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Occurrence of PFAS in Lyon soils: contribution and impact of industrial activities

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As the environmental impact of per- and polyfluoroalkyl substances (PFAS) increases, understanding their deposition to soils near chemical industries is vital to mitigate health and ecological risks. Lyon (France) is a hub of chemical production and faces concerns about PFAS releases into the environment. This study assessed the extent of PFAS deposition from chemical industries located in central Lyon to different parts of Lyon. Through a target analysis using UHPLC-MS, we investigated the concentration of 80 PFAS in 215 Lyon soil samples collected around a fluoropolymer industrial complex. PFAS contamination of eggs in Lyon has been reported by local public health; hence, we evaluated PFAS in soils from a few selected chicken-feeding areas and free-range chicken eggs. High PFAS concentrations (3.8–175 $\mu\text{g kg}^{-1}$) were observed in soils near fluorochemical industries, with levels declining as distance from the industrial site increased. Long-chain PFCAs ($C \geq 8$) and PFSAAs ($C \geq 6$) showed high detection rates (78–100%), with PFUnDA (0.05–106 $\mu\text{g kg}^{-1}$), PFTrDA (0.03–47.6 $\mu\text{g kg}^{-1}$), PFOS (0.1–32.6 $\mu\text{g kg}^{-1}$), PFHxS (0.03–23.7 $\mu\text{g kg}^{-1}$), PFNA (0.02–13.4 $\mu\text{g kg}^{-1}$) and PFOA (0.05–6.8 $\mu\text{g kg}^{-1}$), being the predominant PFAS. The $\sum 14$ PFAS detected in soils from chicken-feeding areas ranged from 3.3–10.5 $\mu\text{g kg}^{-1}$ and 2.1–19.1 $\mu\text{g kg}^{-1}$ in eggs. This confirmed that the chickens are exposed to PFAS from the surrounding soils and other sources through their diet and foraging. Generally, the detection of elevated levels of PFNA, PFUnDA, and PFTrDA suggests an industrial input from the production of Surflon® at the Pierre-Bénite site. A significant negative correlation (p -value < 0.001) was observed between PFAS concentrations across the zones and their distance from the chemical industry. The variability in PFAS distribution may be influenced by wind direction, which has likely transported airborne PFAS from its source near the industrial complex and extending towards other parts.

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Environmental significance

This study provides clear evidence of ambient PFAS deposition in a dataset of 215 Lyon soils, with contamination levels changing based on proximity to a major chemical industrial site using and producing PFAS. The observed soil PFAS concentrations are associated with prevailing wind patterns, thus indicating a direct atmospheric deposition of PFAS to Lyon soils.

1 Introduction

The ubiquitous detection of per- and polyfluoroalkyl substances (PFAS) in environmental matrices worldwide is predominantly due to accelerating industrial activities. As the environmental footprint of PFAS continues to grow, understanding ambient PFAS deposition to soils near industrial sites is vital to mitigate both ecological and human health risks. PFAS are a group of synthetic compounds known for their exceptional stability, durability and functionality in extreme conditions, driving their extensive use across several industrial sectors.^{1,2}

Glüge *et al.* identified more than 200 industrial uses for more than 1400 individual PFAS.² These uses were found in various industries, including chemical, aerospace engineering, biotechnology, building and construction, energy sectors, mining, machinery and equipment, nuclear, photographic, textile, wood, automotive, pharmaceutical, and oil and gas.² Chemical industries have a vast use for PFAS, ranging from the production of plastics and rubbers, coatings, paints and varnishes, fire-fighting foams, pesticides, cosmetics and personal care products, floor coverings including carpets and floor polish, paper and packaging, pipes, pumps, fittings, and liners, among others.^{1–3}

The exceptional properties of PFAS that promote its use within these industries include hydrophobic and oleophobic properties, low surface tension and the ability to lower the

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surface tension of liquids, low refractive index, non-reactivity, long-term stability without degradation, non-flammability and high-temperature durability.^{2,4}

PFAS exhibit remarkable stability, which enables them to resist degradation processes in natural environments, leading to their accumulation across various environmental matrices, including air,^{5,6} water,^{7,8} and sewage sludge/biosolids,^{9–12} soils^{13–15} and crops.^{16–18} Consequently, PFAS have become a notable environmental contaminant, detected even in isolated regions distant from their initial sources.^{19–21} PFAS persistence and bioaccumulative nature have raised concerns about their potential to negatively impact ecosystems and human health.^{3,22}

PFAS have been widely detected in human samples, raising concerns about their long-term health impacts.^{23–26} Several studies have linked PFAS exposure to a range of severe health conditions, including kidney and testicular cancers, liver damage, developmental toxicity, reproductive disorders, endocrine disruption, increased cholesterol levels, ulcerative colitis, thyroid disorders, and immune system impairments.^{27–32} These findings highlighted the potential risks of PFAS accumulation in the human body and the importance of further research and regulatory measures to reduce exposure.

Recent studies have revealed the role of airborne emissions in the distribution of PFAS in the environment.^{5,33,34} When PFAS are released into the atmosphere through industrial contamination, they can travel significant distances before being deposited onto soils and vegetation. Ambient deposition plays a crucial role in the accumulation of PFAS in surface soils, particularly in remote areas.²⁰ Once PFAS are deposited, they can persist in the soils for an extended period, potentially bioaccumulating in crops and entering the food chain.^{34–37}

Airborne PFAS deposition to soils can occur through wet or dry deposition.¹ Wet deposition occurs primarily during precipitation events, such as rainfall or snowfall, whereas dry deposition involves the direct settling of particulate matter (PM) from the atmosphere onto soil surfaces. PFAS distribution in PM has been confirmed to be aerosol particle size-dependent,³⁸ with several studies confirming PFCA distribution in fine to ultrafine size fractions (peak at 0.5–1 μm PM size).^{39–43} However, PFOS has also been reported to be associated with coarse particles.⁴¹ Studies have confirmed PFAS contaminations in precipitations (wet deposition)^{3,34,44,45} and in atmospheric particulate matter (dry deposition)^{6,38,40,46,47} however, under normal circumstances, wet deposition has been confirmed to be the major pathway for atmospheric PFAS deposition to surface soils, estimated to be as much as 20 times higher than dry deposition.⁴⁶

Chen *et al.* investigated 22 PFAS compounds in precipitation samples collected from 28 cities across mainland China in their study. The study confirmed detection rates higher than 80% for 10 PFAS, including TFA, PFOA, PFOS, PFPrA, PFBA, PFHxA, PFHpA, PFNA, PFDA, and PFUnA. A higher PFAS flux was recorded in precipitations from cities with higher industrial activities (200–3400 ng per m^2 per day) compared to cities without industrial activities (63–1700 ng per m^2 per day).⁴⁴ These values were higher than PFAS daily reflux recorded in the US (1.3 to 47.4 ng m^{-2} per day).²

In a recent study, Berthoud *et al.* analyzed the levels of 52 PFAS compounds in rainwater collected from urban, rural, and coastal locations across France during autumn 2022.⁷ The study confirmed the dominance of PFNA (4.6 ± 1.4 – 31.0 ± 9.2 ng L^{-1}), PFUnA (7.8 ± 2.5 – 17.7 ± 5.7 ng L^{-1}), and PFHxA (5.1 ± 1.8 – 14.0 ± 4.9 ng L^{-1}).⁷

Schroeder *et al.* investigated the presence and distribution of PFAS in industrial airborne emissions emission to land in Vermont and Eastern New York State, USA. The study confirmed uninterrupted PFAS contamination in soils from inhabited areas into conserved forest lands. These reports confirmed airborne PFAS deposition to soils near industrial sites and transfer further away from contamination sites.³⁴

Assessing PFAS concentrations across various land use types, such as forests, agricultural land, remote areas, and urban soils, as well as comparing to the levels found in contaminated sites—such as industrial soils, AFFF-contaminated soils, and biosolid-amended soils—can offer valuable insights into the background levels of PFAS in soils and the mechanisms of atmospheric deposition.^{13,37,48} In a study conducted by Zhu *et al.*, 66 soil samples were collected from Vermont US, targeting publicly owned lands (*e.g.*, municipal building lawns, municipal parks and communal gardens, school lawns, and urban forests), which were not close to or located near any suspected PFAS sources such as industries, firefighting training facilities or air base.²¹ The study confirmed a concentration of $\sum 17$ PFAS ranging from 0.54 to 36 μg per kg d.w. across these sites.²¹ In a recent state-wide survey, 83 forest soil samples in Michigan were analyzed for 28 PFAS to determine the PFAS background levels.⁴⁹ PFAS levels in these samples ranged from 0.1–37 μg kg^{-1} , similar to the concentration range detected in Vermont²¹ (Table 1).

Lyon, France, is recognized as the birthplace of the French chemical industry and remains a center for extensive chemical production.⁵⁰ The industrial site in Pierre-Bénite has played an essential role in the production of fluoropolymers since the late 1950s, operated mainly by Arkema and Daikin.⁵¹ Historically, fluoropolymer production at Pierre Benite's site was initiated by SRF (société des résines fluorés) starting from 1957, which became Atochem in 1983, then Elf Atochem in 1992, Atofina in 1999 and then Arkema in 2004. Daikin implantation in Pierre Bénite started in 2003.⁵² Over the last decades, these industries have manufactured different fluoropolymer products, using a range of PFAS as processing supports and raw materials.⁵³

In the early years (1958–1986), Arkema produced polychlorotrifluoroethylene (PCTFE) using perfluoroalkyl telomers (Forafac) and began producing polytetrafluoroethylene (PTFE) in 1965, utilizing perfluorooctanoic acid (PFOA) as an emulsifier. The use of PFOA was later phased out due to its persistence in the environment and growing regulatory measures against its use.⁵³ From 1984–1996, the industry produced fluorinated acrylic copolymers using perfluoroalkyl telomers (Forafac®) and perfluoroalkyl alkenes (Foralkyl®).⁵³ From 2003 to 2017, the industry transitioned to using Surflon®, a commercial PFAS additive containing 74% PFNA, 16–20% PFUnDA, 5% PFTrDA, and <1% PFOA for the production of polyvinylidene fluoride (PVDF). After 2017, Surflon® was replaced by a 6:2



Table 1 A literature review of reported PFAS background levels in soils compared with the current study

Location	Sample size	Year	PFAS compounds	Concentration ($\mu\text{g kg}^{-1}$)	Reference
North America	33	NS	PFAAs	0.18–8.1	71
Europe	10	NS	PFAAs	0.05–9.61	71
Asia	6	NS	PFAAs	0.21–14.7	71
Africa	5	NS	PFAAs	0.12–1.63	71
South America	3	NS	PFAAs	0.05–0.37	71
Antarctica	1	NS	PFAAs	0.198	71
Paris (France)	32	2010	$\sum 22$ PFAS	0.2–3.2	69
US	10	2012	$\sum 13$ PFAAs	7.9–129	67
China	33	2013	PFOA + PFOS	13.9	72
Uganda	35	2015	$\sum 26$ PFAS	1.7–7.9	18
Japan	10	NS	$\sum 13$ PFAAs	9.4–35.5	67
Mexico	10	NS	$\sum 13$ PFAAs	10.8	67
Sweden	31	2017	$\sum 28$ PFAS	0.40–6.6	68
Korea	243	2017	PFOA, PFOS	0.5–3.5	73
China	153	2018	PFAAs	3–64	74
Vermont-US	66	2018	$\sum 17$ PFAS	5.40–36	21
Belgium	10	2019	$\sum 15$ PFAS	0.8–53	75
Michigan-US	83	2019–2021	$\sum 28$ PFAS	0.1–37	49
Lyon	215	2024	$\sum 80$ PFAS	0.3–167	Current study

fluorotelomer sulfonic acid (6:2 FTS)-based product (Capstone®), as part of efforts to phase out the use of long-chain PFAS.⁵³ Additionally, the industry also declared the production of various perfluoroalkyl iodides (RFI), ranging from 6:2 RFI ($\text{C}_6\text{F}_{13}\text{C}_2\text{H}_4\text{I}$), $\text{C}_6\text{F}_{13}\text{I}$, 8:2 RFI ($\text{C}_8\text{F}_{17}\text{C}_2\text{H}_4\text{I}$), $\text{C}_8\text{F}_{17}\text{I}$, 10:2 RFI ($\text{C}_{10}\text{F}_{21}\text{C}_2\text{H}_4\text{I}$), $\text{C}_{12}\text{F}_{25}\text{I}$ and 12:2 RFI ($\text{C}_{12}\text{F}_{25}\text{C}_2\text{H}_4\text{I}$).⁵³

According to France's General Inspectorate for the Environment and Sustainable Development (IGEDD) report in 2022, Arkema's facility in Pierre-Bénite releases up to 3.5 tons of PFAS every year into the Rhône River.⁵³ As of 2024, the quantity of PFAS released into ducted air by Daikin is estimated to be around 40 g per day, with PFHxA accounting for 95% of the total PFAS, while Arkema releases 6:2 FTS and PFHxA (60 g per day) into the air with traces of PFNA and PFUnDA.⁵³

Due to this historical and continuous use of PFAS in this region, Mourier *et al.*, in their study, confirmed $\sum$ PFAS concentrations reaching 48.7 ng g^{-1} d.w. in sediments collected from a backwater area adjacent to the Rhone River downstream near these fluoropolymer manufacturing industries in Lyon.⁵⁴ Also, the result of the PFAS campaign held by the French TV program "Vert de Rage" in Lyon⁵⁵ confirmed that PFAS concentrations in the Rhône River downstream of industrial plants was over 36 000 times higher than those found 2.5 km upstream of those plants. The Team also reported over 200 ng l^{-1} PFAS concentration in drinking water serving around 60 000 people in South Lyon, compared to the EU's proposed threshold of 100 ng l^{-1} .^{55,56} In a recent study, Teymoorian *et al.* investigated PFAS levels in various water sources in southern Lyon, including ponds, rivers, wells, springs, and tap water. The study confirmed average values of 147, 123, and 71 ng L^{-1} in groundwater, tap water, and surface water samples for the $\sum_{77}$ PFAS.⁸

In another study by Babut *et al.*, $\sum$ PFAS concentrations in aquatic plants harvested from the Rhone River ranged from 4.8 to $7.6 \text{ ng per g d.w.}$ with PFNA, PFUnDA and PFTrDA being the

dominant PFAS detected ($2.0\text{--}2.9 \text{ ng g}^{-1}$). The study also confirmed a 100% PFAS (C9–C13 PFCAs), detection rate in fish from this river, with concentrations varying from 6.6 to $356 \text{ ng per g d.w.}$ ⁵⁷

To understand the extent of airborne PFAS deposition in Lyon soils, Dauchy investigated the concentration of PFAS in 9 surface soils collected 200 m away from a specific PVDF and fluoroelastomer production industry and five outdoor dusts in Pierre-Bénite, Lyon. The study recorded a high detection rate for long-chain perfluoroalkyl carboxylic acids (PFCAs) ($C \geq 9$), with PFUnDA being the predominant PFAS in surface soils ($12\text{--}245 \text{ ng per g d.w.}$) and PFTrDA in outdoor dusts ($<0.5\text{--}59 \text{ ng per g d.w.}$). The study concluded that this contamination likely originated from PVDF and fluoropolymer production industries located near these sample collection sites.⁵⁸

The current study provides a detailed understanding of PFAS contamination in Lyon soils. By collecting samples from various distances, close to and far from industrial facilities, this study mapped the patterns of airborne PFAS deposition and assessed how proximity to fluoropolymer plants affects PFAS levels in the soil. Furthermore, we analyzed a broad spectrum of PFAS compounds, including cationic, anionic, and zwitterionic species, to better understand their use, environmental persistence, and bioaccumulation in the soil. Additionally, this study evaluated PFAS in soils from chicken-feeding areas and free-range chicken eggs. PFAS contamination in eggs has been documented across France, especially in areas with extensive industrial activities.^{59–61} In 2023, the Rhône prefecture (French Government) warned against the consumption of eggs from home-raised chickens due to reports of high PFAS levels in free-range chicken eggs near chemical facilities in south Lyon.⁶¹ Similar concerns also began in Île-de-France when egg samples from 410 municipalities around Paris were analyzed and confirmed to be contaminated with PFAS. Additionally, in the northern French town of Villers-Saint-Paul, tests conducted in



2024 revealed high PFAS levels in eggs, prompting an investigation into a nearby U.S.-owned chemical facility as a potential source of PFAS contamination. Consequently, the French health agency cautioned citizens regarding PFAS-contaminated eggs from domestic henhouses.⁵⁹

By evaluating the range of PFAS in paired chicken feeding areas and the eggs of free-range chickens feeding on those soils, we assessed the industrial footprint on PFAS types and concentrations in the eggs to determine whether soils are a significant source of PFAS exposure to free-range chickens in home gardens. This approach will yield insights into the transport and accumulation mechanisms of PFAS across various environmental matrices in highly industrialized areas.

2 Materials and methods

2.1 Study area and soil sample collection campaign

The soil sampling site – Lyon, is in Southeastern France in the Auvergne-Rhône-Alpes region (Fig. 1). As part of the citizen science collaboration between the Université de Montréal and “Ozon L'Eau Saine,” a group of citizens helped collect the soil samples through a collection campaign across the Greater Lyon metropolitan area. Sampling locations were distributed across the central, north, south, east, and west of the Arkema industrial site located in Pierre-Bénite. To better understand industrial inputs to PFAS levels across the Lyon area, soil samples were collected very close (0.3–2 km), close (2–10 km), and far (10–30 km) from the Arkema industrial site. Additionally, soil samples from agricultural plots receiving sewage sludges and

irrigated/non-irrigated agricultural plots in greenhouses and open fields were collected to verify the impact of sewage sludge application and irrigation on the contamination. During this sampling campaign, 215 topsoil samples were collected at 5–6 cm soil depth using properly washed stainless steel spoons. In addition, soil samples were collected from private poultry house locations for which earlier data on PFAS egg concentration were analyzed and available. Those egg-soil pairs were used to verify the correlation between soil and egg contamination and ranged from 2.5 km from Arkema, 11–15 km south of Arkema and 5 km west of the Arkema site.

A video was created to demonstrate the correct analytical procedures, guiding citizens through appropriate soil sampling steps. Field blanks using clean Ottawa sand were also exposed to the same conditions as the samples to monitor possible cross-contamination during sampling, transport and preparation.

2.2 Chemicals

Certified standard solutions of native and internal standards were purchased from Wellington Laboratories (Guelph, ON, Canada) or the Fluobon Surfactant Institute (Beijing, China). The SI (Table S1) fully describes the native standards and their corresponding internal standard compositions with acronyms.

Acetonitrile, methanol, ammonium fluoride, ammonium acetate, Ottawa blank soil and HPLC-grade water containing 0.1% formic acid were purchased from Fisher Scientific (Whitby, ON, Canada). Additional reagents, including

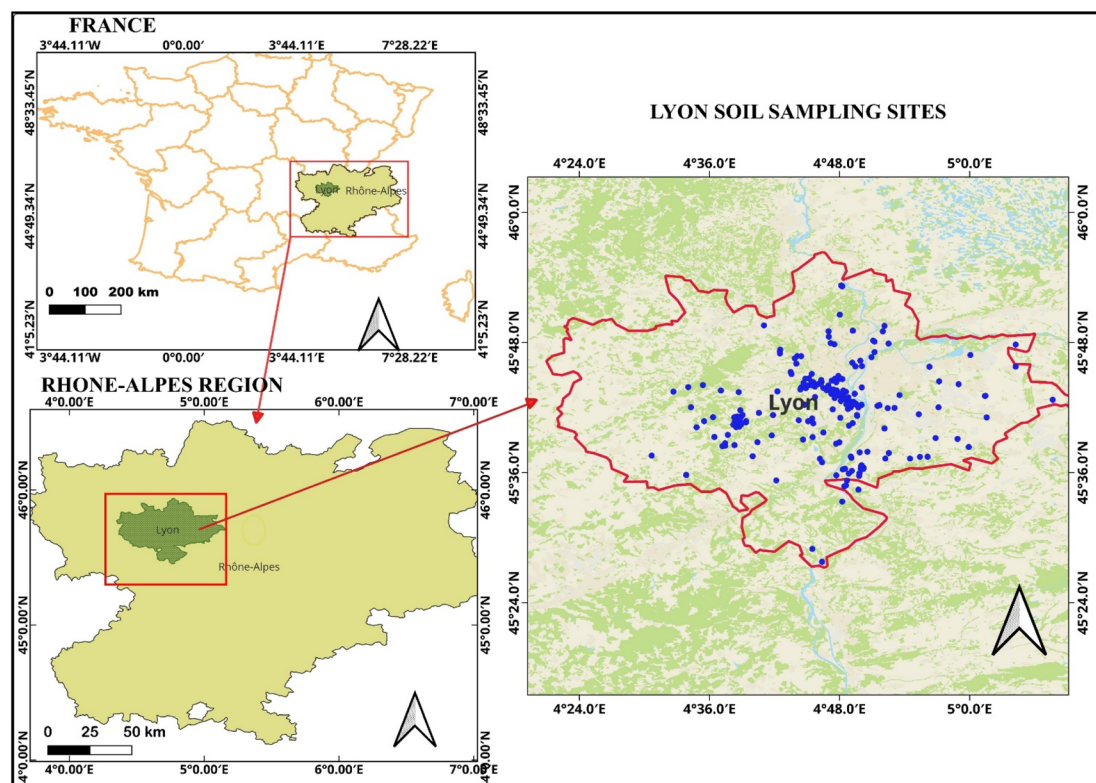


Fig. 1 Map of the study area.



ammonium acetate ($\geq 98\%$ purity) and formic acid (reagent grade, $\geq 95\%$ purity), were obtained from Sigma-Aldrich (St. Louis, MO, USA). Supelclean ENVI-Carb cartridges (500 mg/6 mL) were acquired from Supelco (Bellefonte, PA, USA).

2.3 PFAS extraction from soil samples

Samples were extracted following the validated protocol established by Munoz *et al.*⁶² Upon arrival at the laboratory, soil samples were lyophilized, pulverized and adequately homogenized. For extraction, 1 g of soil was precisely weighed into methanol-pre-washed 15 mL polypropylene tubes. Each sample was spiked with an internal standard solution (10 ppb) and left to equilibrate for 60 min. Extraction was performed in two cycles using methanol containing 100 mM ammonium acetate as the extracting solvent. Each cycle involved an addition of 5.0 mL of the extraction solvent, followed by high-speed vortexing (0.5 min at 3200 rpm), ultrasonication (20 min), and centrifugation (10 min at 6000 rpm). Supernatants from each cycle were transferred to methanol-pre-rinsed tubes, and the combined extracts were concentrated to approximately 4 mL under nitrogen with mild heating (40–45 °C).

The extracts were then carried through a clean-up process using pre-conditioned Supelclean ENVI-Carb cartridges (500 mg, 6 mL), followed by a rinse with 3.0 mL of methanol, reconcentrated to about 0.8 mL under nitrogen and mild heating. A 150 μ L aliquot of the final extract, combined with 30 μ L of HPLC-grade water, was transferred to a polypropylene LC-MS vial, vortexed briefly, and analyzed using ultra-high-performance liquid chromatography coupled with high-resolution mass spectrometry (UHPLC-HRMS).

Analyte separation was achieved using a reverse-phase C18 column (Thermo Hypersil Gold, 100 mm $\times$ 2.1 mm, 1.9 μ m particle size) with a Thermo Hypercarb PFAS delay column (20 mm $\times$ 2.1 mm, 7 μ m) to trap background PFAS contamination. A dual-injection approach was used to analyze 80 targeted PFAS compounds in soil and plant extracts. The first injection, optimized for diPAPs, consisted of mobile phases: (A) H₂O + 0.1 mM NH₄F and (B) MeOH + 0.1 mM NH₄F.¹² The second injection analyzed the remaining 77 PFAS using mobile phases consisting of (A): 0.1% formic acid in HPLC-water and (B): 0.1% formic acid in acetonitrile.

A heated electrospray ionization source with an instant polarity-switching capacity was used. Analyte detection was performed using a Q-Exactive Orbitrap mass spectrometer with a resolution of 70 000 full width at half maximum (FWHM) at m/z 200, controlled by Xcalibur 2.3 software (Thermo Fisher Scientific, Waltham, MA, USA). The mass scan range was set at m/z 150–1000 in full scan MS mode. The free-range chicken eggs were analyzed for 14 PFAAs in a commercial laboratory, and corresponding QA/QC data are presented in Table S3.

2.4 Quality assurance and quality control

To prevent cross-contamination during sampling, soil and plant samples were stored separately in polyethylene ziplock bags and field blanks (blank soil using Ottawa sand) were exposed to sample collection sites and conditions to monitor

contamination during sample movement. Method and instrument blanks were prepared using Milli-Q water and methanol to monitor contamination during extraction and instrumental analyses. Some specific anionic PFAS were identified at low concentrations in the method blanks, and the average values were subtracted from the measured PFAS concentrations across all samples. However, no cationic PFAS were detected above the detection limit in the blank samples. For target analyte quantification, 8-point matrix-matched standard curves for soil samples (0, 0.5, 0.1, 1, 2, 5, 10, and 20 ng g⁻¹) were prepared by spiking samples with native standards and 10 ng g⁻¹ internal standards. The calibration curves' regression coefficients (R^2) were >0.99 for all target analytes. Detailed QA/QC information can be found in Table S2.

The limits of detection (LODs) were derived from the lowest concentration of the respective calibration curves for all 80 PFAS. The compound-specific MLODs obtained for soil samples ranged from 0.005 to 0.5 ng g⁻¹ (Table S2). The limit of quantitation (LOQ) was set at three times the LOD.

Recovery tests for native standards in both matrices were conducted to assess the accuracy and efficiency of the extraction process. The recoveries of 80 target analyte standards were evaluated by spiking composite samples (composed of mixed grab samples from all analyzed soils) with 25 ng g⁻¹ of native standards and 10 ng g⁻¹ of internal standards. The QC accuracy ranged from 57–118%, while matrix recovery of target analyte standards in the composite soil samples ranged from 75% to 125% (Table S2).

2.5 Estimation of human exposure to soil PFAS via the CSOIL model

To evaluate the potential exposure of Lyon residents to PFAS, the CSOIL model was used to calculate the total estimated daily intake of PFAS from the soil.⁶³ The CSOIL model estimates the daily intake of contaminants by considering overall exposure through soil ingestion, dermal contact, and inhalation.⁶³ Soil can be accidentally ingested *via* unwashed hands or vegetables in adults and could be ingested intentionally by children. Once ingested, the contaminant in the soil is released into the digestive tract and then absorbed into the body. Inhalation of soil is a significant exposure route, mainly through inhaling indoor or outdoor dust. Factors such as wind velocity, soil particle size, texture, and humidity can influence soil inhalation. Dermal exposure to contaminants from soil is expected to be minimal because human skin has protective layers.

The equation of each exposure path is presented in eqn (1)–(3)

$$\text{EDI ingestion} = \frac{\text{Csoil} \times \text{SDI} \times \text{Fa}}{\text{BW}} \quad (1)$$

$$\text{EDI inhalation} = \frac{\text{Csoil} \times \text{IASP} \times \text{Fr} \times \text{Fa}}{\text{BW}} \quad (2)$$

$$\text{EDI dermal} = \frac{\text{Csoil} \times \text{AEXP} \times \text{Fm} \times \text{DAE} \times \text{DAR} \times \text{TB} \times \text{FR} \times \text{Fa}}{\text{BW}} \quad (3)$$



$$\Sigma \text{EDI} = \text{EDI ingestion} + \text{EDI inhalation} + \text{EDI dermal} \quad (4)$$

$$\text{Risks Quotient} = \frac{\sum \text{EDI} (\text{ng kg}^{-1} \times \text{bw per day})}{\text{TDI} (\text{ng kg}^{-1} \times \text{bw per day})} \quad (5)$$

where C_{soil} is the soil PFAS concentration (ng mg^{-1}), SDI : daily soil ingestion (adult = 100 mg per day, children 200 mg per day),^{14,64} F_a : the relative adsorption factor (adult = 0.6 mg per day, children = 0.5),¹⁴ IASP : the inhaled amount of soil particles (children: 3.13×10^{-7} kg per day, adult: 8.33×10^{-7} kg per day), F_r : the soil particle retention factor in the lung (0.75), AEXP : the exposed surface area of skin (children: 0.05 m^2 , adult: 0.09 m^2),⁶⁵ F_m : the matrix factor dermal uptake (0.115),¹³ DAE : the degree of skin coverage (children: 0.0051 kg m^{-2} , adults: 0.038 kg m^{-2}),¹³ DAR : dermal absorption velocity (children: 0.01 h^{-1} , adults: 0.005 h^{-1}),¹⁴ TB is the average period exposed to soil

(children 2.86 h per day, adults 1.14 h per day).⁶⁵ BW is the average body weight (for children: 25 kg, for adults: 75 kg),⁶⁴ and TDI is the established tolerable daily intake value. The TDI used to calculate the risk quotient (RQ) in this study is the value established by EFSA for $\Sigma 4$ PFAS (0.63 ng per kg bw per day), PFOA (0.9 ng per kg bw per day), and PFOS (1.9 ng per kg bw per day).

This study assessed PFAS concentrations in soil on a dry basis, while egg samples were analyzed on a wet-weight basis.

2.6 Statistical analysis

Statistical analysis was conducted using IBM SPSS Statistics (Version 27) and JMP® (Version 18.1, SAS Institute Inc., Cary, NC, 1989–2023). The Shapiro–Wilk was utilized to evaluate the data distribution. The Wilcoxon non-parametric test was used

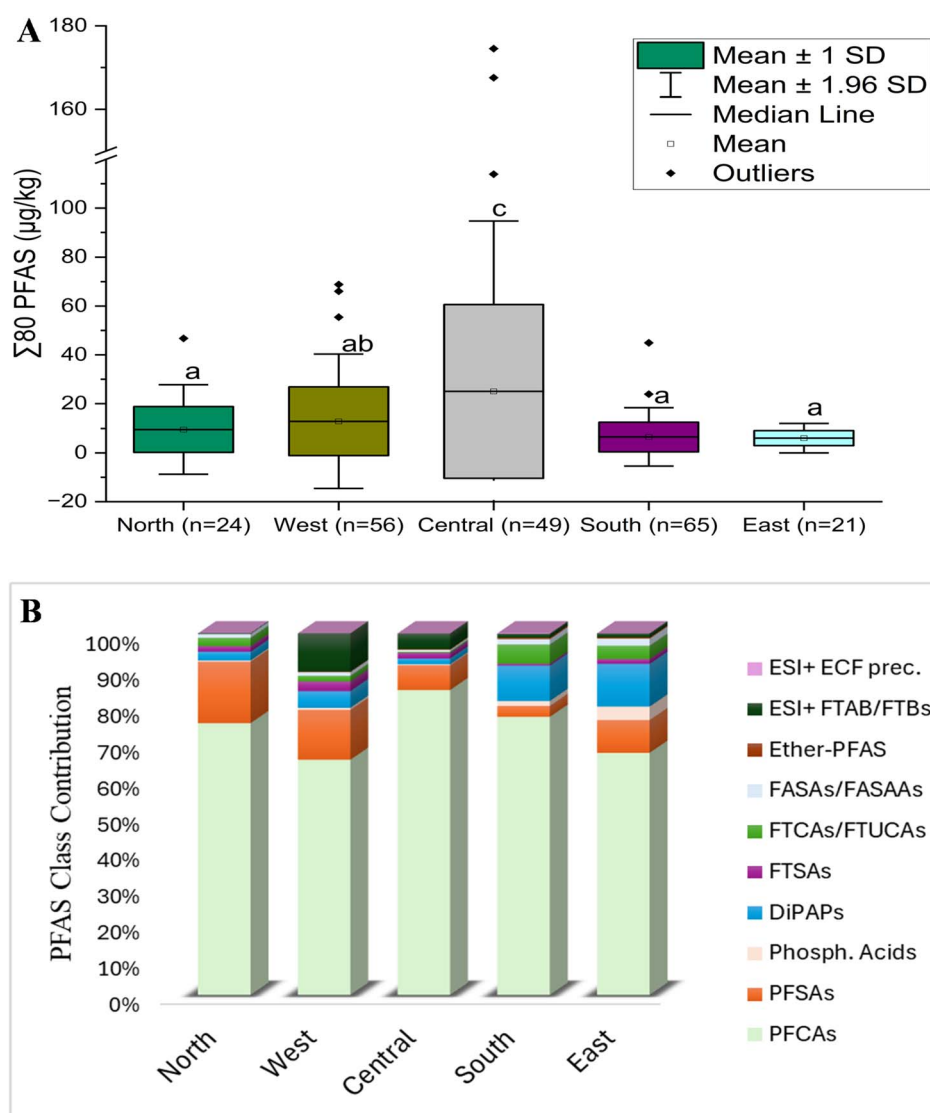


Fig. 2 (A) The sum of 80 PFAS concentrations across Lyon. Wilcoxon Non-parametric test: different letters indicate statistically significant differences ($p < 0.05$). Pairwise p -values: Lyon west vs. Lyon south ($p < 0.0001$), Lyon west vs. Lyon east ($p = 0.0097$), Lyon west vs. Lyon central ($p = 0.0012$), Lyon south vs. Lyon east ($p = 0.651$), Lyon south vs. Lyon central ($p < 0.0001$), Lyon north vs. Lyon central ($p = 0.0006$). (B) Percentage contribution of different PFAS superclasses.



to identify significant differences among the variables in non-normally distributed data.

3 Results and discussion

3.1 PFAS occurrence and concentration levels across Lyon

Across the Greater Lyon area, this study recorded concentrations ranging from 0.6–175 $\mu\text{g kg}^{-1}$ for the $\Sigma 80$ PFAS (Fig. 2A, and Table S3). PFAS concentrations varied across Lyon (central, north, south, west and east of the Arkema industrial site), with the highest levels recorded in central locations (3.8–175 $\mu\text{g kg}^{-1}$), where the Arkema PFAS chemical facility is situated (Fig. 2A).

Next to the central parts were samples collected west of the industrial site with concentrations ranging from 1.6–69 $\mu\text{g kg}^{-1}$ (average value = $13 \pm 14 \mu\text{g kg}^{-1}$, $n = 56$), while the north zone ranged from 1.9–47 $\mu\text{g kg}^{-1}$ ($n = 24$). In contrast, southern and eastern locations exhibited lower PFAS concentrations, with values ranging from 1.2–45 $\mu\text{g kg}^{-1}$ (average value = $6.5 \pm 6.1 \mu\text{g kg}^{-1}$, $n = 65$) and 0.6–11.3 $\mu\text{g kg}^{-1}$ average = $6.0 \pm 3.1 \mu\text{g kg}^{-1}$ ($n = 21$) respectively, indicating minimal deposition in those areas. The south and eastern soils showed some site-specific contamination but were not as high as the central and western soils. Statistical analysis revealed a significant difference between PFAS concentrations in the central zones and others ($p < 0.001$). Statistically, east, north, and south soils showed similar PFAS levels, while west soils displayed intermediate contamination (Fig. 2A). This trend strongly suggests that industrial inputs have contaminated soils within the central, western and northern zones.

Fig. 3 revealed the spatial distribution of PFAS across Lyon, with the prevailing winds showing direction, speed, and frequency across the city's cardinal zones. The prevailing wind in Lyon is the Mistral, a strong north-to-south ($N \rightarrow S$) wind that blows through the Rhône Valley primarily from the north (N, NW, NE), with occasional south (S, SW) winds in specific periods.⁶⁶

The spatial distribution of $\Sigma 80$ PFAS in Lyon soils suggests that airborne transport plays a significant role in the contamination patterns, especially within the west, north, and southern parts of the Arkema site. The higher PFAS concentrations found in the west and northern zones suggest these areas are mainly downwind from the Arkema industrial site, resulting in increased airborne PFAS deposition. In contrast, the lower PFAS concentrations in the east may be due to its upwind position relative to the dominant wind patterns, which decreases the potential for significant atmospheric deposition. The moderate PFAS levels detected in the south suggest some degree of deposition, likely influenced by variations in wind direction and occasional downwind transport from the Arkema industrial site during the Mistral season. Increased PFAS concentrations of up to 45 $\mu\text{g kg}^{-1}$ are reported in some soils from the southern zone and may be attributed to other factors, including the potential application of biosolids in certain areas.

PFAS concentrations observed in Lyon soils ($\Sigma 80$ PFAS = 0.6–175 $\mu\text{g kg}^{-1}$) are significantly higher than those reported in most background PFAS levels reported in different studies across different regions (Table 1). The closest comparison is the background level across the U.S. ($\Sigma 13$ PFAAs = 7.9–129 μg

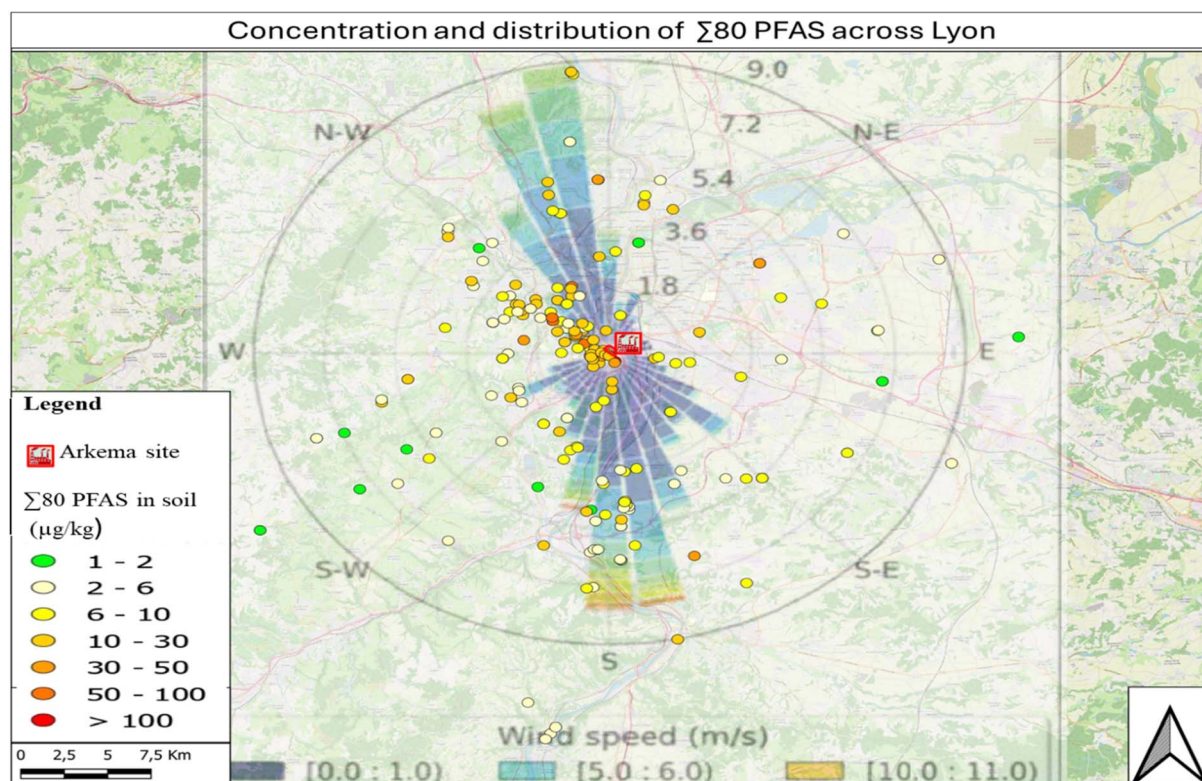


Fig. 3 Map of PFAS concentration and distributions with wind direction across Lyon: prevailing wind indicates wind speed and direction.



kg^{-1}),⁶⁷ which also suggests substantial contamination, likely from industrial sources.⁶⁷ Other regions, such as Sweden ($\sum 28$ PFAS = $0.40\text{--}6.6 \mu\text{g kg}^{-1}$),⁶⁸ Paris ($\sum 22$ PFAS = $0.2\text{--}3.2 \mu\text{g kg}^{-1}$),⁶⁹ Japan ($\sum 13$ PFAAs = $9.4\text{--}35.5 \mu\text{g kg}^{-1}$), Vermont-USA ($\sum 17$ PFAS = $5.40\text{--}36 \mu\text{g kg}^{-1}$),²¹ and Michigan-USA ($\sum 28$ PFAS = $0.1\text{--}37 \mu\text{g kg}^{-1}$),⁴⁹ reported considerably lower levels, emphasizing the extent of PFAS accumulation in Lyon due to industrial activities. Michaud *et al.* investigated the concentration of PFAS in soils from long-term experimental sites located in mainland France, specifically in Nancy (Grand-Est), Feucherolles (Île-de-France), Colmar (Grand-Est), and the French Overseas Territories in the southwestern Indian Ocean, covering the period from 1974 to 2017. In their study, the $\sum 75$ PFAS in soils without organic waste products (OWP) were less than $3 \mu\text{g kg}^{-1}$, while concentrations in soils receiving OWP increased up to $20 \mu\text{g kg}^{-1}$.⁷⁰ In contrast, PFAS concentrations in the Lyon soils in our study increased up to $175 \mu\text{g kg}^{-1}$, further confirming the influence of industrial activities on PFAS levels in Lyon's environment.

3.2 Industrial footprint on PFAS profiles in Lyon soil

The 80 PFAS analyzed in this study were classified into 10 PFAS superclasses (Fig. 2B, and Table S4). Among these superclasses, perfluorocarboxylic acids (PFCAs) were the dominant class across all the zones, contributing 84% (central of Arkema site), 76% (south), 75% (north), 67% (east), and 65% (west) of the total PFAS detected (Fig. 2A). The prevalence of PFCAs, particularly long-chain PFAS (C8 to C14), suggests a strong industrial influence, as these compounds are widely used as surfactants in fluoropolymer production.^{17,48} The detection frequencies of PFAS revealed a distinct industrial footprint, particularly from Surflon-related compounds, which are known to originate from fluorochemical manufacturing.⁵⁸

Across the zones, 18 PFAS were frequently detected in the samples, with detection rates ranging from 90–100% (PFHxA, PFOA, PFNA, PFOS, and PFUnA), 60–80% (PFPeA, PFHxS, 6-2 dipap, PFTrDA, and PFDA) and PFBS, PFOcDA, 6-2 FTS, 6-2 FTAB, PFDoA, PFTeDA were present in 10–50% of the analyzed soils (Fig. S1).

In the central zone, where major chemical industries are situated, Surflon-related compounds PFUnA ($0.9\text{--}106 \mu\text{g kg}^{-1}$), PFNA ($0.3\text{--}13 \mu\text{g kg}^{-1}$), and PFTrDA ($0.7\text{--}47 \mu\text{g kg}^{-1}$) were the dominant PFAS (Fig. 4), accounting for 35, 16, and 10% of the total mass of PFAS detected, respectively (Fig. 5A). The detection of Surflon-related PFAS was also reported in an earlier study by Dauchy⁵⁸ where nine soil samples from this specific central zone were analyzed. The presence of long-chain PFCAs at high concentrations is unusual, and their occurrence within the central zone strongly suggests that they were caused by direct emissions from industrial activities located within this area.

In the northern side of Arkema site, the dominant PFAS included PFNA ($0.02\text{--}11 \mu\text{g kg}^{-1}$), PFHxS ($0.1\text{--}24 \mu\text{g kg}^{-1}$), PFUnA ($0.1\text{--}6.7 \mu\text{g kg}^{-1}$), PFOS ($0.3\text{--}3.3 \mu\text{g kg}^{-1}$), and PFOA ($0.1\text{--}1.8 \mu\text{g kg}^{-1}$), accounting for 25%, 19%, 13%, 12%, and 8% of the total PFAS, respectively (Fig. 4, and 5A). A similar trend was observed in the west, with PFNA ($0.1\text{--}17 \mu\text{g kg}^{-1}$), PFUnA (0.2--

$21 \mu\text{g kg}^{-1}$), PFOS ($0.2\text{--}11 \mu\text{g kg}^{-1}$), and PFOA ($0.1\text{--}4.9 \mu\text{g kg}^{-1}$) contributing 16%, 13%, 12%, and 5% of the total PFAS (Fig. 4, and 5A). PFAS compounds associated with Surflon remain prevalent in the area near the Arkema industrial site, indicating the potential impact of atmospheric deposition or other contamination routes.

Lower concentrations of Surflon-related PFAS were recorded in the southern and eastern zones, where industrial influence is less pronounced and also less impacted by prevailing winds. Within the east, PFOS ($0.1\text{--}2.9 \mu\text{g kg}^{-1}$) and PFOA ($0.1\text{--}1.02 \mu\text{g kg}^{-1}$) were recorded, while in the south, PFOS ranged from $0.2\text{--}32.6 \mu\text{g kg}^{-1}$ (average = $1.4 \mu\text{g kg}^{-1}$), and PFOA varied from $0.2\text{--}6.8 \mu\text{g kg}^{-1}$ (average = $0.6 \mu\text{g kg}^{-1}$). These results suggest that the influence of industrial emissions decreases with distance from the central industrial hub (Fig. 4).

A notable finding was the detection of zwitterionic PFAS; 6 : 2 fluorotelomer sulfonamide alkylbetaine (6 : 2 FTAB, also referred to as 6 : 2 FTSA-PrB) in 15% of samples, with concentrations ranging from $0.1\text{--}52 \mu\text{g kg}^{-1}$ (Fig. 4). While most detections were below $2 \mu\text{g kg}^{-1}$, a sample from the central zone (1.6 km from Arkema) contained $52 \mu\text{g kg}^{-1}$ of 6 : 2 FTAB, and two samples from the west (~ 2 km from Port Édouard-Herriot) had concentrations around $37\text{--}38 \mu\text{g kg}^{-1}$ (6 : 2 FTAB). 6 : 2 FTAB is a major component of some aqueous film-forming foams (AFFFs) used for firefighting. Its presence in the soil near the Port Édouard-Herriot could be linked to past AFFF applications, specifically during the 1987 fire at the oil depot within the port.⁷⁶ The persistent nature of 6 : 2 FTAB and the possibility of leaching into surrounding areas over time seems a plausible explanation for its detection in the analyzed soils.^{77,78}

6 : 2 FTS, being a possible degradation product of 6 : 2 FTAB, was detected in 39% of soil samples collected in central Lyon, with concentrations ranging from 0.1 to $13 \mu\text{g kg}^{-1}$. Also, high detection rates (92–100%) were recorded for PFHxA across the zones, with concentrations ranging from 0.1 to $4.1 \mu\text{g kg}^{-1}$ (central), $0.05\text{--}2.7 \mu\text{g kg}^{-1}$ (south), $0.05\text{--}2.4 \mu\text{g kg}^{-1}$ (west), $0.1\text{--}2.1 \mu\text{g kg}^{-1}$, and $0.03\text{--}0.95 \mu\text{g kg}^{-1}$ (east). The detection of 6 : 2 FTS and PFHxA in this study is consistent with their presence in daily emissions from the Arkema and Daikin chemical industries, as reported in a 2023 industry-mandated report submitted by Arkema and Daikin industries to DREAL.⁵³ This report confirms the use of 6 : 2 FTS at the site since at least 1973 and highlights its presence in industrial discharges, with some PFHxA releases linked to Daikin's discharges.⁵³

The PCA analysis of the dominant PFAS across the zone presented in Fig. 5B further provides insights into the variations in PFAS profiles across the different zones. The central zone was primarily distinguished from other zones by PC1 (54.11%), representing the primary PFAS distribution variance. The West differed from the East and South by PC2 (28.22%), which indicates a secondary variation.

3.3 Influence of proximity to chemical industries on soil PFAS concentration across Lyon

Fig. 6 illustrates the linear relationship between PFAS concentrations and distance from the Arkema industry. A negative



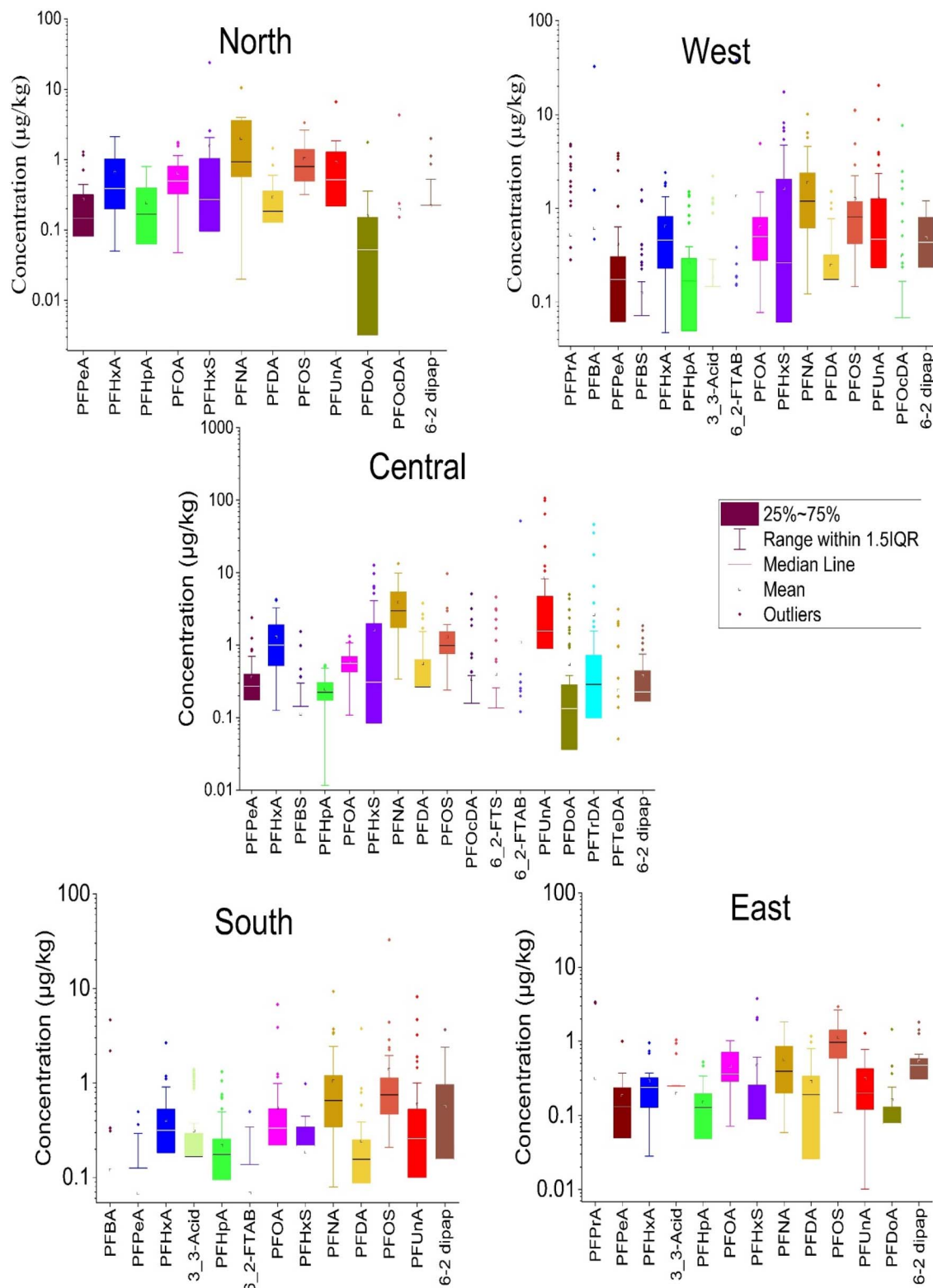


Fig. 4 Concentrations of dominant PFAS across the 5 Lyon zones.

Pearson correlation was observed between distance from the industrial site and the $\sum 80$ PFAS ($r = -0.3266$, $p < 0.0001$), $\sum$ Surflon PFAS ($r = -0.2862$, $p < 0.0001$) and the sum of the four PFAS regulated by the European Food Safety Authority

(EFSA), which include PFOA, PFOS, PFNA, and PFHxS ($r = -0.27$, $p < 0.0001$). A decrease in the concentration of other dominant PFAS was also noted as the distance from the chemical industry increased (Fig. S2).



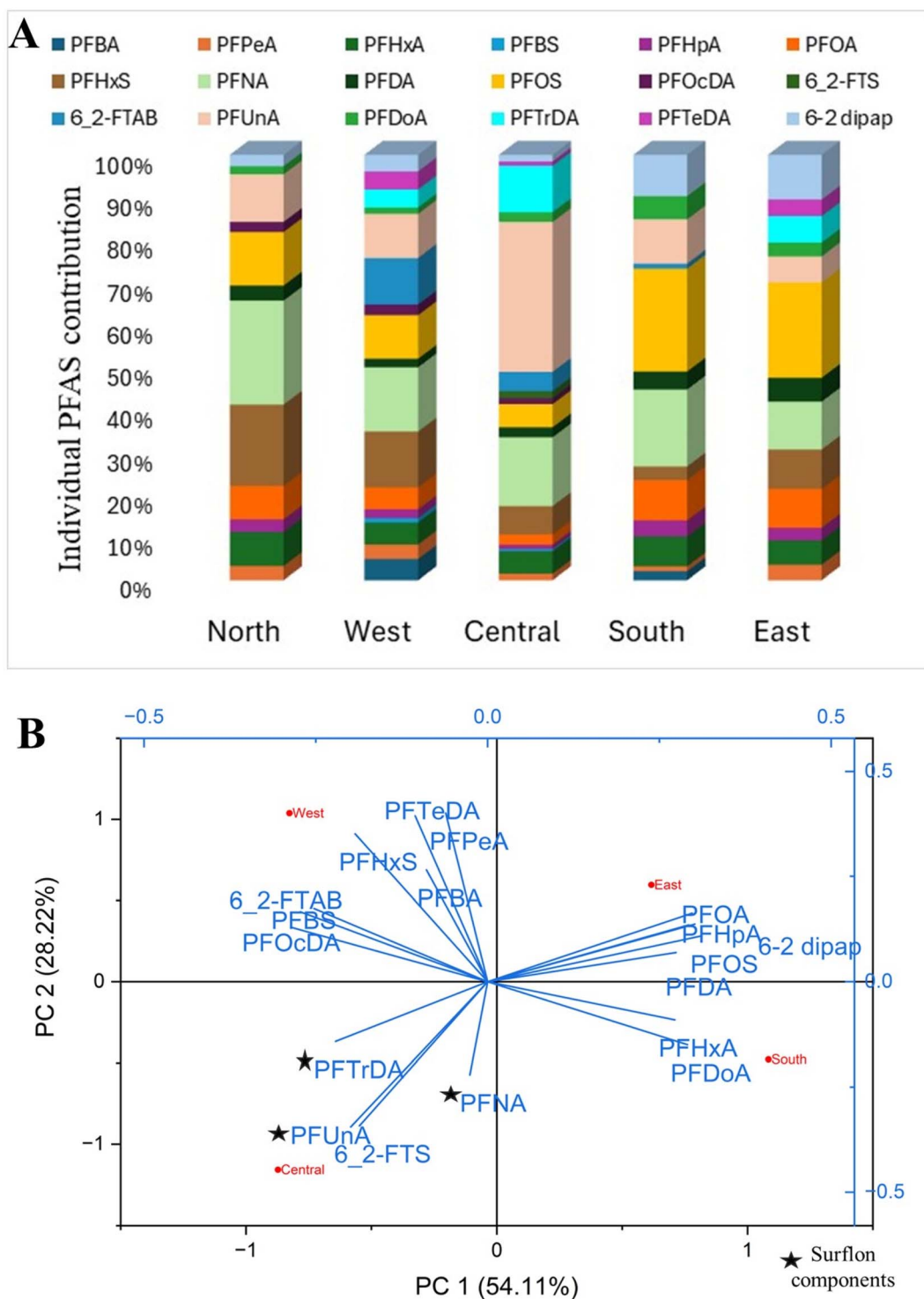


Fig. 5 (A) Contribution of dominant PFAS to the total PFAS concentration across the zones, (B) PCA analysis of the dominant PFAS across the zones.

For soil samples collected within 10 km of the chemical industry, PFAS concentrations increase up to $175 \mu\text{g kg}^{-1}$ ($\sum 80$ PFAS), $148 \mu\text{g kg}^{-1}$ (Surflon PFAS) and $30 \mu\text{g kg}^{-1}$ (sum of 4 PFAS regulated by EFSA), however, a more significant percentage of samples within this distance fall within $10\text{--}50 \mu\text{g kg}^{-1}$ PFAS concentration. The $\sum$ PFOA and PFOS within this distance

ranged from $0.2\text{--}12 \mu\text{g kg}^{-1}$. Between 10 and 20 km away from Arkema, PFAS concentrations in soils showed a notable decline. The $\sum 80$ PFAS values range from 0.1 to $46 \mu\text{g kg}^{-1}$, while the Surflon PFAS values range between 0.1 and $18 \mu\text{g kg}^{-1}$ (Fig. 6 and S2). However, the $\sum 4$ EFSA PFAS increases from $0.4\text{--}27 \mu\text{g kg}^{-1}$ (within $0\text{--}10$ km) to $0.4\text{--}39 \mu\text{g kg}^{-1}$ ($10\text{--}20$ km). While the



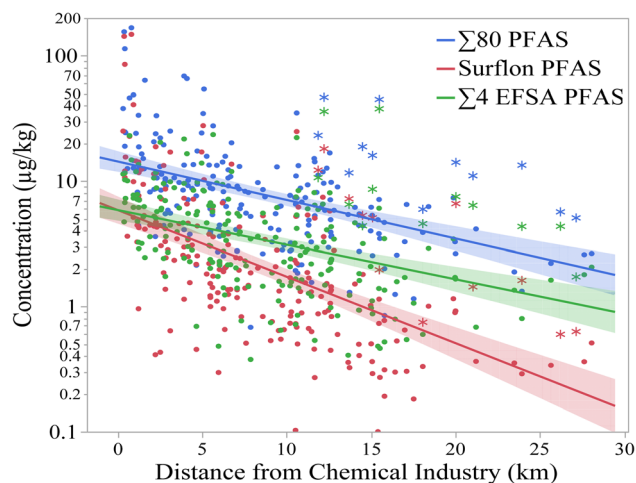


Fig. 6 Correlation between distance from Arkema, $\Sigma 80$ PFAS in Lyon soil and the Σ Surflon PFAS (PFNA, PFUnA and PFTrDA). Pair-wise Pearson correlation: distance & $\Sigma 80$ PFAS ($r = -0.3266$, $p < 0.0001$), distance & Σ Surflon PFAS ($r = -0.2862^*$, $p < 0.0001$), distance & $\Sigma 4$ EFSA PFAS ($r = -0.27$, $p < 0.0001$). Asterisked points (*) in the figure represent locations situated at far distances but downwind of the industrial site.

four PFAS regulated by EFSA include PFOA, PFOS, PFNA, and PFHxS, the increase observed in the $\Sigma 4$ EFSA was due to a rise in PFOS concentration ($32.6 \mu\text{g kg}^{-1}$) recorded in a sample collected 12 km south of the Arkema industry (Fig. S2). The increased PFOS in this soil sample could be due to secondary sources, such as landfills, the application of organic waste products, irrigation of farmland with contaminated water or past contamination. Within this distance, the sum of PFOS and PFOA ranged from 0.2 to $37 \mu\text{g kg}^{-1}$ (Fig. S2).

Beyond 20 km from Arkema, PFAS concentrations decreased to lower levels. Recorded concentrations within this distance range from $1.3\text{--}13.4 \mu\text{g kg}^{-1}$ ($\Sigma 80$ PFAS), $0.2\text{--}1.6 \mu\text{g kg}^{-1}$ (Σ Surflon PFAS), $0.7\text{--}6.4 \mu\text{g kg}^{-1}$ ($\Sigma 4$ EFSA PFAS) and $0.2\text{--}6.4 \mu\text{g kg}^{-1}$ (PFOA + PFOS). Within 15 to 28 km, the role of airborne PFAS deposition was quite visible. The Mistral wind prevalent in Lyon is a strong northwesterly wind that blows through the Rhône Valley. While PFAS concentration in the eastern soils decreased to a background level, within the north, west and southern parts, where the prevailing winds are active, PFAS concentrations still increased up to $45 \mu\text{g kg}^{-1}$ for the sum of 80 PFAS within 15 km (points asterisked within Fig. 6).

The negative correlation between PFAS concentrations and distance from the Arkema industry generally revealed the role of point source emissions and atmospheric transfers in spreading PFAS in the environment. The notable drop in Surflon PFAS levels, compared to the $\Sigma 80$ PFAS, suggests that these specific compounds are closely linked to industrial activities and emissions. In contrast, $\Sigma 4$ EFSA PFAS and the sum of PFOA and PFOS show a wider spread in their presence. Consistent with these findings, high PFAS concentrations have been reported in soils near chemical industries, while levels decrease to background concentrations in areas far from industrial activities.^{21,22,34,46,47,79}

The exponential plot presented in Fig. S3 gives a more precise overview of the influence of proximity to the industrial site and the role of atmospheric deposition on the distribution and concentration of some of the dominant PFAS in Lyon, including PFOA and PFOS. Generally, low coefficients of determination (R^2) values were recorded, indicating a weak exponential decline for each PFAS. However, the R^2 values for surflon PFUnA (0.27), PFTrDA (0.21), and PFNA (0.21) were higher than the very low R^2 values recorded for PFHxS (0.021), PFOS (0.008), and PFOA (0.005). These R^2 values of surflon PFAS revealed point source contamination with a decrease as distance increased. The detection of these PFAS at a distance far from the contamination source revealed the role of atmospheric deposition. Additionally, Arkema and Daikin confirmed the release of PFHxS into the environment in 2024.⁵³ The compound was detected in 100% of the analyzed samples, regardless of the distance, further confirming the role of airborne deposition from a point source to remote areas. For PFOA and PFOS, the near-zero R^2 value indicates that they are present everywhere, regardless of proximity to the point of contamination; however, most of the time, the compounds were detected at background concentrations. This could result from historical contamination, atmospheric deposition, and emissions from diverse sources.

Generally, an overview of PFAS concentrations in Lyon soils, categorized by sample percentiles, is presented in Fig. 7. The percentile classification approach helps identify the number of samples with PFAS background levels, moderate contamination, and possible contamination hotspots. The first 60% of samples ($n = 125$) had concentrations ranging from $0.68\text{--}10.9 \mu\text{g kg}^{-1}$ for the $\Sigma 80$ PFAS, likely representing background contamination levels. These levels are comparable with PFAS background levels reported in the US ($\Sigma 17$ PFAS = $5.4\text{--}36 \mu\text{g kg}^{-1}$),²¹ Sweden ($\Sigma 28$ PFAS $0.5\text{--}6.6 \mu\text{g kg}^{-1}$),⁶⁸ and Mexico ($\Sigma 13$ PFAS $0.4\text{--}10.8 \mu\text{g kg}^{-1}$).⁶⁷ For samples within the 61st to 95th percentile ($n = 80$), concentrations increased up to $63.8 \mu\text{g kg}^{-1}$, suggesting possible industrial inputs. The increase in Surflon

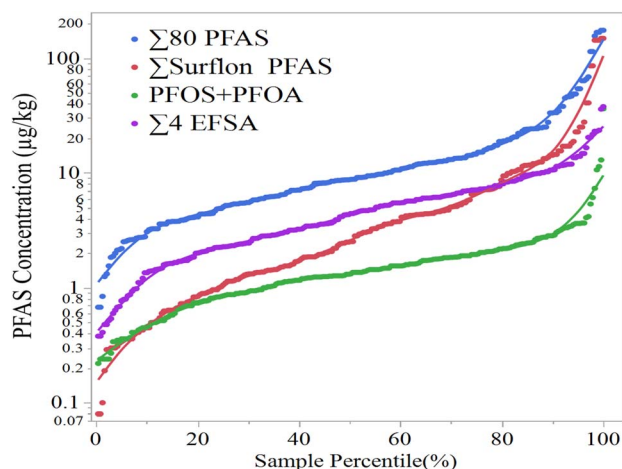


Fig. 7 PFAS Percentile in all soil samples (Surflon:PFNA, PFUnA, PFTrDA), sum of EFSA PFAS is the sum of PFAS regulated by the European Food Safety Authority (PFOA, PFOS, PFNA, and PFHxS).

PFAS in samples within this percentile further confirms industrial contamination. Samples within the 96th–100th percentile ($n = 10$) contain the highest PFAS levels (64–175 $\mu\text{g kg}^{-1}$), suggesting probable PFAS contamination hotspots within Lyon, which could be linked to the industrial activities in this area. Samples with background PFAS levels accounted for 58% of the analyzed samples, while 42% could be classified as contaminated, likely due to industrial influences.

3.4 Exposure and risk assessment

Risk assessment of hazardous materials (substances capable of causing adverse effects) involves evaluating both the extent of human exposure (the degree of contact with the hazardous material) and the resulting risk (the probability of adverse health effects occurring as a consequence of that exposure).⁸⁰ This study evaluated the risks of human exposure to PFAS in eggs and soil.

3.4.1 Estimation of human exposure to PFAS in soil. The CSOIL model was used to estimate the sum of estimated daily intake ($\sum\text{EDI}$) of PFAS from soil for children and adults by

considering overall exposure through soil ingestion, dermal contact, and inhalation.⁶³ The $\sum\text{EDI}_{\text{PFAS in soil}}$ for $\sum 80$ PFAS (Fig. 8A) varied from 0.004–1.17 ng kg^{-1} per day (children) and 0.006–0.18 ng kg^{-1} per day (adults). The $\sum\text{EDI}$ reported in this study was slightly higher than those reported by Zhao *et al.*, with $\sum\text{EDI}_{\text{PFAS in soil}}$ ranging from 0.009–0.07 ng kg^{-1} per day for the $\sum 16$ PFAS in soil.⁸¹ However, we considered a significantly higher PFAS number compared to their study.

With a daily soil ingestion rate estimated at 100 mg per day (adults) and 200 mg per day (children),^{14,64} $\text{EDI}_{\text{soil ingestion}}$ was the primary contributor to the $\sum\text{EDI}_{\text{PFAS in soil}}$ with a percentage contribution ranging from 99.9 and 99.4% for children and adults, respectively (Fig. 8B). Inhalation and dermal exposure routes contributed between 0.5 to <0.01% (Fig. 8B). These findings were similar to those reported by Xie *et al.* ($\text{EDI}_{\text{soil inhalation and dermal}} = 0.012 \text{ ng kg}^{-1} \text{ bw per day}$) for human exposure to PFAS in soils near and far from a fluorochemical plant in China.¹³ These findings established that ingestion is the principal route of human exposure to PFAS in soil, resulting from hand-to-mouth contact with PFAS-

Risk Assessments

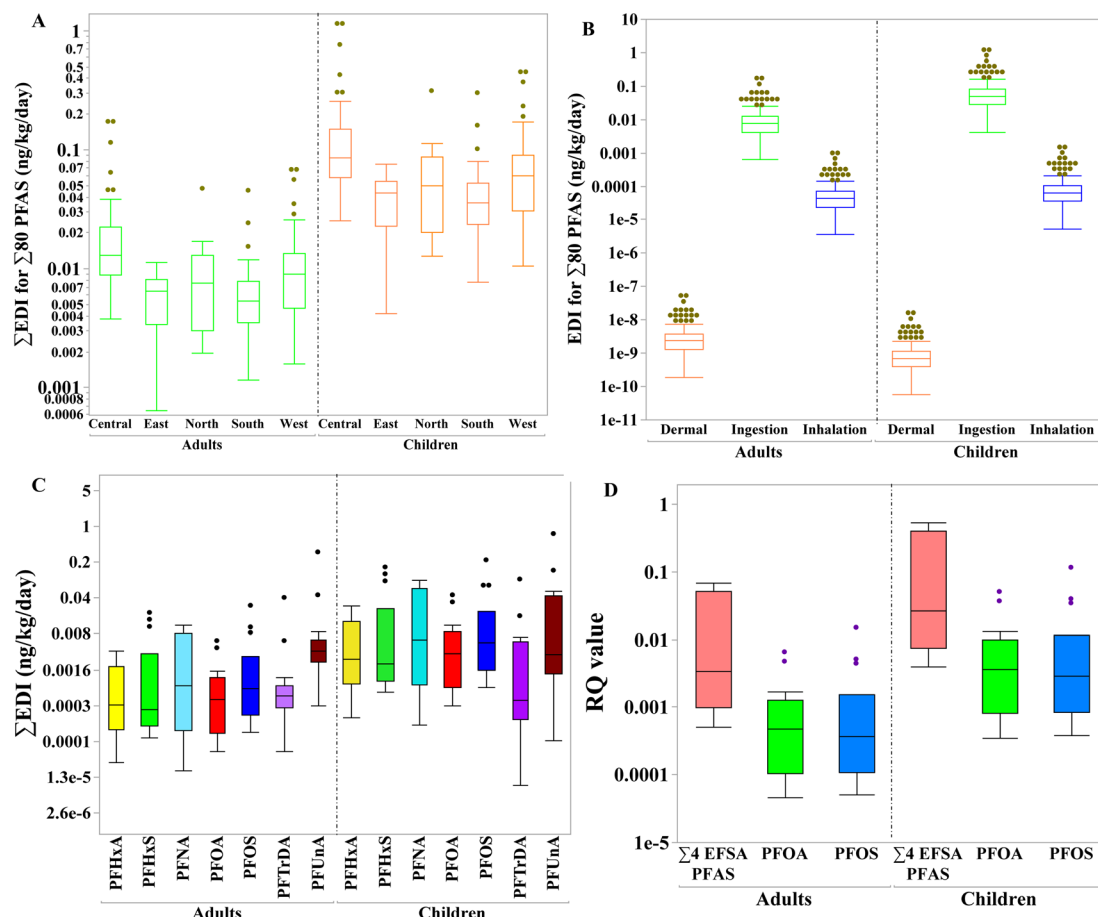


Fig. 8 (A) Sum of estimated daily intakes for $\sum 80$ PFAS in children and adults, (B) estimated daily intakes through different exposure pathways, (C) sum of estimated daily intakes for dominant PFAS, (D) Risk quotient (RQ) values for $\sum 4$ EFSA PFAS, PFOA, and PFOS. RQ values were calculated using the TDI established by EFSA for PFOA ($0.9 \text{ ng kg}^{-1} \text{ bw per day}$), PFOS ($1.9 \text{ ng kg}^{-1} \text{ bw per day}$) and the recent regulation of $0.63 \text{ ng kg}^{-1} \text{ bw per day}$ for the sum of PFOA, PFNA, PFOS, and PFHxS.

contaminated soil, accidental or direct soil ingestion, or consumption of vegetables, fruits or other food contaminated with soil. Children are particularly vulnerable to this pathway due to their habitual hand-to-mouth actions. PFAS can be absorbed into the gastrointestinal tract upon ingestion of PFAS-contaminated soil, consequently entering the circulatory system.⁸²

In contrast to the ingestion exposure route, the negligible contribution from inhalation can be attributed to the non-volatile nature of most PFAS, except some precursor PFAS, such as FTOHs, commonly found in dusts.⁸³ Dermal exposure is minimal due to limited permeability through the skin's outermost layer.⁸⁴

3.4.2 Human risks through PFAS-contaminated soil uptake. The $\sum \text{EDI}_{\text{PFAS in soil}}$ values for individual dominant PFAS detected in the soil (Fig. 8C) were $<1 \text{ ng kg}^{-1}$ per day ($8.7 \times 10^{-6} - 0.7 \text{ ng kg}^{-1}$ per day). The $\sum \text{EDI}$ for PFOA varied from $3.0 \times 10^{-5} - 0.5 \text{ ng kg}^{-1}$ per day (children) and $4.7 \times 10^{-5} - 0.06 \text{ ng kg}^{-1}$ per day (adults), while the $\sum \text{EDI}_{\text{PFAS in soil}}$ for PFOS varied from $0.001 - 0.2 \text{ ng kg}^{-1}$ per day (children) and $9 \times 10^{-5} - 0.03 \text{ ng kg}^{-1}$ per day (adults). Compared with the U.S. EPA reference doses for PFOA (0.03 ng kg^{-1} bw per day) and PFOS (0.1 ng kg^{-1} bw per day), which were proposed in draft form between 2022 and 2023 and finalized in 2024, the maximum $\sum \text{EDI}_{\text{PFAS}}$ values from soil in our study were substantially higher, particularly for children.^{85,86} Furthermore, for PFOA, the EPA derived a cancer slope factor of 0.0293 (ng per kg per day), indicating a potential lifetime cancer risk at extremely low exposure levels. These comparisons suggest that background soil concentrations observed in this study may pose a significant human health risk when evaluated against current USEPA benchmarks. In contrast, the reference doses established by the EFSA were relatively higher than those established by the USEPA. The $\sum \text{EDI}_{\text{PFAS in soil}}$ values obtained for PFOA, PFOS and $\sum 4$ EFSA PFAS were lower than the TDI established by EFSA for PFOA ($0.9 \text{ ng per kg bw per day}$), PFOS ($1.9 \text{ ng per kg bw per day}$)⁸⁷ and $0.63 \text{ ng per kg bw per day}$ for the sum of PFOA, PFNA, PFOS, and PFHxS.⁸⁸ Also, these values were significantly lower than the EDI reported by Xie *et al.* for PFOA ($5.7 - 119 \text{ ng per kg bw per day}$) in an urban soil in China.¹³

The risk quotient (RQ) value for PFOA, PFOS and the sum of four PFAS regulated by EFSA (PFOA, PFNA, PFOS, and PFHxS) were calculated using the ratio of each PFAS $\sum \text{EDI}_{\text{PFAS in soil}}$ and the tolerable daily intake (TDI) established by EFSA for PFOA ($0.9 \text{ ng per kg bw per day}$), PFOS ($1.9 \text{ ng per kg bw per day}$)⁸⁷ and $0.63 \text{ ng per kg bw per day}$ for the sum of PFOA, PFNA, PFOS, and PFHxS.⁸⁸ The RQ values for the $\sum 4$ EFSA PFAS, PFOA, and PFOS were <1 (Fig. 8D). $\text{RQ} < 1$ can be interpreted as intake levels below the threshold that pose a health risk, while values >1 will result in significant health risks. The RQ values recorded in this study indicate that the levels of $\sum 4$ EFSA PFAS, PFOA, and PFOS in the analyzed soil do not pose a significant threat to residents. However, it is crucial to note that the main contributors to total PFAS concentrations in Lyon soils are the Surflon-related PFAS, which are currently not part of the regulated PFAS for which tolerable reference doses are available.

3.4.3 PFAS in soil from chicken feeding area and free-range chicken eggs. The distribution of 14 PFAAs was investigated in domestic soils and corresponding free-range chicken egg samples (Fig. 9). Across the eight paired soil and egg samples analyzed, $\sum 14$ PFAAs ranged from $3.6 - 15.5 \mu\text{g kg}^{-1}$ d.w. (soils) and $2.2 - 20.2 \mu\text{g kg}^{-1}$ wet w. (eggs). Although PFAS concentrations in the free-range chicken egg were higher than recorded in the soils, the statistical test (Tukey Kramer HSD) shows no significant difference between the soils and the paired eggs (Fig. 9). The elevated PFAS concentrations observed in eggs compared to the paired soils could be attributed to the bioaccumulation of PFAS over time as chickens forage on contaminated soil. The detection of Surflon PFAS in the eggs and the lack of statistical difference between Surflon PFAS concentration in eggs and soils further confirmed the influence of industrial activities on PFAS concentrations in Lyon free-range chicken eggs (Fig. 9). Unlike commercial chickens, free-range chickens in residential gardens have uninterrupted access to outdoor enclosures. Consequently, the free-range chickens could be exposed to PFAS by ingesting contaminated soil, soil invertebrates (*e.g.*, worms and insects), dust particles, rainwater or kitchen waste products.⁸⁹ However, Granby *et al.* reported contrasting results, with lower PFAS concentrations in free-range chicken eggs compared to organic eggs collected from farms.⁹⁰

3.4.4 Human exposure to PFAAs through free-range chicken egg consumption. Among the 14 PFAS investigated in our study, 7 PFAS were detected in 100% egg samples with fresh weight concentrations ranging from $0.05 - 7.4 \mu\text{g kg}^{-1}$ (PFDoA), $0.2 - 6.8 \mu\text{g kg}^{-1}$ (PFOS), $0.4 - 5.4 \mu\text{g kg}^{-1}$ (PFNA), $0.2 - 2.6 \mu\text{g kg}^{-1}$ (PFUnA), $0.03 - 2.4 \mu\text{g kg}^{-1}$ (PFDA), $0.01 - 0.2$ (PFHxS), and $0.1 - 0.6 \mu\text{g kg}^{-1}$ (PFOA) (Fig. S5). The concentration of these PFAS in the corresponding soils ranged from $0.01 - 3.0 \mu\text{g kg}^{-1}$ (PFDoA), $0.5 - 1.9 \mu\text{g kg}^{-1}$ (PFOS), $0.3 - 2.9 \mu\text{g kg}^{-1}$ (PFNA), $0.2 - 2.7 \mu\text{g kg}^{-1}$ (PFUnA), $0.1 - 4.3 \mu\text{g kg}^{-1}$ (PFHxS) and $0.02 - 0.3 \mu\text{g kg}^{-1}$ (PFOA). In 2023, the European Union Commission Regulation proposed maximum levels for PFAS in eggs, which are set at $0.30 \mu\text{g kg}^{-1}$ (PFOA), $1.0 \mu\text{g kg}^{-1}$ (PFOS), $0.70 \mu\text{g kg}^{-1}$ (PFNA), $0.30 \mu\text{g kg}^{-1}$ (PFHxS), and $1.70 \mu\text{g kg}^{-1}$ ($\sum 4$ EFSA PFAS).⁹¹ In this study, the recorded values for these PFAS exceeded the proposed maximum levels, with $\sum 4$ EFSA found to exceed limits in 88% of samples, PFOA in 25% of samples, PFNA in 88% of samples, and PFOS in 75% of samples. In contrast, the values for PFHxS remained below the proposed maximum level. In comparison to our study, a similar PFAS footprint was detected in home-produced eggs collected from private gardens located within a 10 km radius of a fluorochemical plant in Antwerp (Belgium);⁸⁹ however, the concentration recorded for PFOS ($0.13 - 241 \text{ ng g}^{-1}$ fresh weight)⁸⁹ was much higher than in our study (PFOS: $0.4 - 5.4 \mu\text{g kg}^{-1}$). The production and use of PFOS in Antwerp have been documented.⁸⁹ In contrast, PFOS was neither used nor produced in the context of fluoropolymer production in Pierre Bénite, except for its possible use for fire drills on this site. A previous study detected a concentration of $4 \mu\text{g L}^{-1}$ of PFOS in the aquifer located south of the chemical platform.⁵²



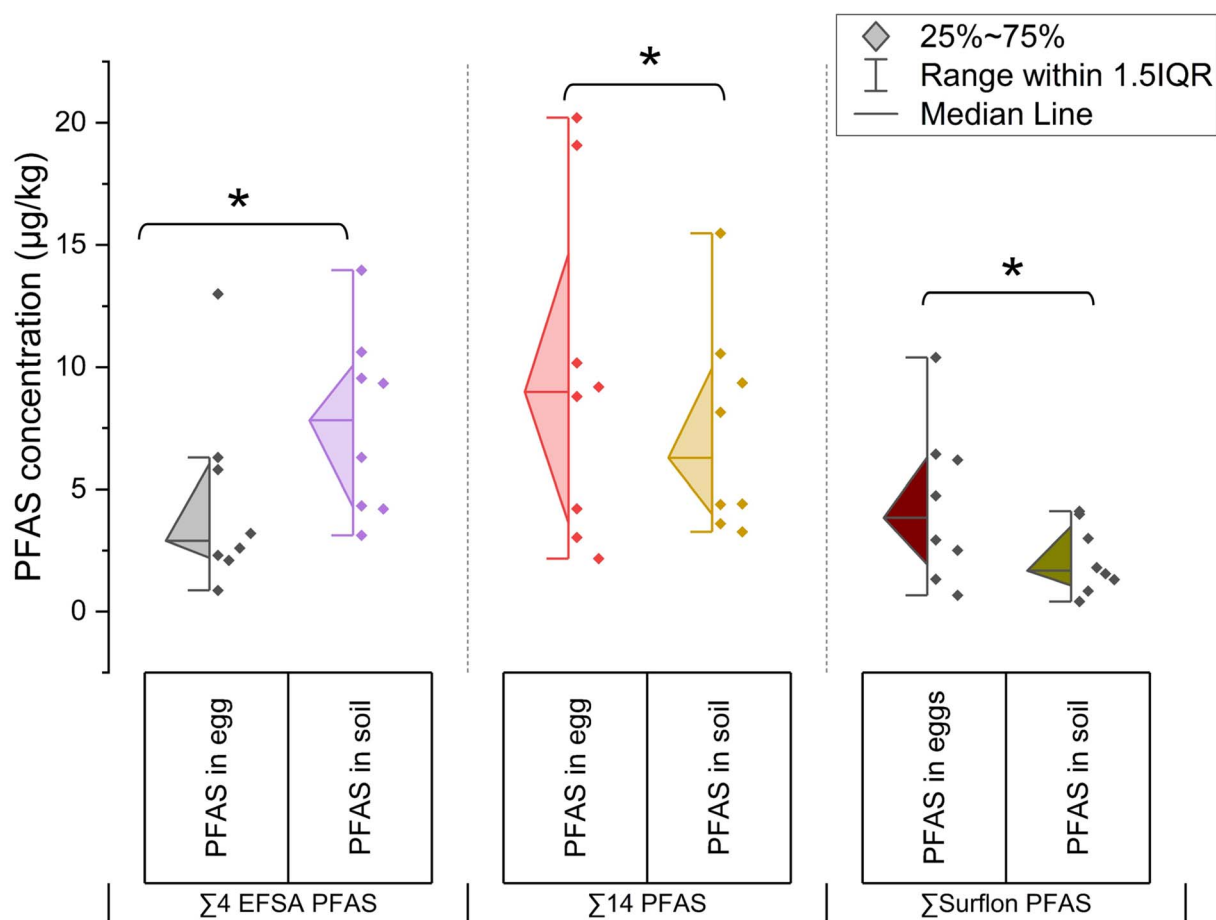


Fig. 9 PFAS concentration in soils from chicken feeding areas ($\mu\text{g kg}^{-1}$ d.w.) and corresponding chicken eggs ($\mu\text{g kg}^{-1}$ fresh weight). A statistical test (Tukey Kramer HSD) showed no significant difference between the soils and the paired eggs.

The ratio of individual PFAS concentrations in eggs and soils was plotted against individual PFAS chain lengths to understand the bioaccumulation potential of various PFAS based on their chain length (Fig. 10). The plot revealed a strong linear correlation between the mean ratio and the PFAS chain length, with a Pearson's r of 0.835 and a p -value of <0.001 . As the PFAS chain length increases, the mean ratio of PFAS concentration in eggs to soil increases.

Due to their high-water solubility, short-chain PFAS are more mobile and likely to leach through the soil rather than adsorb to soil particles. In contrast, long-chain PFAS tend to bind

Long-chain PFAS are more hydrophobic with higher protein-binding affinities, particularly with proteins and lipids found in biological systems. Since eggs contain these proteins and lipids, longer-chain PFAS tend to accumulate in eggs if free-range chickens are exposed to PFAS-contaminated soil or other sources of exposure.^{93–95}

Human exposure to PFAAs through the consumption of free-range chicken eggs can be estimated using the estimated daily intake of PFAS (EDI, ng per kg bw per day), which is generally calculated using eqn (6).⁹⁶

$$\text{EDI} = \frac{\text{Daily egg consumption (g per day)} \times \text{PFAS concentration in eggs (ng g}^{-1}\text{)}}{\text{Body weight (kg)}} \quad (6)$$

hydrophobically to soil organic matter, which can potentially limit their mobility. However, once taken up by an organism, long-chain PFAS tend to bioaccumulate.^{92,93}

In this study, the $\text{EDI}_{\text{PFAS in eggs}}$ was calculated using an average body weight of 25 kg for children (ages 5–11)^{64,97} and 75 kg for adults (≤ 18 years old).⁹⁸ While egg consumption varies across European countries, such as Germany (37 g wet.w per day

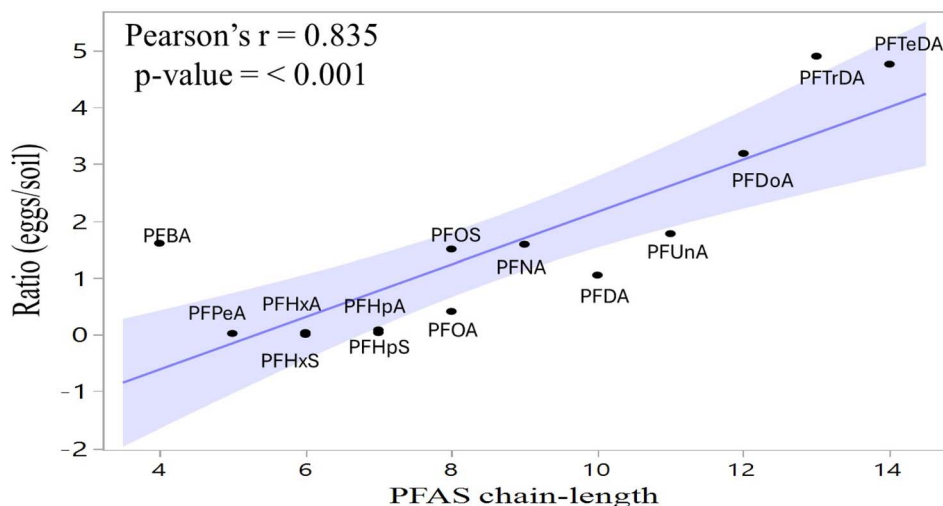


Fig. 10 Correlation between PFAS chain length and the ratio of PFAS concentration in soil from chicken playing area and chicken eggs.

per person)^{99,100} and Denmark (18–24 g wet.w per day per person),¹⁰¹ we assumed an average daily consumption rate of 18 g wet.w. per day per person based on national consumption data for France.¹⁰² However, this may be highly variable and could be much higher for some individuals. The EDI_{PFAS} in eggs, estimated for the $\sum 14$ PFAS, $\sum$ Surflon PFAS, $\sum 4$ PFAS regulated by EFSA, and some other PFAS in free-range chicken eggs, are presented in Table 2. The EDI_{PFAS} in eggs for the $\sum 14$ PFAS analyzed in this study ranges from 1.7–14.6 ng per kg bw per day

(children) and 0.5–4.9 ng per kg bw per day (adults), while the EDI_{PFAS} in eggs for the $\sum$ Surflon varied from 0.5–7.5 ng per kg bw per day (children) and 0.16–2.5 ng per kg bw per day (adults).

3.4.5 Human risk through egg consumption. We evaluated the human risk associated with consuming PFAS-contaminated eggs by comparing the EDI_{PFAS} in eggs with established thresholds set by the EFSA. The reference dose, established as the tolerable daily intake (TDI) by the European Food Safety Authority (EFSA) for PFOS in 2018, was set at 1.9 ng kg⁻¹ bw per day, and 0.9 ng per kg bw per day for PFOA.⁸⁷ The EFSA 2020 regulation proposed a TDI of 0.63 ng per kg bw per day for four PFAS compounds: PFOA, PFNA, PFOS, and PFHxS.⁸⁸ The $\sum 4$ EFSA PFAS recorded in this study varied from 0.62–9.4 ng per kg bw per day (children) and 0.2–2.5 ng per kg bw per day. At maximum concentrations, the EDI_{PFAS} in eggs for $\sum 4$ EFSA PFAS exceeded the established EFSA TDI of 0.63 ng per kg bw per day in both adults and children. For individual compounds, adult exposures to PFOA (0.01–0.14 ng per kg bw per day) and PFOS (0.1–1.6 ng per kg bw per day) were higher than the U.S. EPA reference doses (0.03 ng per kg bw per day for PFOA and 0.1 ng per kg bw per day for PFOS), but remained below the corresponding EFSA TDIs (PFOA: 0.9 ng per kg bw per day; PFOS: 1.9 ng per kg bw per day). In contrast, children's PFOS intake (0.3–4.9 ng per kg bw per day) exceeded the U.S. EPA reference dose across all levels and the EFSA TDI at the maximum PFAS concentration, highlighting the greater vulnerability of children to PFAS exposure. When applying the lower thresholds established by the USEPA,^{85,86} the EDIs recorded for both PFOA and PFOS in this study exceeded the recommended limits across all age groups, especially at maximum PFAS concentration, indicating that all populations considered are at potential health risk. Generally, these findings emphasize that the consumption of home-raised chicken eggs in Lyon may represent a significant source of PFAS exposure, particularly for children.

Fig. 10 also shows that the EFSA target PFAS appear to minimize the risks when compared to the sum of the 14 PFAS measured in the monitoring, or those associated with the

Table 2 Estimated daily intake of PFAS in free-range chicken eggs

EDI _{PFAS} in eggs (ng per kg bw per day)	Children		Adults	
	Min	Max	Min	Max
$\sum 14$ PFAS	1.56	14.55	0.52	4.85
$\sum$ Surflon PFAS	0.48	7.49	0.16	2.50
$\sum 4$ EFSA PFAS	0.62	9.36	0.21	3.12
PFBA	0.09	0.20	0.03	0.07
PFOA ^a	0.03	0.42	0.01	0.14
PFHxS	0.01	0.11	0.00	0.04
PFNA ^a	0.27	3.89	0.09	1.30
PFDA	0.02	1.73	0.01	0.58
PFOS	0.31	4.90	0.10	1.63
PFUnA ^a	0.16	1.87	0.05	0.62
PFDaA	0.04	5.33	0.01	1.78
PFTeDA	0.07	1.73	0.02	0.58
PFTTrDA ^a	0.07	2.09	0.02	0.70

Available data for the minimal risk level (MRL) of PFOA and PFOS

TDI (ng per kg bw per day)	PFOA	PFOS	$\sum 4$ PFAS	References
USEPA	0.03	0.1	X	85 and 86
EFSA	0.9	1.9	0.63	87 and 88
FSANZ	160	20	X	103
ATSDR	20	20	X	104

^a Surflon PFAS.

production of Surflon (which is likely the primary source of contamination around Lyon). Overall, this suggests that consuming contaminated free-range chicken eggs from contaminated areas will significantly increase PFAS intake, posing potential health risks to consumers. These findings highlight the need for continuous PFAS monitoring and prevention.

Conclusion

This study provides clear evidence of ambient PFAS deposition in Lyon soils, with contamination levels varying based on proximity to a primary chemical industrial site that utilizes PFAS. The increased PFAS concentrations observed, particularly near the chemical industries, indicate a direct atmospheric deposition of PFAS to Lyon soils. Additionally, wind patterns influenced PFAS distribution, resulting in high PFAS concentrations in the soils of the western and northern regions, which are located downwind of the Arkema facility. The prevalence of Surflon-related PFAS (PFUnA, PFNA, PFTrDA) in locations closer to the industrial area, along with their decreasing concentrations toward less industrialized zones, further supports a strong link between proximity to emission sources and PFAS distribution. The detection of 6:2 FTS near the Arkema site and PFHxS in all soil samples confirmed a shift in their use in industrial processes within these industrial sites.

The presence of 6:2 FTAB, a zwitterionic PFAS and a significant component of AFFF ($0.1\text{--}52\text{ }\mu\text{g kg}^{-1}$), in the soil near Port Édouard-Herriot, where AFFF was used in 1987, highlights the persistence of this compound. Generally, samples with background PFAS levels accounted for 58% of the total samples analyzed in this study, while 42% could be classified as contaminated, likely due to industrial influence.

Compared with the U.S. EPA reference doses of 0.03 ng per kg bw per day for PFOA and 0.1 ng per kg bw per day for PFOS, the maximum $\sum\text{EDI}_{\text{PFAS in soil}}$ values observed for soil in this study were substantially higher, particularly in children. In contrast, these values were lower than the TDI recommended by EFSA.

The increased PFAS concentrations recorded in the eggs compared to the paired soils in this study revealed the potential bioaccumulation of PFAS in free-range chicken eggs while the chicken forage on PFAS-contaminated soil, with possible contributions from other exposure routes such as contaminated water or feeds, soil invertebrates, rainwater or kitchen wastes. However, the presence of Surflon PFAS in relatively high concentrations strongly suggests that the industrial emission footprint of Surflon PFAS is mainly responsible for the contamination of free-range chicken eggs.

Compared to the proposed maximum levels for specific PFAS in eggs set by the European Union,⁹¹ the concentrations of these PFAS in eggs analyzed in this study exceeded the limits, particularly for PFOA (25% of samples), PFOS (75% of samples), PFNA (88% of samples), and the $\sum 4$ EFSA (88% of samples).

Generally, the $\text{EDI}_{\text{PFAS in eggs}}$ for PFOA and PFOS at maximum exposure levels exceeded the TDIs established by the USEPA. Similarly, the $\sum 4$ PFAS values estimated for all age

groups were higher than the TDI set by EFSA, particularly at maximum PFAS concentrations, indicating a potential health risk for consumers. The EFSA currently have TDI doses established only for the sum of four PFAS, which will need to be updated to include additional PFAS commonly detected in environmental matrices, particularly those prevalent in industrial applications. These findings emphasized the importance of continuous environmental monitoring for a larger number of PFAS, stricter regulatory measures, and further study on PFAS mobility and bioaccumulation in soils and local food sources.

This study also highlights the need for further investigation into the potential contamination of other agri-food products in the affected areas, and the extent to which the widespread groundwater contamination around Lyon may be attributed to leaching from soils or hydrological transfers from contaminated water bodies.

Conflicts of interest

There are no conflicts to declare.

Data availability

The data supporting this article have been included as part of the SI and include the list and details on the analysed PFAS, various statistics and illustrations of the data and the complete dataset with locations. Supplementary information is available. See DOI: <https://doi.org/10.1039/d5va00135h>.

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