoroalkyl Substance (PFAS) Release

Ashley M. Lin, Jake T. Thompson, Jeremy P. Koelmel, Yalan Liu, John A. Bowden, and Timothy G. Townsend*

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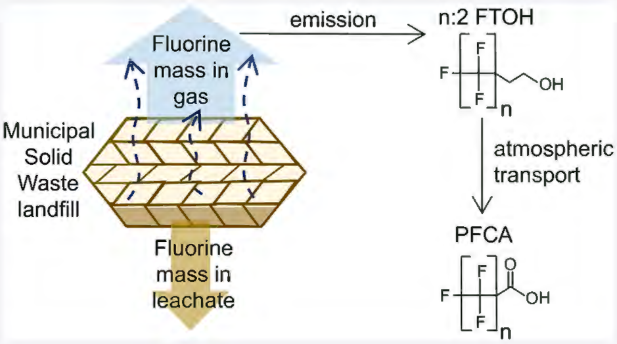
ABSTRACT: The undisclosed and ubiquitous use of perfluoroalkyl and polyfluoroalkyl substances (PFAS) in consumer products has led to a growing issue of environmental pollution, particularly within the solid waste community, where the fate of volatile (neutral) PFAS in landfilled refuse is not well understood. Here, three municipal solid waste landfills in Florida were assessed for neutral PFAS in landfill gas and ionic PFAS in landfill leachate to compare the relative mobility between the two pathways. Landfill gas was directly sampled using a high volume, XAD-2 resin based sampling approach developed for adsorption and analysis of 27 neutral PFAS. Across sites, 13 neutral PFAS were identified from fluorotelomer alcohol (FTOH), fluorotelomer olefin (FTO), secondary FTOH, fluorotelomer acetate (FTOAc), and fluorotelomer methyl acrylate (FTMAc) classes; however, FTOHs dominated concentrations (87–97% total neutral PFAS), with most detections surpassing utilized calibration levels. Even under conservative assumptions, the mass of fluorine leaving in landfill gas (32–76%) was comparable to or greater than the mass leaving in landfill leachate (24–68%). These findings suggest that landfill gas, a less scrutinized byproduct, serves as a major pathway for the mobility of PFAS from landfills.



KEYWORDS: *volatile, emissions, GC, fluorotelomer alcohol*

1. INTRODUCTION

Widespread per- and polyfluoroalkyl substance (PFAS) contamination has been a mounting environmental concern due to their chemical persistence and toxicity to human and biotic health.[1−4] While numerous industries are being confronted with PFAS-related management challenges, the burden of remediation and PFAS removal has often fallen on downstream industries—namely, the solid waste sector.[5−9] Discarded, PFAS-laden consumer products including textiles, wood products, and packaging and commonly landfilled industrial byproducts like MSW incineration ash and wastewater biosolids are known contributors to PFAS loading in landfills.[10−16] Existing research suggests most discarded PFAS mass is retained within landfills[9,17] with liquid-phase byproducts of waste decomposition, leachate and gas condensate, currently considered prevalent pathways for PFAS mobilization.[2,7,9] However, the extent of PFAS release to another major byproduct, landfill gas (LFG), has remained largely unscrutinized.

The bulk of PFAS characterization studies focus on nonvolatile/semivolatile (ionic) perfluoroalkyl acids (PFAAs) measured in liquid and solid matrices, in part because of a high presence and awareness of these species within the PFAS community but largely because analytical capabilities for ionic PFAS measurement are better established.[18−21] Volatile (neutral) PFAS are also utilized in consumer products[13,22−27,27] and have been determined in a few studies on ambient air surrounding landfills and near wastewater treatment plants,[28−32] but a lack of volatile analytical standards and latency in methodological development has hindered the progression of gas phase research in environmental matrices. Whereas PFAS characterization in leachate is established, concentrations ranging from thousands to tens of thousands of nanograms per liter are commonly encountered;[33−38] only two studies characterize volatile PFAS directly in LFG.[39,40] Titaley et al. identified fluorotelomer alcohol (FTOH), fluorotelomer acrylate (FTAc) and fluorotelomer olefin (FTO) homologues in LFG with combined concentrations ranging from 4,600 to 14,000 ng m^{-3} across three landfills. Goukeh et al., only assessing FTOHs, identified higher combined concentrations than Titaley et al., finding ~18,000 ng m^{-3} (sum of 6:2 and 8:2

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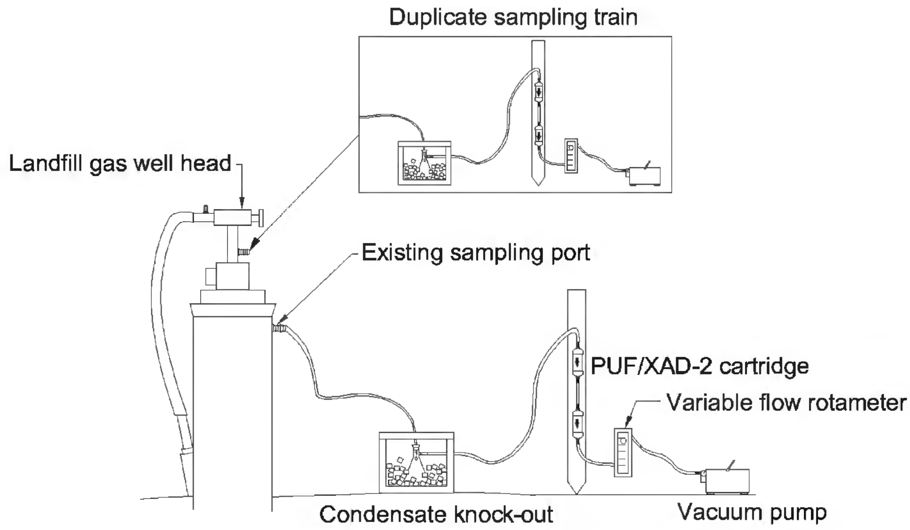

Figure 1. Developed system for sampling neutral PFAS directly from landfill gas well heads.

FTOH) in the one LFG sample examined. These studies suggest PFAS variability in LFG, which motivates further investigation, deploying higher sampling volumes[39] and larger analyte lists[40] to understand the potential presence of other neutral PFAS and distribution among landfills of different regions, compositions, and sizes.

With the ongoing development of PFAS regulation,[19] understanding the partitioning behavior of PFAS in major repositories like MSW landfills grows increasingly critical to minimize environmental and human risk. Unlike leachate, LFG is not always captured by collection systems, and management varies broadly across landfills, ranging from no treatment (i.e., passive venting) to some treatment (i.e., flaring, LFG to energy projects), but current treatment, if any, is not intended for PFAS.[41,42] Emerging research suggests the toxicity of volatile species (specifically 6:2 FTOH) to be significantly higher than their ionic counterparts via the inhalation pathway (a main route of exposure for volatile compounds).[43−47] Further, degradation of neutral species to ionic PFAAs once emitted to the atmosphere is well established.[48−58] The potential for long-range atmospheric transport of PFAS from landfills underscores the importance of considering neutral species and their fate during management to prevent further environmental contamination of highly scrutinized PFAAs such as perfluorooctanoic acid (PFOA) and perfluorooctanesulfonic acid (PFOS). As the only studies on LFG primarily identified FTOHs in LFG, the magnitude and significance of other neutral species remains unclear.

Here, LFG was sampled directly from gas well heads at three MSW landfill locations in Florida using a higher volume sampling protocol. XAD-2 resin sandwiched between polyurethane foam (PUF) was utilized for PFAS capture, then samples were analyzed for 27 volatile/semivolatile (neutral) PFAS via targeted gas chromatography high resolution mass spectrometry (GC-HRMS). To contextualize release in the gas phase, leachate was also collected at each landfill and analyzed for ionic PFAS (n = 93) using ultrahigh pressure liquid chromatography tandem mass spectrometry (UHPLC-MS/MS). The observed LFG and leachate concentrations were normalized on a mass of fluorine basis to compare the potential mobility in gas versus leachate matrices. This study provides foundational data critical for understanding the role of landfills in anthropogenic PFAS release and for informing LFG management.

2. METHODS AND MATERIALS

For brevity, materials and methods associated with ionic PFAS analysis in landfill leachate are provided in section 1 of the Supporting Information (Tables S-1 through S-4).

2.1. Standards and Reagents. Targeted neutral PFAS (≥97% purity, n = 27) were purchased from Wellington Laboratories Inc. (Guelph, ON, Canada), SynQuest Laboratories (Alachua, FL), and Chiron (Stiklestadveien, Trondheim, Norway). Nine classes of neutral PFAS (perfluoroalkane sulfonamides (FASAs), perfluoroalkane sulfonamidoethanols (FASEs), fluorotelomer acetates (FTOAcs), fluorotelomer methyl acrylates (FTMAcs), fluorotelomer iodides (FTIs), fluorotelomer secondary alcohols (sFTOHs), FTOHs, FTAcs, and FTOs) were measured using eight isotopically labeled internal standards (IS) from FASA, FASE, FTOH, and FTMAc classes for quantitation (Table S-5).

2.2. Sample Preparation and Collection. Polyvinyl chloride (PVC) cartridges filled with 4−5 g of Amberlite XAD-2 resin retained between two polyurethane foam (PUF) discs were utilized for PFAS capture.[29,59,60] Before use, XAD-2 sorbent was made PFAS-free through sequential Soxhlet extractions.[61] All cartridge components, sampling vessels, and tubing were sonicated in a mixture of Liquinox and PFAS-free water, rinsed, and then sonicated in methanol and methanol rinsed before use. Once dried and assembled, cartridges were stored in individually sealed polyethylene bags at 4 °C until sampling.

As neutral compounds were the focus of this investigation, aerosolized/particulate-bound PFAS were not specifically targeted for capture; however, a condensate collection system was included to prevent moisture interference. The developed sampling system (Figure 1) consisted of a condensate knockout (borosilicate, barbed Erlenmeyer flask contained in a cold box), two PUF/XAD-2 cartridges (installed in-series), a rotameter for flow control, a portable vacuum pump, and PFAS-free Tygon tubing. Before each sampling event, gas well head connection to the larger landfill gas collection system was disabled to create a neutral to positive pressure, workable for flow through the sampling system, then gas composition/

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Table 1. Average Concentrations (n = 2) of 13 Neutral PFAS (ng m^{-3}) from Three Municipal Solid Waste Landfills in Florida (Site Characteristics Are Provided in Table S-9)[a]

	concentration (ng m^{-3})					
analyte	landfill 1	E	landfill 2	E	landfill 3	E
4:2 FTOH	220		ND		57	
6:2 FTOH	*>9,900*	**170,000**	*>6,000*	**22,000**	*>6,500*	**62,000**
8:2 FTOH	*>6,800*	**200,000**	*>6,000*	**140,000**	*>6,500*	**740,000**
10:2 FTOH	*>5,100*	**14,000**	*>3,000*	**23,000**	*>5,000*	**120,000**
12:2 FTOH	860		1,400		5,000	
5:2 sFTOH	*>2,900*	**8,800**	*>1,700*	**9,000**	*>1,900*	**5,900**
7:2 sFTOH	320		*>1,300*	**13,000**	*>1,400*	**11,000**
8:2 FTO	2,500		1,300		550	
10:2 FTO	650		840		540	
12:2 FTO	97		580		160	
8:2 FTOAc	610		90		490	
10:2 FTOAc	99		19		140	
6:2 FTMAc	3,800		56		150	

[a]Concentrations of 6:2, 8:2, and 12:2 FTOH and 5:2 and 7:2 sFTOH consistently exceeded the upper limit of developed calibration ranges; therefore, both a minimum concentration (assuming the highest calibration concentration) and a maximum extrapolated concentration are provided. Italicized values denote a minimum concentration. Column "E" presents average maximum concentrations. "ND" denotes non-detect measurements. FTAcs, FASAs, FASEs, FTIs, and 8:2 FTMAc were not detected in any samples. Analyte acronyms and details are provided in Table S-5.

temperature was recorded using an Optimax Biogas analyzer (MRU Instruments, Humble, TX). Duplicate sampling trains were connected to existing gas well sampling ports. Approximately 1,200 L was sampled through each train at a flow rate of 5 L min^{-1}. After sampling, PUF/XAD-2 cartridges were sealed and individually stored at ≤4 °C for transport/storage. Quality control (QC) procedures are provided in the SI, section 2.

2.3. Extraction and Analysis. Spent XAD-2 from each cartridge was weighed and transferred to a 50 mL polypropylene centrifuge tube and vortexed, and approximately 2 g aliquoted for extraction. Samples were spiked with a mixture of mass labeled IS (Table S-5), rotated end-over-end for 18 h in 4 mL of 75/25% (v/v) ethyl acetate and methanol, and centrifuged for 10 min at 4,000 rpm. Supernatants were transferred to 15 mL centrifuge tubes, and the extraction process was repeated, combining supernatants from the two-fold extraction. Extracts were concentrated to 3 mL via gentle nitrogen evaporation, aliquoted, and stored no more than 30 days at −20 °C until analysis. QC details are provided in the SI, section 2 (Table S-6 and Figure S-1).

Targeted analysis of 27 neutral PFAS by positive chemical ionization (PCI) with selected ion monitoring (SIM) was conducted using a Thermo Scientific TRACE 1310 gas chromatograph coupled to a Thermo Scientific Orbitrap Exploris GC 240 mass spectrometer (GC-HRMS; see SI, section 2 for details regarding GC separations and instrumentation). A 12-point external calibration curve (from 1 to 2,000 pg μL^{-1}) was developed for quantitation, prepared through serial gravimetrically derived dilutions of primary stock solutions. A mixture of mass labeled IS at concentrations of 150 pg μL^{-1} was added to each calibration level. When a labeled standard was not available for a compound, a labeled standard with a similar retention time or structure was utilized for quantitation (Table S-5).

3. RESULTS AND DISCUSSION

Unexpectedly, several neutral PFAS concentrations in LFG exceeded the implemented calibration levels. Because of considerable exceedance for some compounds, dilution would reduce IS below instrument detection; therefore, in instances where sample concentrations exceeded calibration limits, two concentrations are presented (Equation S-1): a minimum value which assumes the highest calibration concentration and a maximum extrapolated concentration. Fluorine mass release calculations utilize minimum values, preventing overextrapolation while providing a conservative estimate for leachate comparison. Even under these assumptions, substantial concentrations of neutral PFAS, higher than those previously observed, were identified. Future assessments should deploy shorter sampling durations to refine findings.

3.1. Neutral PFAS in Landfill Gas. Except for 4:2 FTOH in one landfill, 13 PFAS were detected in duplicate samples across the three sites (site characteristics are provided in Table S-9). Observed concentrations are displayed in Table 1. At minimum, combined concentrations of neutral PFAS in LFG ranged from 22,000 to 33,000 ng m^{-3}. Considering extrapolated values, total concentrations ranged from 210,000 to 940,000 ng m^{-3}, an order of magnitude higher than those previously reported in LFG.[39]

3.1.1. FTOHs and sFTOHs in Landfill Gas. Like previous studies on LFG and air surrounding landfills, FTOHs dominated neutral PFAS concentrations;[28,31,32,39,40] however, extrapolated concentrations in this study surpassed previous reports in LFG, in some cases by 2 orders of magnitude, and were more comparable (although much lower) to concentrations recently identified in soil vapor near a PFAS manufacturing facility.[55] While there are uncertainties given the degree of extrapolation, the magnitude of FTOHs found in this study compared to existing research suggests fundamental differences potentially related to sampling methodology (e.g., much larger sampling volumes) and/or sampled landfill characteristics (e.g., waste type, age, air intrusion), although these data were not available for comparison. Across the three sites, 6:2, 8:2, and 10:2 FTOH, combined, made up 87 to 97% of total concentrations, but 8:2 FTOH alone constituted 50 to 79%. The shortest and longest analyzed homologues, 4:2 and

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12:2 FTOH, were significantly lower in concentration (Table S-10). This is supported by previous FTOH distributions determined from source fluoro-telomer polymers[62] and observations in LFG, urban air, and air surrounding wastewater treatment/landfill sites.[28−31,39,40,63] Concentrations of 12:2 FTOH were of similar magnitude to those in Titaley et al., but 4:2 FTOH has not been detected in LFG, suggesting MSW landfills to be a previously unidentified potential source of atmospheric 4:2 FTOH.[39]

Secondary FTOHs have not been targeted in gas-phase landfill research but have been identified in condensate associated with LFG collection systems.[37] As intermediary byproducts of 6:2 and 8:2 FTOH biodegradation to PFAAs, 5:2 and 7:2 sFTOH, were unsurprisingly elevated, they were at least an order of magnitude lower than respective parent FTOH homologues.[64−66] All detections of 5:2 sFTOH and two out of three detections of 7:2 sFTOH were above calibration, combined sums attributed to 2 to 10% of total concentrations.

3.1.2. Other Neutral PFAS in Landfill Gas. Other neutral PFAS fell within acceptable calibration ranges and together accounted for 0.22 to 1.9% of total concentrations. FTO homologues have been encountered in other LFG and ambient air studies, but in past assessments 8:2 and 10:2 FTO were below limits of quantitation and 12:2 FTO concentrations were consistently an order of magnitude higher than those reported here.[39,59] To the authors' knowledge, 8:2 and 10:2 FTOAc and 6:2 FTMAc have not been determined in LFG. FTOAcs are not commonly assessed analytes but are associated with fluoropolymer textile treatments and have been identified in one indoor air study from Japan.[67,68] Similarly, 6:2 FTMAc has only been analyzed in a few studies on cosmetics and wastewaters but at lower concentrations.[69−71]

3.2. Comparative Fluorine Mass Release between Landfill Byproducts. Normalizing PFAS concentrations on a fluorine basis allows comparisons to be drawn between different matrices and PFAS types (e.g., gas−liquid, neutral−ionic, precursor−terminal). This methodology is widely used to assess the "mass balance" of PFAS within systems, given that the long-term environmental fate of measurable PFAS is transient, whereas the mass of fluorine is conserved.[17,72,73]

Here, the same approach is utilized to compare the PFAS mobility in leachate versus LFG pathways. Neutral (Table 1, minimum values) and ionic (Table S-4) PFAS concentrations in LFG and leachate from this study were individually normalized to a mass of fluorine (Equation S-2) using compound specific fluorine mass fractions (Table S-8). Summed fluorine masses in leachate and LFG were then scaled according to site-specific annual generation volumes reported for each landfill (Table S-9).[41] A caveat of this comparison is the absence of measurements for neutral species in leachate and ionic species in LFG; however, the literature suggests FTOHs (the dominant neutral class identified) predominantly exist in the gaseous phase, while PFAAs exist in liquid or particulate phases.[28,74] Subsequent research should assess neutral and ionic compounds in both matrices to validate findings and further elucidate the PFAS behavior in landfills.

Even utilizing minimum concentrations observed in LFG, equal magnitudes of fluorine release are observed between LFG and leachate at each site (Figure 2)—contrasting from existing estimates of PFAS mass flow from landfills.[9] Existing

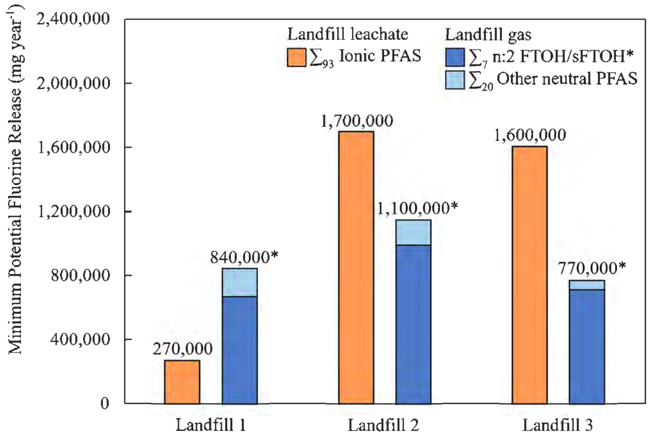

Figure 2. Annual fluorine mass release in landfill leachate versus landfill gas (LFG) from three municipal solid waste landfills in Florida. Fluorine masses in leachate are derived from ionic PFAS ($\sum_{93}$ PFAS) concentrations measured in leachate from each site (Table S-4) multiplied by the annual leachate generation volume and scaled using each detected compound's fluorine mass fraction (Tables S-8, S-9). The same methodology was applied for neutral PFAS ($\sum_{27}$ PFAS) in LFG by using the average of minimum concentrations (Table 1). Asterisked (*) values denote input FTOH/sFTOH concentrations which were above calibration levels developed for this study and therefore assumed to be at the highest calibration concentration. Consequently, these findings should be viewed as minimum values which conservatively estimate the magnitude of PFAS mobility in leachate versus LFG.

estimates, based on limited data, suggest that most PFAS mass mobilized from landfills releases through leachate (~62%).[9] However, our data from Landfill 1, showing over 76% fluorine release in LFG, along with substantial masses released by LFG in Landfills 2 and 3 (at minimum 40% and 32%, respectively), indicate that LFG may serve as an equal, likely greater, conduit of PFAS mobility from landfills than leachate, concurring with previous reactor studies on FTOH volatilization and neutral/ionic PFAS assessments of select waste materials.[17,75,76]

At least 79 to 92% of the fluorine mass in LFGs were derived from FTOH/sFTOH classes, with minimal contribution from FTOs, FTOAcs, and FTMAcs. In this conservative assessment, fluorine from LFG surpassed leachate in only Landfill 1. Although actual fluorine emission from LFG is higher than reported here, the elevated ratio of gas-to-leachate generation at Landfill 1 likely caused this difference (Table S-9). Landfill 2, the largest site, demonstrated the highest combined fluorine release from leachate and LFG, followed by Landfill 3, and then Landfill 1, corresponding to descending waste mass in place at each location.

4. IMPLICATIONS

This study provides fundamental data about neutral PFAS in LFG from MSW landfills. Unexpectedly, FTOH/sFTOH detections in LFG from this study exceeded implemented calibration levels; subsequent research should deploy shorter sampling durations. Regardless, even under more conservative assumptions these findings suggest that LFG, largely unscrutinized for PFAS, contains similar or greater magnitudes of PFAS compared to leachate, mostly attributed to midlength FTOH homologues. As landfills can be viewed as unabating PFAS repositories, the significance of LFG management in mitigating the long-term, long-range atmospheric transport of

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neutral PFAS, and subsequently derived PFAAs, cannot be understated. Unlike landfill leachate, LFG collection systems (when in place) are not fully efficient, collecting an estimated ~50−70% of generated biogases.[77] Though this is a considerable collection efficiency of biogas and presumably neutral PFAS, management of captured LFG fractions varies globally, from no treatment to degrees of carbon filtration and thermal treatment (i.e., flaring, advanced renewable natural gas technologies). Because the feasibility of PFAS destruction through thermal treatment remains unclear, research is needed to determine the treatment/removal efficiency of existing LFG management technologies. Considering the range of LFG capture efficiency, the retention and emission of neutral PFAS via fugitive emissions (i.e., migration through the waste layer) should also be examined, along with the role of landfill waste type, age, and temperature in neutral PFAS variability.

■ ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acs.estlett.4c00364.

> Acronyms, structures, and instrumental parameters for neutral PFAS via GC-HRMS and ionic PFAS via LC-MS/MS; detailed QC information and results; and fluorine mass balance/extrapolation details and assumptions (PDF)

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Notes

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Full length article

Unveiling PFAS hazard in European surface waters using an interpretable machine-learning model

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ABSTRACT

Per- and polyfluoroalkyl substances (PFAS), commonly known as “forever chemicals”, are ubiquitous in surface waters and potentially threaten human health and ecosystems. Despite extensive monitoring efforts, PFAS risk in European surface waters remain poorly understood, as performing PFAS analyses in all surface waters is remarkably challenging. This study developed two machine-learning models to generate the first maps depicting the concentration levels and ecological risks of PFAS in continuous surface waters across 44 European countries, at a 2-km spatial resolution. We estimated that nearly eight thousand individuals were affected by surface waters with PFAS concentrations exceeding the European Drinking Water guideline of 100 ng/L. The prediction maps identified surface waters with high ecological risk and PFAS concentration (>100 ng/L), primarily in Germany, the Netherlands, Portugal, Spain, and Finland. Furthermore, we quantified the distance to the nearest PFAS point sources as the most critical factor (14%–19%) influencing the concentrations and ecological risks of PFAS. Importantly, we determined a threshold distance (4.1–4.9 km) from PFAS point sources, below which PFAS hazards in surface waters could be elevated. Our findings advance the understanding of spatial PFAS pollution in European surface waters and provide a guideline threshold to inform targeted regulatory measures aimed at mitigating PFAS hazards.

1. Introduction

Owing to their resistance to heat, water, and oil, over 14,000 per- and polyfluoroalkyl substances (PFAS) are extensively utilized in various industrial and consumer applications, such as in nonstick cookware, firefighting foams, food containers, and anti-staining fabrics (Ackerman Grunfeld et al., 2024; Evich et al., 2022). However, throughout their life cycle, from manufacture to disposal, PFAS are released into various environmental media, including surface water, soil, and air (Bonato et al., 2025; Evich et al., 2022; Podder et al., 2021). For example, PFAS may enter surface water via industrial and municipal wastewater discharge (Huang et al., 2025; Salvatore et al., 2022). Given that surface water supplies half of all drinking water globally (FAO, 2024; Wang et al., 2023), it may represent an important pathway of PFAS exposure in both humans and organisms. Owing to the ubiquity of PFAS in aquatic environments, global concerns regarding their persistence, mobility, and potential for adverse health impacts are increasing (Cousins et al., 2022; Park et al., 2024; Sims et al., 2022).

PFAS are associated with adverse health effects in humans and wildlife, such as kidney cancer, infertility, liver effects, and altered immune function (Chiriac et al., 2023; Cordner et al., 2024; Steenland et al., 2010). In response to the potential risks posed by PFAS, the European Union has proposed guidelines for PFAS concentrations in drinking water, setting limits of 500 ng/L for the sum of all PFAS or 100 ng/L for the sum of 20 selected PFAS (Cappelli et al., 2024; EU, 2020). Considerable efforts have also been made to investigate the occurrence of PFAS in European surface waters, primarily at the regional scale (e.g., the Danube River (Ng et al., 2022) and the River Rhine (Möller et al., 2010)), while investigations at the national (e.g., Germany (Göckener et al., 2023)) level are limited. Recently, “the Forever Pollution Project”

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compiled a large dataset from various sources, recording 265,621 measurements of PFAS concentrations in surface waters across Europe (Cordner et al., 2024). However, this dataset is primarily descriptive (Moghadasi et al., 2023), and a considerable portion of European surface waters lacks records. To date, the spatially explicit patterns of PFAS risk in continuous surface waters across Europe, as well as the key spatial drivers underlying these patterns, remain largely unexplored.

Generating a high-resolution map of the PFAS risk in European surface waters can both provide important insights into the underlying risk patterns and inform targeted mitigation strategies. Despite its importance, comprehensive continental monitoring of PFAS in surface waters remains challenging because of the substantial costs and technical complexities involved. Machine-learning modelling offers a promising approach for predicting the PFAS risk in surface waters where monitoring data are scarce, owing to its excellent ability to establish nonlinear relationships between known contamination data and relevant environmental parameters (Chen et al., 2025; Chen et al., 2024; Podgorski and Berg, 2022). Recent modelling practices have demonstrated its application to the mapping of PFAS risks in Chinese surface water (Hu et al., 2023; Huang et al., 2025), US surface water (Breitmeyer et al., 2024) and groundwater (Park et al., 2024; Tokranov et al., 2024), as well as European soil (Moghadasi et al., 2023). In this study, we hypothesized that an interpretable machine-learning model could accurately predict PFAS concentrations, as well as the associated risks, in European surface waters, while also identifying the key spatial drivers underlying these patterns.

To test this hypothesis, we first compiled data on the concentrations of 20 selected PFAS at 9,985 surface water sites in 32 European countries (Fig. S1) and then calculated the cumulative ecological risk of PFAS at these sites by using the risk quotient (RQ) method (Bureau, 2003). Subsequently, two interpretable eXtreme Gradient Boosting (XGBoost) models were developed by establishing nonlinear relationships between the PFAS data and 20 relevant environmental parameters. These two XGBoost models allowed us to generate the first detailed maps of the total concentration and ecological risk of PFAS in European surface waters at a spatial resolution of 2 km. Furthermore, the concentration map was utilized to estimate the population affected by surface waters with PFAS levels exceeding the European Drinking Water guideline of 100 ng/L. Finally, the interpretable XGBoost algorithm was applied to identify the key influencing factors and threshold distance of PFAS point sources, so as to mitigate PFAS hazards in European surface waters.

2. Materials and methods

2.1. PFAS concentration

The following 20 PFAS, listed in the European Drinking Water Directive 2020/2184 (EU, 2020), were selected: perfluorobutanoic acid, perfluoropentanoic acid, perfluorohexanoic acid, perfluoroheptanoic acid, perfluorooctanoic acid (PFOA), perfluorononanoic acid, perfluorodecanoic acid, perfluoroundecanoic acid, perfluorododecanoic acid, perfluorotridecanoic acid, perfluorobutane sulfonic acid, perfluoropentane sulfonic acid, perfluorohexane sulfonic acid, perfluoroheptane sulfonic acid, perfluorooctane sulfonic acid (PFOS), perfluorononane sulfonic acid, perfluorodecane sulfonic acid, perfluoroundecane sulfonic acid, perfluorododecane sulfonic acid, and perfluorotridecane sulfonic acid.

Data on the PFAS present in surface waters across Europe were retrieved from the "Forever Pollution Project" (https://foreverpollution.eu/) (Cordner et al., 2024). To assess the occurrence of the 20 PFAS in European surface waters and to minimize possible biases, we applied the following selection criteria and statistical analysis: (i) only surface water sites located in continental Europe were included, excluding those on islands or in seas; (ii) the PFAS concentrations in the most recent year were used to reflect the current pollution level, where sites were sampled over multiple years; (iii) PFAS concentrations above the limit of quantification (LOQ) were recorded and assigned a value of 0 in case of the concentrations reporting as "below LOQ"; and (iv) all PFAS concentration units were converted to ng/L. After employing these quality control procedures, a final dataset of 20 PFAS from 9,985 sites in surface waters across 32 European countries was compiled for further analysis (Fig. S1).

2.2. Ecological risk assessment

We assessed the ecological risk of PFAS in European surface waters by using the common RQ method (Bureau, 2003). The RQ of each PFAS was calculated by comparing the measured concentration (MEC) in surface waters with the existing ecotoxicity thresholds (i.e., the predicted no-effect concentration [PNEC]), as follows (Rodrigues et al., 2024; Tang et al., 2025):

$$RQ = MEC / PNEC \tag{1}$$

$$PNEC = \min\ [(EC_{50} \text{ or } LC_{50}) / AF] \tag{2}$$

where EC_{50} and LC_{50} are the median effective and lethal concentrations for aquatic organisms at three trophic levels (i.e., algae, invertebrates, and fish), respectively, which were obtained from the USEPA ECOTOX Database (https://cfpub.epa.gov/ecotox/), as shown in Table S1. The assessment factor (AF) was set to 1,000 (Rodrigues et al., 2024). Accordingly, the individual RQ of each PFAS was calculated based on the most sensitive taxonomic group from three species (Rodrigues et al., 2024).

Given the similar mode of action of PFAS (Mu et al., 2022), the risk of PFAS mixture was evaluated by summing the RQs of all individual PFAS (RQ_{mix}), performed using the concentration addition model (Loewe and Muischnek, 1926). The ecological risk of PFAS mixtures was classified into the following four levels based on the RQ_{mix} values: no risk ($RQ_{mix} < 0.01$), low risk ($0.01 < RQ_{mix} < 0.1$), moderate risk ($0.1 < RQ_{mix} < 1$), and high risk ($RQ_{mix} > 1$).

2.3. Preprocessing of variables for machine-learning models

To mitigate the differences in analytical precision among different PFAS data sources and focus on identifying the regions of concern, we developed two machine-learning classification models, with reference to previous studies (Lombard et al., 2024; Podgorski and Berg, 2020; Tokranov et al., 2024; Xiao et al., 2024) and our recent works (Chen et al., 2025; Chen et al., 2024). One model was developed to predict the four ecological risk levels (i.e., $RQ_{mix} < 0.01$, $0.01 < RQ_{mix} < 0.1$, $0.1 < RQ_{mix} < 1$, and $RQ_{mix} > 1$) of PFAS in European surface waters. The other model, which had four PFAS concentration classes (i.e., <1, >1 to < 10, >10 to < 100, and > 100 ng/L), was developed to map the PFAS pollution level in European surface waters, and to estimate the affected population. The boundaries for the four PFAS concentration classes were selected based on the regulatory thresholds, environmental pollution levels, and analytical limitations: (i) The European Drinking Water Directive 2020/2184 recommends a sum of 20 selected PFAS below 100 ng/L (Cappelli et al., 2024; EU, 2020); (ii) concentrations less than 10 ng/L are considered a low level of PFAS contamination, while levels exceeding 10 ng/L are of environmental concern, as categorized by the "Forever Pollution Project" (https://pdh.cnrs.fr/en/about/); and (iii) the average LOQ of the 20 PFAS was approximately 1 ng/L, according to the analytical method outlined in the Technical guidelines of the Directive from the European Parliament and of the Council (EU, 2024). A LOQ below 1 ng/L may increase the risk of false positive results (Ruffle et al., 2023).

Twenty predictor variables, representing established associations and serving as proxies for factors potentially influencing the spatial distribution of PFAS in surface waters, were selected (details in Text S1 and Table S2). These variables encompassed the climate, land use, soil, geographic, agricultural, and socioeconomic parameters associated with each site. The variables were further standardized using the Z-score method (Zhao et al., 2023). Additionally, they were converted into grid cells at a resolution of 2 km × 2 km, enabling the two machine-learning models to predict the PFAS concentration and ecological risk in grid cells without PFAS data.

2.4. Model development and evaluation

For the two models, four concentration or ecological risk levels, and the 20 variables in each grid cell were designated as the response and predictor variables, respectively. The entire dataset was partitioned into 10 spatial clusters by using the K-means clustering algorithm based on the spatial locations, with eight randomly selected clusters assigned to the training set (80%) and the remaining two clusters forming the test set (20%) used for model performance evaluation. To optimize the model hyperparameters, a 10-fold cross-validation method was employed, in which the training set was further divided into 10 clusters using K-means clustering. In each of the ten rounds, nine randomly selected clusters were used for the training subset, while the remaining cluster served as the validation subset. This spatial partitioning strategy was used to mitigate potential spatial biases by ensuring that nearby locations were not simultaneously included in either the training or the test sets.

Based on a comparison of the performances of four common machine-learning algorithms (XGBoost, feedforward neural network, support vector machine, and K-nearest neighbor), XGBoost was selected as the final model owing to its superior performance (Table S3-S4). Model performance was assessed by the accuracy, sensitivity, specificity and the Area Under the ROC (receiver operator characteristic) curve (AUC). The hyperparameters of the two final XGBoost models were: n_estimators, max_depth, learning_rate, min_child_weight, gamma, subsample, scale_pos_weight and colsample_bytree (Table S5).

The two final XGBoost models were applied to the prediction dataset to predict the total concentration and ecological risk levels of the 20 PFAS in European surface waters at a 2-km spatial resolution. The grid map of European surface waters was downloaded from Natural Earth Database (https://www.naturalearthdata.com/). The machine-learning models were performed using the sklearn package in Python 3.11.5.

2.5. Model interpretation and uncertainty analysis

The XGBoost models were interpreted using the SHapley Additive exPlanations (SHAP) method, which evaluates the contribution of each predictor variable and calculates the marginal effect of predictor variables on a model's predictions. A positive SHAP value indicates a positive contribution, whereas a negative value indicates a negative contribution (Chen et al., 2025). Furthermore, we applied a generalized additive model (GAM) (Chen et al., 2022; Jia et al., 2024) to analyze the relationships between the predictor variables and their corresponding SHAP values. The SHAP analysis was performed using the shap package (Lundberg et al., 2020) in Python 3.11.5. The GAM fitting was developed based on the LinearGAM package (Daniel et al., 2018) in Python 3.11.5.

To perform an uncertainty analysis of the model predictions, a Monte Carlo simulation approach was employed (Zhao et al., 2023). The simulation involved 100 iterations, where different training sets were generated using a resampling technique. For each iteration, the XGBoost model was trained on a resampled training set and then used to predict the prediction dataset. The standard deviations of the predictions in each grid cell across all iterations were computed to quantify the uncertainty. The average value of the standard deviation for each country was further calculated to assess the impact of uneven PFAS sites (Fig. S1) on model uncertainty. Finally, uncertainty maps of the predictions of the two models were generated based on the calculated standard deviation at a resolution of 2 km.

2.6. Estimation of population affected by PFAS exceedance in surface waters

As previously mentioned, a value of 100 ng/L for the sum of the 20 selected PFAS is the recommended European Drinking Water guideline (Cappelli et al., 2024; EU, 2020). We defined grid cells with predicted PFAS concentrations exceeding a threshold of 100 ng/L as excessive regions. To estimate the population affected by surface waters with PFAS concentrations exceeding 100 ng/L, we calculated the affected population residing in excessive regions by multiplying the total population by the model probability and the proportion of domestic drinking water consumption from untreated surface water. Country-level data on the proportion of domestic drinking water consumption sourced directly from untreated surface water bodies, such as rivers, dams, lakes, ponds, streams, canals, and irrigation canals, were downloaded from the JMP database (https://washdata.org/monitoring/drinking-water). Owing to the lack of data on the proportion of domestic drinking water consumption from untreated surface water in each grid cell, we assumed that this data was uniform within a country. Data on the European population density in the year 2020 at 1-km resolution were obtained from the GPWv4 dataset (CIESIN, 2018). Accordingly, we calculated the affected population living in excessive regions by country, as follows:

$$\text{people}_{\text{total}} = \text{people}_{\text{urban}} + \text{people}_{\text{rural}} \tag{3}$$

$$\text{people}_{\text{urban}} = \text{density}_{\text{urban}} \times \text{perc}_{\text{urban}} \times pro \tag{4}$$

$$\text{people}_{\text{rural}} = \text{density}_{\text{rural}} \times \text{perc}_{\text{rural}} \times pro \tag{5}$$

where $\text{people}_{\text{total}}$, $\text{people}_{\text{urban}}$, and $\text{people}_{\text{rural}}$ represent the total, urban, and rural potentially affected populations, respectively. $\text{density}_{\text{urban}}$ and $\text{density}_{\text{rural}}$ are the population densities in the urban and rural regions in 2020, respectively. $\text{perc}_{\text{urban}}$ and $\text{perc}_{\text{rural}}$ are the urban and rural proportions of domestic drinking water usage from untreated surface water, respectively. *pro* is the predicted probability of PFAS concentration exceeding 100 ng/L by the XGBoost model.

3. Results and discussion

3.1. Occurrence of PFAS at the surface water sites

Our *meta*-analysis compiled a dataset comprising 25,801 concentration records for 20 PFAS at 9,985 surface water sites in 32 European countries (Figs. S1-S2). The PFAS levels, predominantly measured within the past five years (2020–2024) and accounting for 70.4% of the total data (Fig. S3), provided preliminary insights into the current status of PFAS contamination in European surface waters. Nevertheless, these sites exhibited an uneven spatial distribution, with a concentration in Western (e.g., France and the United Kingdom) and Southern Europe (e.g., Italy) (Figs. S1-S2). Of the 9,985 sites, 31.2% either did not detect any PFAS or had levels below the quantification limit. In addition, PFAS mixtures were present at 4,725 (47.3%) of the 9,985 sites, with a maximum of 18 detected PFAS (Fig. S4). Among the 20 PFAS, PFOS had the highest detection frequency (54.3%), followed by PFOA (37.2%) (Fig. S5). Previous *meta*-analysis has also reported PFOA and PFOS as the most commonly detected PFAS, with detection frequencies of 81%–90% in global surface waters (Sims et al., 2022). This was likely attributed to their widespread historical production and use in applications such as waterproof coatings and firefighting foams, as well as their designation as priority analytes in PFAS monitoring studies (Muir and Miaz, 2021; Pistocchi and Loos, 2009).

3.2. Prediction map of PFAS concentration in surface waters

Generating a high-resolution map of the PFAS concentration in European surface waters is essential for understanding PFAS pollution patterns and identifying priority areas requiring intervention. Our study developed an XGBoost model to predict the total concentration of the 20 PFAS in European surface waters at a 2-km resolution (Fig. 1). The model exhibited a high predictive performance, with an accuracy, AUC, sensitivity, and specificity of 0.92, 0.99, 0.92, and 0.87, respectively (Fig. 1a and Table S3). These metrics indicate the capability of the model to accurately forecast unknown PFAS concentration patterns. The uncertainty of the prediction map was evaluated by calculating the standard deviation in each grid cell, which exhibited a median, average, and maximum of 0.13, 0.13, and 0.20, respectively. Despite the uneven geographic distribution of PFAS sites across European countries (Fig. S1), the predictive performance of the model remained consistent and satisfactory. This was evidenced by the fact that the average standard deviation values by country ranged from 0.10 to 0.15, close to the overall average of 0.13 (Fig. S6).

The PFAS concentration prediction map (Fig. 1b) showed that approximately 63% of European surface waters exhibited PFAS concentrations below 10 ng/L, suggesting a generally low level of PFAS contamination across the continent. Notably, 37% of European surface waters were predicted to have PFAS concentrations of environmental concern (>10 ng/L), primarily located in regions such as Spain, Germany, Romania, Ukraine, and Serbia.

To identify priority regions, that is, regions most urgently requiring water quality management, we defined regions with PFAS concentrations exceeding the European Drinking Water guideline of 100 ng/L as "excessive regions". Excessive regions were located predominantly in Germany and the Netherlands (e.g., the Rhine River and its tributaries), Portugal (e.g., the Sorraia and Sado Rivers), Spain (e.g., the Genil, Zújar, and Guadalimar Rivers), and Finland (e.g., the Kitinen River) (Fig. 1b). Compared to regions with low PFAS levels, excessive regions with high PFAS concentrations were generally located closer to PFAS sources, had higher population densities, and underwent more socioeconomic activity (Fig. S7). For instance, the Rhine River, one of Central Europe's largest waterways with a dense population, suffers from severe PFAS contamination caused by substantial wastewater discharge from over 2,800 treatment plants, as well as intensive industrial activities, including those of nearly 10% of global chemical industries (Li et al., 2023) and three PFAS production facilities (Fig. S8).

To the best of our knowledge, this study is the first to perform a model prediction of PFAS concentrations in European surface waters, providing a data-driven foundation for understanding PFAS contamination patterns across Europe. The predictions derived from our model were generally consistent with the findings obtained from previous regional-scale monitoring efforts. For instance, a previous investigation reported that the concentrations of 26 target PFAS at 40 river sites along the Swedish coast were 1–60 ng/L (Nguyen et al., 2017), which is consistent with our predicted concentration ranges below 100 ng/L (Fig. 1b). Additionally, a *meta*-analysis found that the PFAS concentrations were below 10 ng/L in most European waters (Domingo and Nadal, 2019). An important advancement of our study is the identification of excessive regions, for which no monitoring data had been reported, such as the Sorraia and Sado Rivers in Portugal, and the Kitinen River in Finland (Fig. 1b). For individuals residing in these excessive regions, we recommend both avoiding the direct consumption of surface water and switching to purified bottled water. Our prediction map, particularly for excessive regions, can serve as a guide for raising awareness for public consumption, future monitoring, and PFAS removal options tailored to local surface-water conditions.

3.3. Population affected by PFAS exceedance in surface waters

To assess the potential impact of PFAS contamination in surface waters on humans, we quantified the number of individuals affected by surface waters with PFAS concentrations exceeding the European Drinking Water guideline of 100 ng/L. In 29 of the 44 European countries, we estimated that approximately 7,749 individuals were affected by surface waters with PFAS concentrations exceeding 100 ng/L. Geographically, the affected populations were concentrated in Central and Southwestern Europe (Fig. 2a). Germany was the predominant contributor, accounting for up to 28.74% (Fig. 2b), followed by Spain (19.87%), the Netherlands (14.31%), and France (12.94%). The remaining 25 countries collectively contributed to less than 25% of the total affected population.

3.4. Ecological risk of PFAS in surface waters

Of the 9,985 sites, PFAS posed a potential ecological risk at 14% of the sites (Fig. S9). Specifically, PFAS posed no risk at most sites (86.39%), followed by low (11.18%), moderate (1.88%), and high (0.55%) risks. Notably, the above analysis results were highly dependent on existing monitoring data. Given both the uneven distribution of PFAS and the lack of PAFS measurements in numerous surface waters (Fig. S9), site-specific risk assessments may not comprehensively capture the PFAS risks in European surface waters.

To investigate the spatially explicit patterns of ecological risks posed by PFAS in European surface waters, we developed an XGBoost model using known PFAS risk data and 20 environmental spatial variables. This

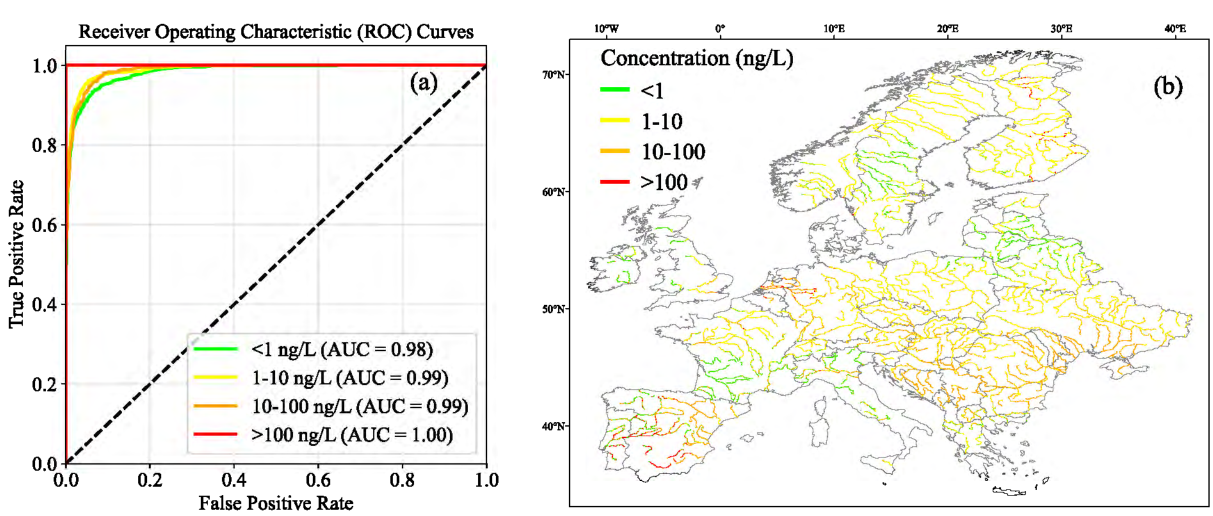

Fig. 1. European mapping of PFAS concentrations in surface waters. (a) The area under curve (AUC) of the receiver operator characteristics (ROC) in an XGBoost model. A higher AUC value indicates a better ability of this XGBoost model to distinguish the four concentration levels of PFAS. (b) The map of total concentration of 20 PFAS in European surface waters with prediction results by the XGBoost model.

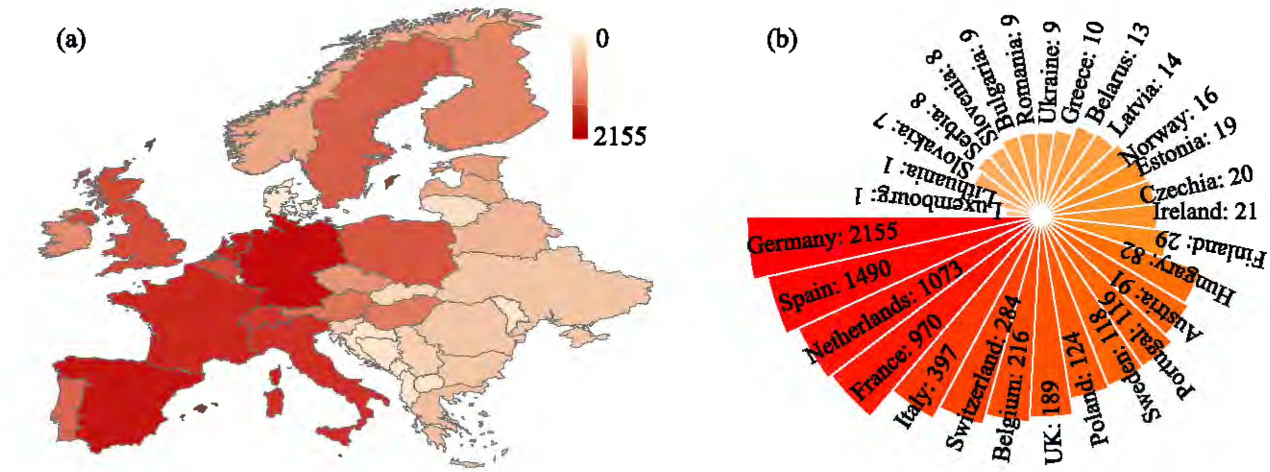

Fig. 2. Population by country affected by surface waters with PFAS concentrations exceeding the European Drinking Water guideline of 100 ng/L. Geographical distribution (a) and national ranking (b) of affected populations.

model performed very well, as evidenced by its accuracy, AUC, sensitivity, and specificity of 0.97, 0.93, 0.94, and 0.93, respectively (Fig. 3a and Table S3). In addition, the uncertainty in the model prediction was evaluated by calculating the standard deviation for each grid cell. The standard deviation values ranged from 0 to 0.19, with average and median values of 0.06 and 0.04, respectively, suggesting low uncertainty in the model (Fig. S10). Although the average standard deviation varied among countries, ranging from 0.03 to 0.135, this was not significantly related to the number of sampling sites in each country (Fig. S10). Overall, a higher uncertainty was observed in grid cells with higher ecological risk levels, such as those in Finland and Spain. This finding was consistent with the results of previous machine-learning models applied to groundwater (Xiao et al., 2024) and soil (Zhao et al., 2023).

Using the developed XGBoost model, we successfully mapped the ecological risk levels of PFAS in European surface waters at a resolution of 2 km (Fig. 3b). The analysis results revealed that approximately 96% of the surface waters were predicted to have no ecological risk (i.e., $RQ_{mix} < 0.01$), which aligns with the results of previous regional monitoring studies conducted in the Llobregat River in Spain (Campo et al., 2015) and in the Danube River (Ng et al., 2022). Only 4% of European surface waters were predicted to be at potential ecological risk (i.e., $RQ_{mix} > 0.01$), primarily located in the Rhine River and its tributaries in Germany, the Sorraia and Sado Rivers in Portugal, and the Zújar, Genil and Guadalimar Rivers in Spain. Additionally, surface waters with the high ecological risk of PFAS (i.e., $RQ_{mix} > 1$) were identified in Eastern Finland, such as the Hossanj and Kemijoki Rivers. These surface waters with potential PFAS risks generally coincided with surface waters with high PFAS concentrations. Overall, the ecological risk of PFAS in European surface waters was relatively low, but surface waters with a high ecological risk of PFAS require further attention and enhanced monitoring.

3.5. Contributing factors

Understanding the effects of natural and anthropogenic factors on the spatial distribution of PFAS contamination in European surface waters is crucial for implementing targeted PFAS mitigation strategies. This study quantified the relative contributions of 20 factors by using the SHAP method (Fig. 4 and Fig. S11). In the two XGBoost models, the

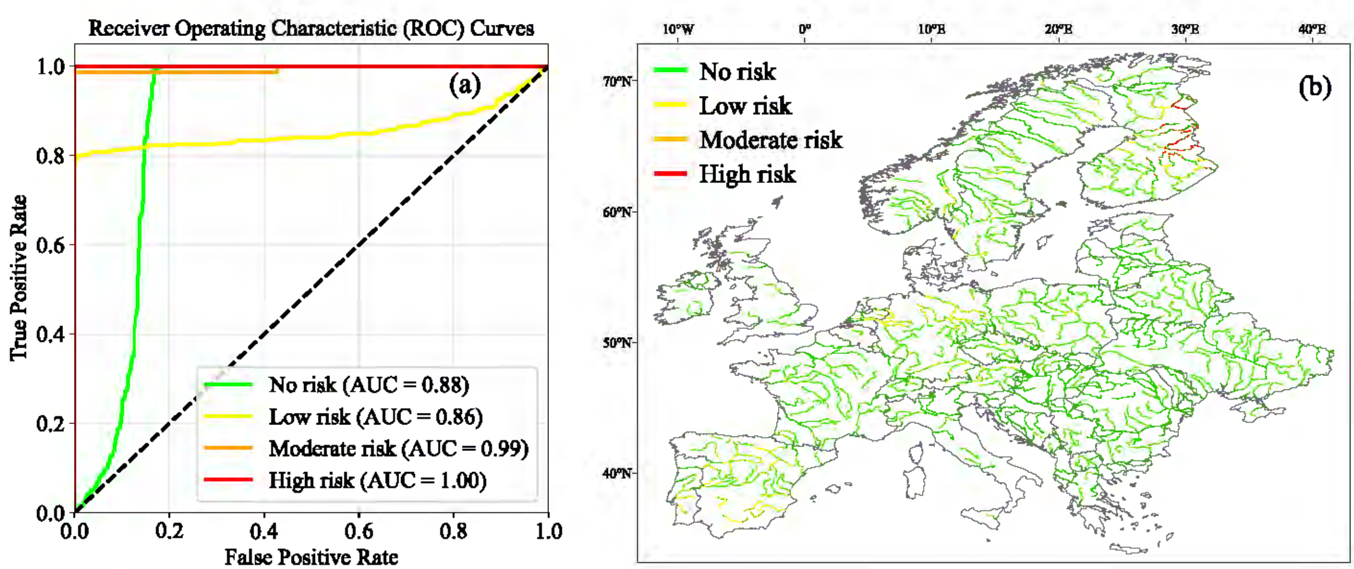

Fig. 3. European mapping of ecological risk of PFAS in surface waters. (a) The area under curve (AUC) of the receiver operator characteristics (ROC) in an XGBoost model. A higher AUC value indicates a better ability of this XGBoost model to distinguish the four ecological risk levels of PFAS. (b) The map of ecological risk of 20 PFAS in European surface waters with prediction results by the XGBoost model. The ecological risk of PFAS was divided into four levels based on the total risk quotient (RQ_{mix}): $RQ_{mix} < 0.01$, no risk; $0.01 < RQ_{mix} < 0.1$, low risk; $0.1 < RQ_{mix} < 1$, moderate risk; and $RQ_{mix} > 1$, high risk.

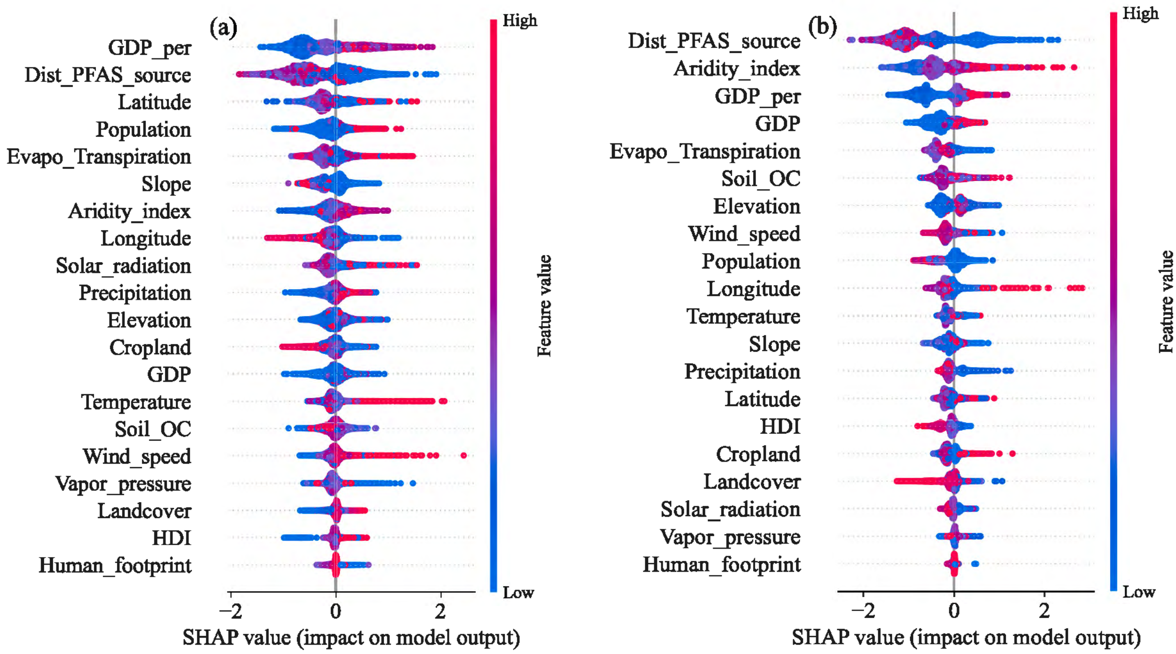

Fig. 4. Shapley additive explanation (SHAP) waterfall diagram of two XGBoost models for predicting total concentration (a) and ecological risk (b) of PFAS. SHAP values greater than 0 indicate a positive effect, and vice versa. The color of the point represents the magnitude of the variable value. Feature descriptions are provided in Table S2.

dominant influencing factors were socioeconomic parameters (41.0%–43.1%), including the distance to the nearest site potentially containing, using, or emitting PFAS, gross domestic product (GDP), GDP per capita, population, human development index, and human footprint parameters. This result was expected because population growth and increased socioeconomic activity are generally accompanied by an increased production and usage of PFAS (e.g., food containers and fire-suppressing foams) (Evich et al., 2022), thereby contributing to elevated PFAS emissions.

The second major contributing factor was climate-related conditions, including temperature, precipitation, wind speed, solar radiation, water vapor pressure, evapotranspiration, and the aridity index, which cumulatively contributed 28.4%–30.4%. These climatic conditions can significantly affect the environmental behavior and fate of PFAS in surface water, including their transport, storage, transformation, and dilution (Huang et al., 2025). Soil properties were another important factor, accounting for 12.6%–13.8%. The topographical features of elevation and slope can alter the volume, velocity, and direction of river flow (Sheikholeslami and Hall, 2023), thereby influencing the transport and fate of PFAS in surface water.

3.6. *Management for mitigating PFAS pollution*

Of the 20 factors, the distance to the nearest site potentially containing, using, or emitting PFAS (Dist_PFAS_source) was identified as the most critical factor in the two XGBoost models, accounting for 13.8%–18.5% of the model contributions (Fig. 4 and Fig. S11). Lower values of Dist_PFAS_source had positive SHAP values (Fig. 4), suggesting that the probabilities of a high PFAS concentration (i.e., >100 ng/L) and ecological risk (i.e., RQ_{mix} > 1) increased with decreasing Dist_PFAS_source value. This finding could be attributed to the tendency for sites in close proximity to PFAS sources (e.g., fire training facilities, metal-coating facilities, landfills, and waste treatment plants) to accumulate higher concentrations of these compounds. A recent study also recognized Dist_PFAS_source as a key factor driving the PFAS distribution in US groundwater (Tokranov et al., 2024).

To provide a scientific foundation for precise regulation, we further applied a GAM method to determine the relationship between Dist_PFAS_source values and the corresponding SHAP values, thereby exploring how Dist_PFAS_source influenced the PFAS concentration and ecological risk in surface waters (Fig. 5). The GAM analysis revealed that, when the Dist_PFAS_source values were below 4.1–4.9 km, the SHAP values for both XGBoost models were positive, indicating a positive effect. Conversely, when the Dist_PFAS_source values exceeded this threshold of 4.1–4.9 km, the SHAP values were negative, suggesting that the probabilities of a high PFAS concentration (i.e., >100 ng/L) and ecological risk (i.e., RQ_{mix} > 1) decreased with increasing Dist_PFAS_source value. Accordingly, we defined the critical distance (i.e., 4.1–4.9 km) as the tipping point at which Dist_PFAS_source transitioned from having a positive influence to having a negative influence. Taken together, we recommend that facilities with potential PFAS sources (e.g., PFAS production facilities, fire training stations, landfills, and wastewater treatment plants) be located at least 4.1–4.9 km away from surface waters to mitigate the adverse effect of PFAS contamination to humans and ecosystems.

4. Limitations and prospects

To the best of our knowledge, this study presents the first comprehensive assessment of the adverse impacts of PFAS contamination in European surface waters on human and ecosystem health. However, this study had several limitations and uncertainties that warrant further consideration. First, the PFAS concentrations and predictor datasets were obtained from diverse sources, an issue inherent in large-scale machine-learning studies (Chen et al., 2025; Podgorski and Berg, 2022; Tokranov et al., 2024; Xiao et al., 2024). Although we resampled all predictor datasets to a 2-km resolution and developed XGBoost classification models to mitigate the effects of varied data sources, the disparity in data quality might have still introduced uncertainties into the XGBoost models. Pursuing a more uniform data resolution and quality for comprehensive prediction should be a focus of future modeling studies. Second, the European Union has established two guidelines: the sum of 20 selected PFAS should not exceed 100 ng/L, and the sum of all PFAS should not exceed 500 ng/L (Cappelli et al., 2024; EU, 2020). Incorporating a broader range of PFAS into our assessment may significantly increase the number of regions with high ecological

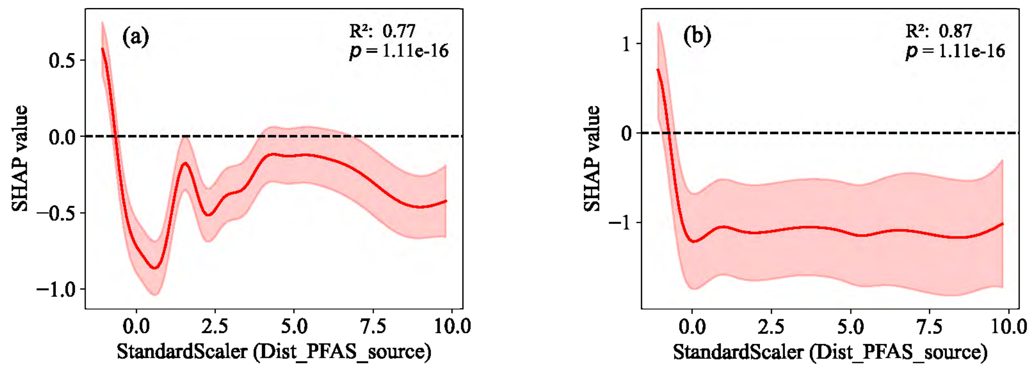

Fig. 5. Partial dependence plots of the Shapley additive explanation (SHAP) value against the standardized Dist_PFAS_source in two XGBoost models for predicting total concentration (a) and ecological risk (b) of PFAS. SHAP values greater than 0 indicate a positive effect, and vice versa. The red curves and pink shadings are the fitted lines and the 95% confidence intervals, respectively, using a generalized additive model. Dist_PFAS_source represents the distance to the nearest site potentially containing, using, or emitting PFAS. The value of Dist_PFAS_source was standardized using Z-score method. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

risks and affected populations. Third, the estimated affected populations were calculated by multiplying the total population by the model probability and the proportion of domestic drinking water consumption from untreated surface water at the country level, because of the lack of higher-resolution data on surface water usage (Podgorski and Berg, 2020; Xiao et al., 2024). Fourth, our assessment focused solely on surface water, without accounting for groundwater. Future studies should conduct a comprehensive assessment incorporating a wider range of PFAS and higher-resolution data on water usage, while considering both surface and groundwater. Finally, the distance to the nearest potential PFAS source site (i.e., Dist_PFAS_source) was used as a predictor variable in our XGBoost models. We did not calculate the distance to each individual type of PFAS point source, owing to the limited availability of data on many source types as model inputs. Future research should measure or collect more comprehensive data on different types of PFAS sources, such as PFAS production facilities, airports, fire training stations, landfills, and wastewater treatment plants. This would enable the determination of critical distance thresholds for each source type, which could inform more targeted regulatory measures to mitigate PFAS hazards in surface waters.

5. Conclusion

This study developed two XGBoost machine-learning models based on data on 20 PFAS from 9,985 surface water sites across Europe, as well as 20 relevant environmental parameters. By applying the two XGBoost models, we mapped the concentrations and ecological risk levels of PFAS in continuous European surface waters at a resolution of 2 km. These maps have implications for raising awareness of PFAS pollution in European surface waters, guiding future environmental monitoring and providing information on PFAS removal strategies tailored to local conditions. Furthermore, we estimated that nearly eight thousand individuals, living mainly in Central and Southwestern Europe, were affected by surface waters with PFAS concentrations exceeding the European Drinking Water guideline of 100 ng/L. Using a combination of the SHAP and GAM method, we identified Dist_PFAS_source as the most important contributor (13.8%–18.5%) influencing the PFAS concentration and ecological risk. We then examined the relationship between Dist_PFAS_source and SHAP values, which enabled us to determine a critical distance threshold (4.1–4.9 km). This threshold provides a scientific basis for decision-makers to precisely regulate potential point sources of PFAS (e.g., PFAS production facilities, landfills, and wastewater treatment plants), which should be located at least 4.1–4.9 km away from surface waters to safeguard humans and ecosystems.

CRediT authorship contribution statement

Li Zhao: Writing – original draft, Methodology, Investigation, Data curation. **Jian Chen:** Writing – review & editing, Supervision, Methodology, Investigation, Formal analysis, Conceptualization. **Jiaqi Wen:** Writing – review & editing, Formal analysis. **Yangjie Li:** Writing – review & editing, Methodology, Funding acquisition. **Yingjie Zhang:** Writing – review & editing, Formal analysis. **Qunyue Wu:** Writing – review & editing, Funding acquisition. **Gang Yu:** Writing – review & editing, Supervision.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.envint.2025.109504.

Data availability

Data will be made available on request.

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Dear Benton County Commissioners,

I oppose Coffin Butte landfill's expansion.
Clean air and water are precious vital resources for all of us in Benton County.
A feeling of ease in our communities because our elected officials consistently stand up for our safety, health and welfare, is vitally important.
Promises have been broken over the years by the owners and operators of Coffin Butte landfill. And our county has failed in its oversight.
The landfill expansion if approved weighs on our common resources to a great extent over an extended period of time, affecting our grandchildren and their children.

It's Wet Here

This post-war landfill is in a very wet part of a wet valley.
Adair Village gets 51 inches of rain, on average, per year.
At Arlington, an alternative regional landfill with large capacity and accessible by rail, the average annual rainfall is less than 9 inches.

There is a strong link between wet conditions and leachate production in landfills.
Every droplet of water that splashes down on an open landfill cell will slowly trickle through the trash, transforming into a concentrated liquid waste known as leachate.

Moisture encourages bacterial decomposition, which is the primary process for methane generation in landfills.

Landfill Gas

Billowing tarps, tears and odors indicate the release of methane and landfill gasses into our air. These gasses also contain PFAS. "According to an EPA-funded study recently published in the peer-reviewed Environmental Science and Technology Letters, PFAS could be escaping landfills via gas at concentrations similar to — if not higher than — liquid leachate."
I have attached the research paper entitled *Landfill Gas: A Major Pathway for Neutral Per- and Polyfluoroalkyl Substance (PFAS) Release.*

In the paper, researchers noted that they have “detected “unexpectedly” high levels of PFAS in landfill gas, adding to a growing body of evidence on how “forever chemicals” leave waste sites.”

PFAS Forever Chemicals

All over the world, PFAS in landfills are growing. In Europe and the UK research has been forward thinking and robust. I cite here a Guardian article from November 4, 2024 telling of a project seeking to remove PFAS forever chemicals from leachate that contaminates groundwater and surface water - and can cause health problems, including kidney and testicular cancer.

“Processes intended to decontaminate noxious liquid landfill waste before it enters rivers and sewers have been found to increase the levels of some of the worst toxic chemicals, a study has shown.

Landfills are well known to be a main source of PFAS forever chemicals – or per- and polyfluoroalkyl substances – but the new study shows that the treatment plants designed to clean up the liquid waste can instead boost the levels of banned PFAS such as PFOA and PFOS, in some cases by as much as 1,335%.”

https://www.theguardian.com/environment/2024/nov/04/decontamination-of-landfill-waste-leads-to-increase-in-toxic-chemicals-says-study

Currently, PFAS are ubiquitous in surface waters- and that means the Willamette River, into which untold numbers of gallons of PFAS containing landfill leachate from Coffin Butte have been released untreated after being transported to the Corvallis and Salem water treatment plants.

Solutions do not exist to “treat” PFAS forever chemicals. It behooves us to lessen the amount of toxic leachate in our region by not approving the expansion of Coffin Butte in this very wet part of the Willamette Valley.

Local and global concerns regarding the persistence of PFAS, how they move through the environment, and the potential for adverse health impacts of PFAS are increasing. Here in Benton County, we have the ability to make a decision in order to safeguard the health of our population and our natural resources.

A Big Liability

The Coffin Butte owners and operators have not been good stewards. They have not been good partners. There are many incidences of violations and mishandling of the confidence and trust placed in them.

Coffin Butte landfill is not a resource to us - rather it is a mountainous and growing liability and a source of real health and environmental concerns.

The landfill's expansion would further impinge upon our rights to our health and our finite natural resources of clean air and water. There are consequences of leachate that percolates into groundwater, or that is disposed of in the Willamette River. Consequences of PFAS that burp into the air along with landfill gasses. Those PFAS forever chemicals are percolating into our bodies and natural environments.

From Politico, October 2025- *A group of 24 European politicians whose blood was tested for toxic PFAS chemicals over the summer all had the substances in their bodies, the NGOs involved in the testing revealed Tuesday.*
"I tested positive for four substances, and three of them can harm unborn children, act as endocrine disruptors, cause liver damage, and are suspected of being carcinogenic," said Danish Environment Minister Magnus Heunicke in a written statement, describing his results as a "frightening reality."

PFAS in our environments are ubiquitous locally and globally. "Owing to their resistance to heat, water, and oil, over 14,000 per - and polyfluoroalkyl substances (PFAS) are extensively utilized in various industrial and consumer applications, such as in nonstick cookware, firefighting foams, food containers, and anti-staining fabrics."

Please oppose this expansion.
Thank you for your diligence.

Susan Walenza
1415 NW Greenwood Place
Corvallis

PFAS in the body

PFAS in Surface Water

Landfill Gas- Major Pathway for PFAS Release

Decontamination of Leachate Leads to Increase in Toxic Chemicals