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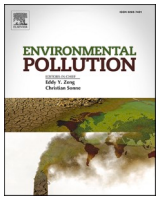

New insights from an eight-year study on per- and polyfluoroalkyl substances in an urban terrestrial ecosystem☆

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ABSTRACT

Per- and polyfluoroalkyl substances (PFAS) were analysed in a high number of terrestrial samples of soil, earthworm, bird eggs and liver from red fox and brown rat in an urban area in Norway from 2013 to 2020. PFOS and the long chain PFCAs were the most dominating compounds in all samples, proving their ubiquitous distribution. Other less studied compounds such as 6:2 FTS were first and foremost detected in earthworm. 8:2 FTS was found in many samples of fieldfare egg, sparrowhawk egg and earthworm, where the eggs had highest concentrations. Highest concentrations for both 6:2 FTS and 8:2 FTS were detected at present and former industry areas. FOSA was detected in many samples of the species with highest concentrations in red fox liver and brown rat liver of 3.3 and 5.5 ng/g ww.

PFAS concentrations from the urban area were significantly higher than from background areas indicating that some of the species can be suitable as markers for PFAS emissions in an urban environment. Fieldfare eggs had surprisingly high concentrations of PFOS and PFCA concentrations from areas known to be or have been influenced by industry. Biota-soil-accumulation factor and magnification calculations indicate accumulation and magnification potential for several PFAS.

Earthworm and fieldfare egg had average concentrations above the Canadian and European thresholds in diet for avian wildlife and predators. For earthworms, 18 % of the samples exceeded the European threshold (33 ng/g ww) of PFOS in prey for predators, and for fieldfare eggs, 35 % of the samples were above the same threshold. None of the soil samples exceeded a proposed PNEC of PFOS for soil living organisms of 373 ng/g dw.

1. Introduction

Per- and polyfluoroalkyl substances (PFAS) have been widely used in many industrial and commercial applications for over 70 years. The chemical and thermal stability of the hydrophobic carbon-fluorine alkyl chain, combined with a hydrophilic functional group, lead to highly useful and enduring properties in surfactants and polymers. Applications include textile stain and water repellents, grease-proof, food-contact paper and other food contact materials used for cooking. Surfactant applications that take advantage of the unparalleled aqueous surface tension–lowering properties include processing aids for fluoropolymer manufacture, coatings, and aqueous film–forming foams (AFFFs) used to extinguish fires involving highly flammable liquids. Numerous additional applications have been described, including floor polish, ski waxes, and water-proof coatings of textile fibers (Glüge et al., 2020). PFAS degrade in the environment only to a very limited extend or degrade to other persistent PFAS. As a consequence, PFAS have been detected worldwide in the environment, wildlife, and humans. A large body of studies and reviews have focused on how these substances are transported in the environment, and to what extent and how humans and wildlife are exposed and their potential toxic effects (Abunada et al., 2020; De Silva et al., 2021; Fenton et al., 2021). Furthermore, the potential for atmospheric long-range transport of a number of PFAS has been demonstrated (Faust, J., 2023; D'Ambro et al., 2021; Lai et al., 2016) and toxic effects on biota including humans were for example discussed by Dickman et al. (2022), DeWitt (2015), Cai et al. (2021) and

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Khazaee et al. (2021). The regulation and policy processes of the PFAS group is in centre of EU's chemicals strategy for sustainability.[1] PFOS, PFOSF, PFOA, their salts and related compounds, are recognized as persistent organic pollutants (POPs) and banned in the Stockholm convention. In Norway, PFOS and PFOA are banned, and the C9–C14 PFCAs and PFHxS[2] (see Table S1) are on the Norway's Priority List of Hazardous substances as well as being included in the candidate list of substances of very high concern for Authorization in ECHA. The POPs Review Committee of the Stockholm convention is currently reviewing long chain PFCAs (LC-PFCAs), their salts and related compounds.[3] The national authorities of Norway, Sweden, Denmark, Germany and the Netherlands have recently submitted a restriction proposal to ECHA to ban a majority of PFAS-substances from products being used and sold within the European Union.[4]

In addition to the well-known PFAS mentioned above, probably several thousands PFAS are on the global market, and the chemical identities of many are confidential (Wang et al., 2017). Emissions and leakage to the environment are unavoidable throughout the whole lifecycle of PFAS containing products. For example, perfluoro-4-ethylcyclohexane-sulfonate (PFECHS) was detected for the first time in atmospherically derived samples and biota, with a potential source being attributed to aircraft hydraulic system leakage and aqueous film forming foams (AFFFs) for fire fighting (MacInnis et al., 2017). Pan et al. (2017) reported the occurrence and bioaccumulation of hexafluoropropylene oxide trimer acid in surface water and fish (Pan et al., 2017). Gebbink et al. (2017), published findings of the PFOA replacement chemical GenX at all downstream river sampling sites with the highest concentration (812 ng/L) at the first sampling location downstream from a production plant in the Netherlands, proving the necessity of measuring for a broad range of emerging PFAS. Recently it was found that when the sulfluramid ETFOSA was applied to soil it led to FOSA, FOSAA and PFOS formation after a couple of weeks (Guida et al., 2023).

Urban environments are known to act as diffuse point sources for pollutants circulating in society, PFAS just being some of them. It has also been found that indoor concentrations of PFAS exceeded significantly those outdoor, suggesting indoor emissions driving outdoor contamination (Goosey and Harrad, 2012). Some knowledge exists of the extent of PFAS emitted to urban ecosystems, but first and foremost to water and wastewater (Kurwadkar et al., 2022; Podder et al., 2021), and very little insight of PFAS in terrestrial urban ecosystems (but see Fremlin et al., 2020, 2023).

In our study we aimed at providing insights to PFAS occurrence, bioaccumulation and magnification potential and risk assessment in terrestrial species in the city of Oslo, Norway. Non-invasive sampling of the species was a prerequisite in our program, especially for bird eggs, since sparrowhawk and tawny owl are protected species. Biomagnification and trophic magnification factors have clear definitions where diet connected species are used in the models. Three species in the project were known to be connected through diet. Earthworm is known to be the major diet of fieldfare, and fieldfare is known to be an important prey of sparrowhawk, but we lacked the PFAS concentrations in whole birds. We therefore wanted to investigate if the more effective and non-invasive sampling of bird eggs also could be used in order to increase the insight on potential trophic magnification.

This study, funded by the Norwegian Environment Agency, has been part of an eight-year monitoring program of contaminants in species belonging to an urban terrestrial ecosystem covering soil, species from low trophic levels (earthworms) to higher trophic levels such as birds (fieldfare, tawny owl and sparrowhawk) as well as mammals (red fox, brown rats).

2. Material & methods

2.1. Sample collection and study site

The following samples and species were collected between 2013 and 2020: 55 earthworms (*Lumbricidae*) (2013–2020), 33 soil samples (2015–2020), 57 fieldfare eggs (*Turdus pilaris*) (2015–2020), 50 sparrowhawk eggs (*Accipiter nisus*) (2014–2019), 38 tawny owl eggs (*Strix aluco)* (2015–2017 and 2020), 66 red fox livers (*Vulpes vulpes*) (2014–2020) and 58 brown rat livers (*Rattus norvegicus*) (2015–2020), see Table 1. The urban sampling area was the urban area of the capital city Oslo of Norway, see Fig. 1. The locations of soil, earthworm (EW), fieldfare (FF) egg and brown rat (BR) samples were mainly in the city centre of Oslo, while sparrowhawk (SH) eggs also had some locations further away; and tawny owl (TO) eggs, and some of the red fox (RF) animals, were in the border area of the city municipality, up to 40 km away from the city centre (see Figs. S1–S6). Samples from background areas were available for the years 2013 and 2014. The background areas for the earthworm were Åmotsdalen (Dovre) in 2013 and Aust-Agder and Telemark in 2014. The latter area was also the background area for sparrowhawk eggs collected in 2014. Background data for fieldfare egg in 2014 were from Åmotsdalen, Dovre, and red fox in 2014 were from Oppdal, see Figs. S1, S3, S4 and S5.

On average, one soil and one earthworm sample were collected at five locations each year. From year 2015, soil and earthworm were collected at the exact same sites. Up to ten samples were collected for the bird eggs and liver samples (red fox and brown rat), dependent on the availability of samples. Each sample of fieldfare egg contained two eggs per nest per year.

Sampling took place during April/May for bird eggs, June to August for soil and earthworm, fall for red fox, and late fall/wintertime for brown rat. Fieldfare eggs were collected in close vicinity of soil and earthworm locations, depending on occurrence of nests. The upper layer of 0–20 cm of soil was sampled at each site where three sampling spots in an area of few meters comprised one pooled soil sample. Earthworm samples consisted of at least 10–15 individuals per site. To purge their guts, earthworms were kept in aluminium covered plastic containers, and lined with moist paper sheets for three days before being stored at − 20 °C. The soil, earthworm and liver samples were first packed in aluminium foil and inserted in plastic containers or plastic bags before storage at − 20 °C.

Bird eggs were collected under permission of the Norwegian Environment Agency. The first laid egg is known to have higher contaminant concentration than second and third laid eggs; but the laying order of the eggs was not taken into account when collecting the eggs to avoid disturbing the nest more than necessary. The eggs were kept individually in polyethylene bags in a refrigerator (+4 °C), before being shipped. The whole content of the eggs was removed from the shell, homogenized and transferred to clean glass vials for storage at − 20 °C.

Brown rats were caught in residential areas of Oslo during fall and wintertime using clap-traps. The traps were usually inspected daily, and the rats were placed in the freezer as fast as possible on the day of collection. Red foxes were collected from legal hunting in the forest surrounding the city and accidental roadkill. All samples from all species were stored at −20 °C until homogenization and sample preparation.

2.2. PFAS analysis

The following PFAS were targeted (see Table S1): perfluorooctanesulfonamide (FOSA), perfluorobutanesulfonate (PFBS), perfluoropentanesulfonate (PFPeS), perfluorohexanesulfonate (PFHxS), perfluorooctanesulfonate (PFOS), perfluorononanedsulfonate (PFNS), perfluorodecanesulfonate (PFDS), perfluorohexanoate (PFHxA),

[1] https://echa.europa.eu/hot-topics/perfluoroalkyl-chemicals-pfas.
[2] https://echa.europa.eu/documents/10162/40a82ea7-dcd2-5e6f-9bff-6504c7a226c5.
[3] https://chm.pops.int/Implementation/IndustrialPOPs/PFAS/Overview/tabid/5221/Default.aspx.
[4] https://echa.europa.eu/da/-/echa-publishes-pfas-restriction-proposal.

Table 1
PFAS concentrations (ng/g ww) for earthworm (EW), Fieldfare egg (FF), Sparrowhawk egg (SH), Tawny owl egg (TO), Red fox liver (RF), Brown rat liver (BR) and soil (ng/g dw). Mean; Median (separated by semicolon) and min-max range below, are given for PFAS with more than 60 % detection rate in the various samples from the urban area (Oslo). Compounds not fulfilling 60 % detection in any sample are not shown, and min-max range are shown for other compounds with detections below 60%. n: number of samples (included LOD values). LOD values were substituted with 0.5*LOD and are marked in italic.

	Soil n = 33 (2015–2020)	EW n = 55 (2013–2020)	FF n = 57 (2015–2020)	SH n = 50 (2014–2019)	TO n = 38 2015–17; 2019	RF n = 66 (2014–2020)	BR n = 58 (2015–2020)
PFHxS	21 % DF	2.81; 1.28	0.37; 0.26	0.54-;0.25	53 % DF	0.24; 0.17	36 % DF
	0.001–3.76	*0.001*–40.5	*0.001*–2.70	*0.001*–4.25	*0.001*–3.83	*0.001*–2.07	*0.001*–2.96
PFHpS	15 % DF	27 % DF	0.49; 0.22	0.72; 0.38	55 % DF	0.10; 0.06	43 % DF
	0.001–0.72	*0.001*–14.3	*0.001*–7.91	*0.001*–6.16	*0.001*–0.60	*0.001*–0.73	*0.001*–3.16
PFOS	8.64; 1.33	42.2; 8.53	56.3; 23.3	53.5; 31.8	14.4; 9.73	16.1; 10.0	46.6; 22.9
	0.01–162	*0.05*–955	*0.007*–601	3.18–367	0.57–61.0	1.35–144	1.33–272
PFDS	3 % DF	11 % DF	3.04; 0.51	1.53; 0.60	0.31; 0.08	25 % DF	2.61; 0.25
	0.002–0.70	*0.001*–4.21	*0.02*–33.0	*0.001*–16.0	*0.002*–1.83	*0.001*–1.84	*0.001*–27.1
PFHpA	0.21; 0.11	1.50; 0.43	35 % DF	18 % DF	0 % DF	39 % DF	0 % DF
	0.003–0.91	*0.001*–27.3	*0.002*–0.87	*0.001*–0.86		*0.001*–0.86	
PFOA	0.76; 0.38	1.65; 0.73	0.81; 0.55	0.96; 0.48	18 % DF	0.22; 0.17	52 % DF
	0.03–3.34	*0.003*–6.99	*0.018*–6.25	*0.001*–5.54	*0.001*–3.19	*0.001*–0.67	*0.001*–10.1
PFNA	0.27; 0.13	0.54; 0.29	1.06; 0.79	1.31; 0.91	0.42; 0.14	1.17; 1.05	2.46; 0.76
	0.004–1.68	*0.003*–2.87	*0.018*–5.03	0.13–4.85	0.01–5.14	*0.06*–3.26	*0.04*–35.4
PFDA	0.23; 0.07	0.86; 0.27	2.86; 1.73	2.14; 1.53	0.97; 0.57	1.30; 0.85	3.84; 1.62
	0.01–1.59	*0.003*–6.86	*0.09*–18.3	0.12–9.18	0.05–10.4	*0.001*–4.77	*0.001*–20.3
PFUnDA	58 % DF	5.45; 0.44	2.90; 2.20	3.68; 2.59	1.20; 1.02	1.36; 0.69	2.27; 1.02
	0.001–5.31	*0.003*–261	*0.01*–11.7	0.36–16.0	0.08–7.94	0.03–9.66	0.19–9.83
PFDoDA	36 % DF	3.42; 0.83	9.26; 6.16	7.66; 5.85	2.62; 1.20	0.80; 0.34	5.07; 1.89
	0.001–0.27	*0.003*–63.5	*0.001*–59.3	0.32–34.3	*0.01*–38.1	*0.001*–9.63	0.19–44.8
PFTrDA	18 % DF	3.51; 1.12	7.12; 5.58	9.06; 5.39	2.04; 1.16	0.85; 0.37	2.52; 0.60
	0.002–0.54	*0.003*–107	*0.002*–60.7	0.61–59.1	0.20–10.6	*0.001*–14.7	0.01–14.1
PFTeDA	12 % DF	2.37; 1.28	9.41; 5.81	8.21; 5.99	1.98; 0.74	0.39; 0.14	2.39; 0.58
	0.002–0.15	*0.003*–24.5	*0.002*–137	0.42–36.7	0.10–34.8	*0.001*–3.07	*0.001*–18.6
PFHxDA[c]	0 % DF	0.62; 0.35	1.14; 0.37	0.69; 0.53	11 % DF	31 % DF	18 % DF
		0.01–2.56	*0.01*–26.7	*0.004*–3.66	*0.004*–0.23	*0.001*–2.47	*0.01*–0.48
FOSA	0 % DF	32 % det	0.11; 0.06	0.15; 0.06	21 % DF	0.27; 0.06	0.38; 0.08
		0.001–0.25	*0.001*–1.04	*0.001*–2.12	*0.001*–0.58	*0.001*–3.32	*0.01*–5.51
6:2 FTS[d]	9 % DF	6.34; 0.09	16 % DF	10 % DF	18 % DF	17 % DF	28 % DF
	0.005–0.89	*0.001*–115	*0.004*–6.87	*0.006*–0.24	*0.001*–5.55	*0.001*–0.97	*0.001*–4.40
8:2FTS[e]	4 % DF	0.36; 0.24	2.47; 0.34	2.37; 0.52	46 % DF	30 % DF	52 % DF
	0.007–0.08	*0.001*–1.10	*0.007*–55.1	0.01–47.6	*0.004*–1.90	*0.001*–8.15	*0.001*–16.4
sumPFCA[a]	2.12; 1.04	20.3; 6.85	34.4; 27.2	33.4; 24.5	9.75; 5.51	6.52; 4.67	19.8; 8.73
	0.15–6.99	0.93–389	2.01–316	2.02–140	0.59–110	0.91–44.7	1.06–113
sumPFAS[b]	11.4; 3.56	71.4; 18.0	97.2; 58.6	91.5; 63.7	25.5; 20.1	23.7; 16.3	71.2; 33.1
	0.52–173	1.59–1087	3.25–691	6.03–480	1.24–157	2.26–181	3.17–347

DF: Detection frequency.
[a] sumPFCA.: PFHxA, PFHpA, PFOA, PFNA, PFDA, PFUnDA, PFDoDA, PFTrDA, PFTeDA, PFHxDA.
[b] sumPFAS: FOSA, PFBS, PFPS, PFHxS, PFHpS, PFOS (L-PFOS), PFNS, PFDS, PFHxA, PFHpA, PFOA, PFNA, PFDA, PFUnDA, PFDoDA, PFTrDA, PFTeDA, PFHxDA, 6:2 FTS, 8:2FTS.
[c] PFHxDA analysed 2017–2020.
[d] 6:2 FTS analysed 2015/2016-2020.
[e] 8:2 FTS analysed 2016–2020.

perfluoroheptanoate (PFHpA), perfluorooctanoate (PFOA), perfluorononanoate (PFNA), perfluorodecanoate (PFDA), perfluoroundecanoate (PFUnDA), perfluorododecanoate (PFDoDA), perfluorotridecanoate (PFTrDA), perfluorotetradecanoate (PFTeDA), perfluorohexadecanoate (PFHxDA), 6:2 fluorotelomer sulfonic acid (6:2 FTS), 8:2 fluorotelomer sulfonic acid (8:2 FTS), where details on the extraction and analytical method can be found in Herzke et al. (2023) and Jouanneau et al. (2022). In brief, 1–2 g of homogenated sample was extracted with methanol, concentrated and treated with suspensive Envicarb. After centrifugation, aliquots were added to aqueous ammonium acetate buffer and analysed by LC/MS/MS. Isotopic dilution method was used, and all concentrations are stated on a wet weight basis. Contaminant analyses were conducted at NILU, The Fram Centre, Tromsø, Norway.

2.3. Quality control

The QA/QC measures applied followed the procedure described in Herzke et al. (2023). In brief, blank samples and a standard reference material (human serum INSPQ within the Arctic Monitoring and Assessment Program ring test) were used to assure the quality and control for repeatability and precision of the targeted PFAS method. One blank and one standard reference material were analysed every 10 samples to verify quality of the prepared samples, test reproducibility and precision of the method (see SI for more information).

2.4. Stable isotopes of N ($\delta^{15}N$) and C ($\delta^{13}C$)

The analysis was performed by IFE (institute for Energy Technology) after the method of Dvergedal et al. (2023). Briefly, approximately 1.0 mg of each sample was transferred to a Sn capsule. The capsules with sample were combusted in the presence of O2 and Cr2O3 at 1700 °C in a EA1110 elemental analyser from Thermo Scientific. Reduction of NOx to N2 was done in a Cu oven at 650 °C. H2O was removed in a chemical trap of Mg(ClO4)2 before separation of N2 and CO2 on a 2 m Poraplot Q GC column. N2 and CO2 were directly injected on-line to a Delta XP isotope ratio mass spectrometer (IRMS) from Thermo Scientific, for determination of $\delta^{13}C$ and $\delta^{15}N$ along with the weight % of C and N (Accuracy and precision, see SI).

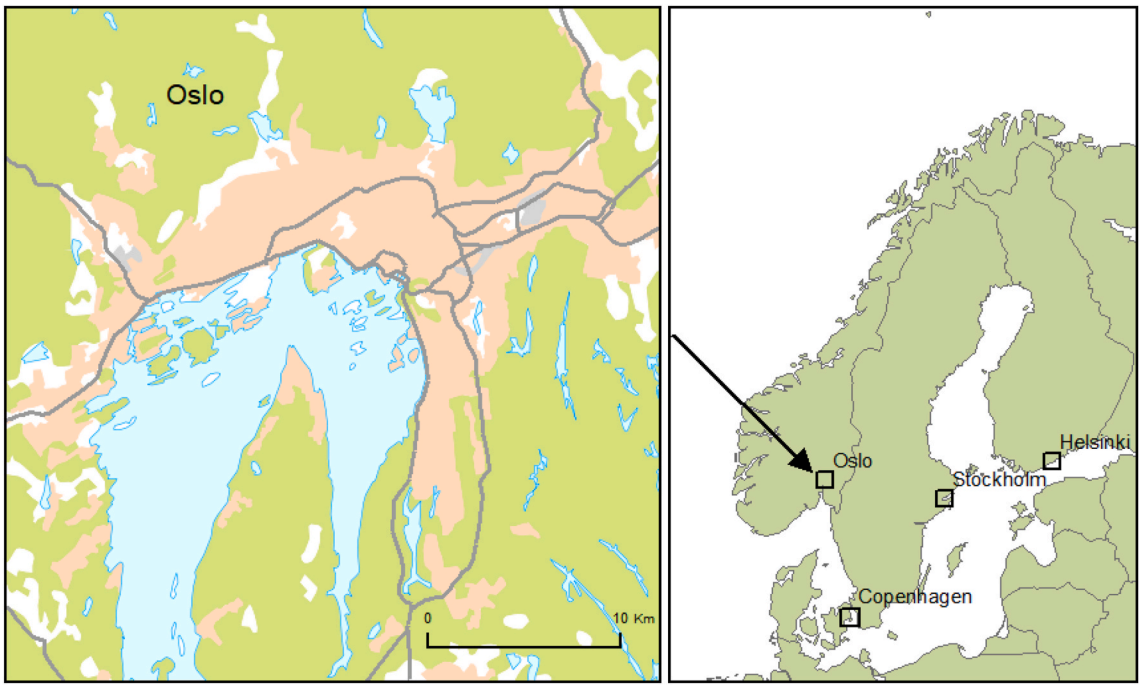

Fig. 1. Map of Norway and the sampling area around the capital city Oslo. Urban areas (densely populated; pink, industry; grey), parks and agriculture (white), forest (green) and highways (grey lines).

2.5. Bioaccumulation and magnification potential

2.5.1. BSAF

Biota-soil-accumulation factor (BSAF) with wet weight concentration in earthworm (EW) divided by soil on a dry weight basis, BSAF = C_{EW} (ng/g ww)/C_{Soil} (ng/g dw), was calculated as done for PFAS in freshwater sediment and biota (Munoz et al., 2022). Only detected concentrations in both soil and earthworm in the same location per year were used. Total organic carbon content in soil was only available for 2016 samples and not used in the calculations.

2.5.2. Prey-predator relationship and magnification

It is well known that earthworm is the main food resource for fieldfare (Arvidson, 2023; Haas, 1985; Wiklund, 1984). The sparrowhawk is a specialist predator on small bird species, and fieldfare is known to be one of the avian prey species of sparrowhawks in southern Norway (Selås, 1993), see Figure S7. Hence, earthworm, fieldfare and sparrowhawk are linked in a food chain, and we assumed that they represent three trophic levels, (Figure S7). As such, we used this food chain approach to discuss and evaluate the magnification of PFAS and relation to stable $\delta^{15}N$ isotope (a metric of trophic position). The other terrestrial species in our study are much weaker connected through diet, and none of them form a chain of three species. Brown rats are opportunistic omnivores that will consume almost anything in the urban areas including human food waste. Red foxes that visit the city may potentially prey on these brown rats, but they are also generalists and their diet may include invertebrates, birds and eggs, rodents, rabbits, roe deer fawn, hunting offal (guts etc) and human waste (Jahren et al., 2020). Tawny owls prefer voles and other small rodents, such as wood mice, in their diet (Vik, 2017). Furthermore, they breed outside the city centre and they don't go inside the city centre to hunt rats. This differs from sparrowhawks, which may breed in parks inside and forest areas outside the city centre, and are commonly observed hunting small birds inside the city centre (Artsobservasjoner, 2024).

For sampling of bird, bird egg was chosen to fulfil ethical and non-invasive sampling. We do not have the information about the relationship of PFAS concentrations from bird egg to whole bird, but Jouanneau et al. (2022) found linear relationship between female plasma and their eggs for most PFAS compounds, indicating significant maternal transfer of PFAS to bird eggs. Especially for the first laid egg, but levels were only 73% in the second laid egg (Jouanneau et al., 2022) Egg-levels of other types on contaminants are also known to reflect the contamination of the egg-laying female (Bianchini et al., 2022; Cifuentes et al., 2003), and in a study of great tits, organochlorine egg concentrations were found to be similar to concentrations in adipose tissue of adult great tits (Dauwe et al., 2006). PFAS levels in bird eggs were therefore used as approximation for PFAS levels in adult female bird and may to an extent also resemble the concentrations in young chicks. Sparrowhawk is known to prey on both adult birds and chicks.

Magnification factor from earthworm to fieldfare egg, C_{FFegg}/C_{EW} was calculated due to the fact that earthworm is a major diet of fieldfare. Calculations were done for locations were both earthworm and fieldfare egg were sampled annually, and only for detected concentrations in both species.

2.5.3. Trophic magnification

The relationship between the stable nitrogen $\delta^{15}N$ values and PFAS concentrations were evaluated. Fieldfare eggs were not sampled in 2014, and no sparrowhawk eggs in 2020; calculations were therefore carried out for the period of year 2015–2019 where all three species were sampled each year, although only two samples of sparrowhawk eggs in 2019. These three species were sampled in the spring and summer months over the years. The three species were habituating a larger area of the city of Oslo reflecting an average for the general urban area, and over the period of 5 years.

The trophic level can be determined from stable N isotope ratios, $\delta^{15}N$ (Borgå et al., 2012). Our TL equations are based on the approach for determining trophic magnification factors for application under the European Union Water Framework Directive (Kidd et al., 2019; Huang et al., 2022), i.e. TL (consumer) = 2 + ($\delta^{15}N$− $\delta^{15}N_{baseline}$)/3.4, where 3.4 is the isotopic enrichment factor ($\Delta^{15}N$) between the trophic levels, with reference to freshwater ecosystems (Post, 2002). For comparison, in a Canadian terrestrial food web study it was determined and used an enrichment factor of 2.88 (Fremlin et al., 2020; Fremlin et al., 2023). We

used the same TL equation, but with an isotopic enrichment factor of 2.4 based on the estimated $\delta^{15}N$ enrichment from diet to bird in common cormorants (Mizutani et al., 1991), and also since average $\delta^{15}N$ difference between fieldfare (i.e. egg) and the major prey earthworm was 2.4.

Due to the use of bird eggs and small $\delta^{15}N$ gap between fieldfare egg and sparrowhawk egg, and as such not fulfilling the requirements of a TMF calculation (Borgå et al., 2012; Kidd et al., 2019), our TMF values should be interpreted as an indicative TMF, and first and foremost if TMF is higher or lower than 1.

Indicative TMF for the relationship earthworm (EW)- fieldfare (FF) egg -sparrowhawk (SH) egg was calculated from the slope of a regression between the chemical concentration and trophic level (TL) of the three species.

The trophic level (TL) per species was calculated per sample for each species relative to the species representing the lowest position, assuming an isotopic enrichment factor $\Delta^{15}N$ of 2.4. Earthworm was used as a base level and assumed at trophic level of 2 in agreement with the results from Potapov et al. (2019), and as used in the terrestrial study of Fremlin et al., (2020); Fremlin et al. (2023).

The following equations were used to estimate the TL per sample of the three species:

$$TL_{EW} = 2 + (\delta^{15}N_{EW} - \delta^{15}N_{EWmean})/2.4$$

$$TL_{FF} = 2 + (\delta^{15}N_{FFegg} - \delta^{15}N_{EWmean})/2.4$$

$$TL_{SH} = 2 + (\delta^{15}N_{SHegg} - \delta^{15}N_{EWmean})/2.4$$

TMFs were calculated as the power of 10 of the slope (b) of the linear regression between log concentration and the sample's TL. PFAS are not hydrophobic compounds (Kelly et al., 2009), and calculations in this study were performed on wet weight basis.

Log [compound] = a + *b*TL, Indicative-TMF = 10^b

To ensure low uncertainty in the estimations, the TMFs were only calculated for compounds with a detection frequency of 60 % or higher. These indicative TMF values should be interpreted as an approximate to reveal if TMF is likely to be above or below 1, and as an average for an area covering the larger surroundings of Oslo city over the time span of year 2015–2019.

2.6. Data treatment and statistics analysis

Concentrations below LOD were substituted by ½*LOD values for statistical analysis. Such arbitrary censorship of less-than values can be problematic, since several descriptive statistical parameters (mean, median, var, sd, se) as well as test statistics (t, r, p) can be affected if a large fraction of non-detects is substituted. We therefore chose to use a conservative criterion and only provide descriptive statistics or statistical tests on compounds with >60% detected, leaving us with very few substituted values for most compounds. The median would, thus, be retained in all cases and the mean would also be very close to the true value. For compounds with <60% detection, the detection rates are given to demonstrate that the detection rate was low, along with max and min values only. This conservative and robust approach will limit the potential effect of applied censorships and substituted values.

We applied the software R (R Core Team, 2020) for statistical computation of TMF, visualization of isotopic niche space, and making whisker and box plot, and the software Microsoft Excel (Microsoft Corporation, 2020) to make bar charts and relative frequency bar charts.

Isotopic niche spaces were calculated with $\delta^{15}N$ and $\delta^{13}C$ values from years 2015–2019 of earthworm (whole animal), fieldfare (eggs) and sparrowhawk (eggs) and illustrated with ellipses covering 95% of the stable isotope data for each species (SIBER package in R, Jackson et al., 2011). Stable isotope means were compared using a post hoc test (package emmeans in R, Lenth, 2023). We used the base package in R (R Core Team, 2020) to calculate the TMF values of PFAS (2015–2019 earthworm, fieldfare egg and sparrowhawk egg), along with Pearson correlation coefficient (R) and coefficient of determination (R^2). The slope estimate (b) was obtained for the linear regression of log compound on trophic level, and significant p-values ($p < 0.05$) mean that 'b' is significantly different from zero. Positive values of 'b', thus, indicate magnification, while negative values indicate elimination.

PFOS concentrations in species with both background and urban data were visualized with whisker and box plot (base package in R, R Core Team, 2020). The upper and lower boundaries of the box are representing the 25th (Q1) and 75th percentile (Q3). The whiskers represent the minimum and maximum values without outliers. Outliers are values outside 1.5 x inter quartile range (IQR) from Q1 or Q3, respectively.

3. Results and discussion

3.1. PFAS in urban terrestrial environments

The PFAS with highest detection frequencies (DF) in the various samples were PFOS, PFOA, PFNA and PFDA (Table 1). The DF of PFOS was 100 % in all the samples from Oslo, except for one earthworm and one fieldfare sample.

As shown in Table 1 and illustrated by Fig. 2; fieldfare egg from Oslo contained the highest average $sumPFAS_{20}$ concentrations close to 100 ng/g, closely followed by sparrowhawk egg. Earthworms and brown rat liver had average $sumPFAS_{20}$ of approximately 71 ng/g ww, followed by tawny owl egg (25.5 ng/g ww) and red fox liver (23.7 ng/g). When comparing median values of $sumPFAS_{20}$, sparrowhawk egg had slightly higher concentration than fieldfare egg, which was followed by brown rat liver, tawny owl egg earthworm and red fox liver. When evaluating the PFAS composition in the samples (Fig. 2), PFOS is dominating in all samples. However, when comparing the PFCAs only, differences between mammalian and avian samples become clear. While bird eggs are more characterised by LC-PFCAs with carbon chain length of C_{12}, C_{13} and C_{14}, we find PFCAs from C_9 to C_{13} dominating the PFCA pattern in mammalian liver samples. Both varying PFCA exposure as well as different translocation properties of PFCAs in liver and eggs could be the explanation. The highest C_{12}, C_{13} and C_{14} concentrations of PFCAs in fieldfare egg were detected in a popular skiing area (Holmenkollen).

3.1.1. Soil

The soil samples from Oslo revealed fewer detected PFAS compared to the biological samples. PFOS was detected in all soil samples with an average of 8.64 ng/g dw (Table 1). Except for PFOS, the other perfluoroalkane sulfonates had detection rates in the range of 0–21 %. PFBA was not detected, while PFOA was detected in all the soil samples followed by PFDA (85 %), PFUnDA (82 %), PFHpA (73 %) (for additional DF see Table 1). In soil samples, the highest concentrations of $sumPFAS_{20}$ and PFOS were measured at the industry location Alna during the years 2016–2018.

Topsoil samples collected from 2017 to 2018 in China were analysed for various PFAS (Ma et al., 2022). The selected sampling sites were related to background or residential. Mean PFOA (0.47 (0.023–1.45) ng/g dw) dominated over mean PFOS (0.10 (<MDL-1.62) ng/g dw). These concentrations were lower than the data from the Oslo urban area, especially for PFOS (see Table 1). Higher concentrations of PFOS and PFOA in various soil samples from other countries and areas have been reported in a recent review by Cai et al. (Cai et al., 2021 and references herein). The sumPFCA and sumPFSA concentrations from Europe were 0.06–3.64 ng/g dw and <LOD-3.27 ng/g dw, respectively, and comparable to our soil median value for sumPFCA and sumPFSA from Oslo, indicating similar urban sources of PFAS throughout Europe.

PFOS concentration in soil samples (n = 12) from Canada varied from 0.38 to 11.3 ng/g dw (Fremlin et al., 2023). Two soil samples from Richmond, Canada had highest concentrations of 3.63 and 11.3 ng/g dw (Fremlin et al., 2023). Soil from six locations in Belgium, in an area not far away from the 3 M fluorochemical plant in Antwerp, had PFOS

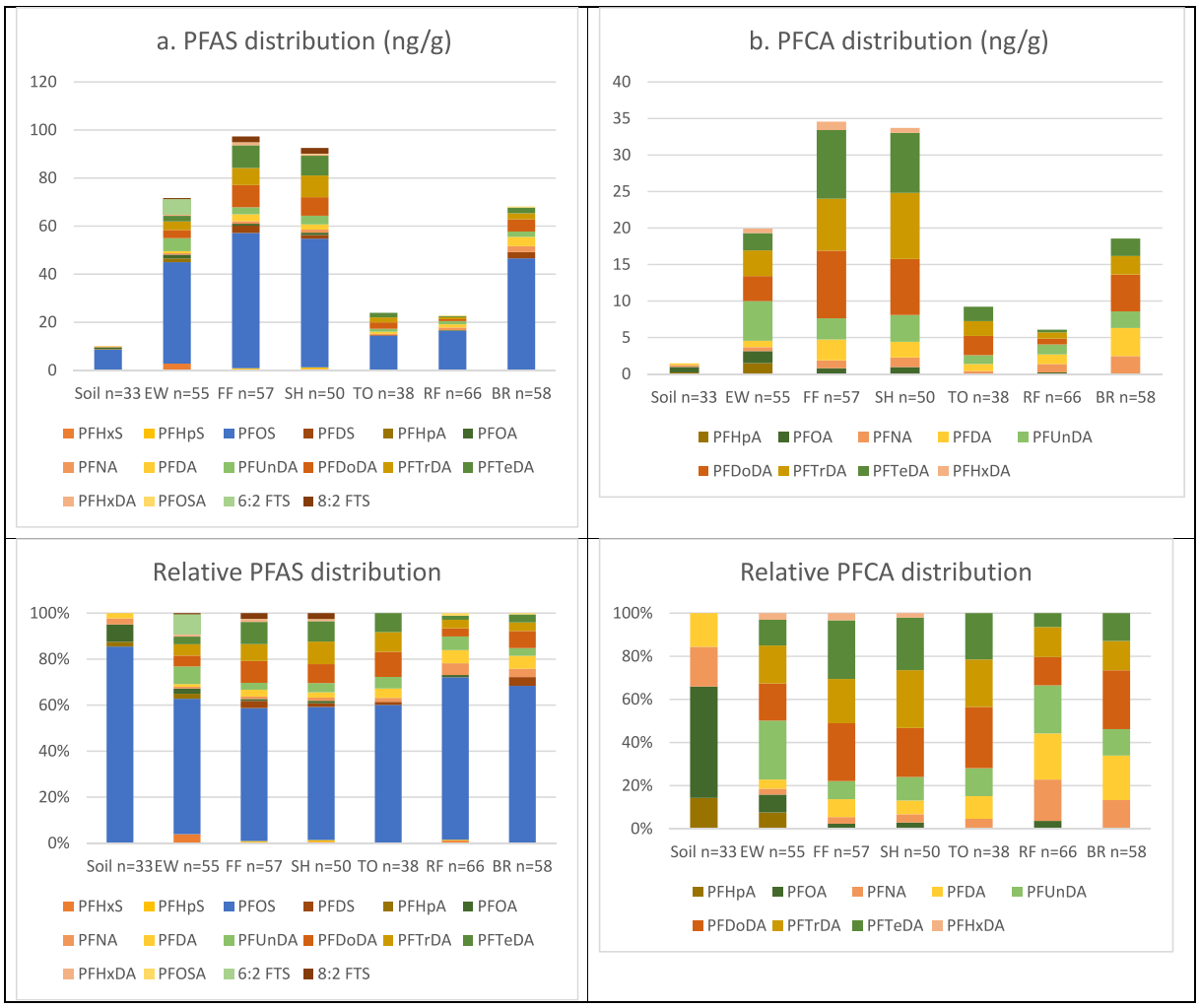

Fig. 2. Distribution shown as average concentrations of PFAS (2a) and PFCA (2b) compounds, and their relative distribution in soil, earthworm (EW), Fieldfare egg (FF), Sparrowhawk egg (SH), Tawny owl egg (TO), Red fox liver (RF) and Brown rat liver (BR). All biota samples in ng/g ww and soil in ng/g dw.

concentrations in the range of 2.3 ± 1.10 to 12.6 ± 5.6 ng/g dw (Groffen et al., 2023).

3.1.2. *Earthworm*

For earthworms, accumulation of contaminants do not only imply a risk to the earthworm population, but also to many vertebrate species feeding on them. The accumulation of pollutants is greatly influenced by physicochemical properties of the pollutants and environmental conditions, and may vary largely among the different PFAS (Zhao et al., 2013).

A larger number of PFAS were detected in the earthworm samples from Oslo. PFOS had an average concentration of 42.2 ng/g (median 8.53 ng/g) due to some very high concentrations at industrial sites; i.e the maximum values in Table 1. The site Alna had highest sumPFAS$_{20}$ (100–1164 ng/g ww) and PFOS (69–1030 ng/g ww) concentrations in the period 2016–2019. PFUnDA dominated the PFCA pattern with an average concentration of 5.45 ng/g and maximum concentration of 261 ng/g ww. Although PFOS concentrations at the present industrial area (Alna) and near the former landfill (Grønmo) had lower concentrations in earthworms in the later years (2018–2020), they were still 5–10 times higher than the average of the other locations. One earthworm sample from the former airport Fornebu in 2017 had higher PFUnDA (261 ng/g) than PFOS (159 ng/g) and a high concentration of PFTrDA (107 ng/g ww), and another sample from a skitrack area (Voksenkollen) in 2016 had elevated 6:2 FTS of 115 ng/g ww compared to PFOS (33.5 ng/g) in the same sample. The most important pollution point sources for high concentration of PFOS in earthworm and fieldfare egg are shown in Fig. S2.

Soil, earthworms (*Eisenia fetida*) and Bank voles (*Myodes glareolus*) were investigated in a forest area 15 km from the city of Trondheim, Norway, and a skiing area (Grønnestad et al., 2019). The average PFOS in soil and earthworm samples near Trondheim were from 25 to 60 times lower than PFOS from the Oslo urban area (Table 1), and the PFOS levels in earthworms near Trondheim were comparable to the non-urban PFOS levels in our study. In the Trondheim study, higher concentrations of PFCA compounds were detected at sites near skiing areas as also observed in samples from skiing areas in the Oslo urban study for soil and earthworm.

Earthworm samples (n = 12) from a Canadian study of a terrestrial ecosystem (Fremlin et al., 2023) had much lower concentrations than the Oslo earthworm samples.

3.1.3. *Bird eggs*

Among the bird species, highest average and median concentrations of PFOS, sumPFCA$_{10}$ and sumPFAS$_{20}$ were detected in the fieldfare and sparrowhawk eggs compared to tawny owl eggs. The concentrations of PFOS in fieldfare and sparrowhawk eggs were comparable, but fieldfare eggs revealed higher average and maximum concentrations for PFOS, and also for some of the other PFAS, over the years 2014–2020. The highest PFOS concentrations in fieldfare eggs were detected at the

former landfill Grønmo. It was expected that PFAS concentrations would be higher in sparrowhawk compared to fieldfare since sparrowhawk is known to be a predator of fieldfare, other thrushes, and smaller birds. In addition, other contaminant data from the program period revealed that the lipophilic PCB congeners such as CB-153 was 18 times higher in sparrowhawk egg compared to fieldfare egg, see Table S5. From the concentrations of this hydrophobic persistent organic pollutant, a strong linkage of sparrowhawk as a predator of fieldfare and other birds was expected. The explanation to the opposite pattern of PFAS and PCB is likely connected to the different physicochemical properties with the hydrophilic (or protein associated) PFAS and the hydrophobic and lipid soluble PCBs, the diet of the various species and their differing metabolic capacities. Earthworm is a major diet of fieldfare, and earthworm from Oslo contained relatively high PFAS concentrations which is expected to be an explanation for the high PFAS levels in fieldfare egg.

One fieldfare egg sample from a ski track area (Holmenkollen) in 2018 contained the highest detected concentrations of PFDoDA (59.3 ng/g), PFTrDA (60.6 ng/g), PFTeDA (137 ng/g) and PFHxDA (26.7 ng/g) in fieldfare egg, and with a relatively low PFOS (18 ng/g) concentration. In the majority of the other observed samples, PFOS was the dominating PFAS, and highest at Grønmo.

Few available PFAS data are reported from urban areas for egg of the same bird species and the terrestrial mammals that were part of the Oslo study.

PFAS analysis of eggs of great tits (*Parus major)* near a fluorochemical plant in Flanders, Belgium, revealed that the PFAS concentrations were among the highest observed in birds (Groffen et al., 2017) with median concentration of PFOS of 10380 ng/g ww. Follow up studies revealed that despite the high concentrations, there was limited evidence of reproductive impairment (Groffen et al., 2019).

The area in northeast Michigan has some of the highest recorded PFAS exposure in birds in the United States, with geometric mean for total PFAS in tree swallow (*Tachycineta bicolor*) eggs ranging from 554 to 954 ng/g ww (Custer et al., 2019). There were no demonstrable effects of PFAS exposure observed on reproduction nor on most physiological responses (Custer et al., 2019).

Further, perfluorinated chemicals were investigated in a large study of European starling (*Sturnus vulgaris)* eggs across Canada; eggs collected in 2009–2012 and 2014, at locations such as landfills, industrial and urban environments (Gewurtz et al., 2018). In general, PFAS concentrations in eggs collected at landfill and industrial areas had highest concentrations. This is in agreement with what we find in the study from Oslo where the former landfill Grønmo had highest concentrations for fieldfare egg followed by the industry area of Alna. The median PFOS concentrations in starling eggs from year 2014 at landfills across Canada had large variations in data with median values of PFOS from 41 to 659 ng/g ww. This concentration range is comparable to the concentration range of PFOS in fieldfare eggs from the urban environment around Oslo.

In a very recent study, PFAS were investigated within a terrestrial avian food web of an urbanized region of Metro Vancouver, British Columbia, Canada (Fremlin et al., 2023). Samples were collected in 2016, and PFOS was the dominating compound in the avian food web where highest concentration was detected in the eggs of the apex predator Cooper's hawk (*Accipiter cooperii*). The mean value of PFOS based on 11 Cooper's hawk eggs was 138 ng/g ww. Of the PFCA compounds, PFTeDA (25 ng/g ww) and PFDoDA (19.8 ng/g ww) showed highest concentrations in Cooper's hawk which also dominated together with PFTrDA in the bird eggs from Oslo (see Table 1). PFAS were also reported in whole body of European Starlings (*Sturnus vulgaris*), American Robin (*Turdus migratorius*) and thrushes in the study of Fremlin et al. (2023), where the mean concentrations (ng/g ww) of PFOS in 6 samples of each species were 54.8, 52.6 and 34.8 of European Starling (n = 5), American Robin (n = 6) and trushes (n = 6), respectively. The mean PFOS concentrations from whole body European Starlings and American Robin were comparable to the mean PFOS concentration of sparrowhawk (53.5 ng/g ww) and fieldfare egg (56.3 ng/g ww) from our study, 2014–2020.

Tawny owl eggs revealed lower detection frequencies and concentrations than both fieldfare and sparrowhawk eggs from Oslo area. The concentrations were at average 4–5 times lower than in the other two bird species. The tawny owl eggs were only sampled approximately 40 km from Oslo city, and this might be an explanation for the lower PFAS concentrations compared to the two other bird species.

In a temporal trend study of PFAS in tawny owl eggs (1986–2019) from central Norway, the average and median concentrations of PFOS were 10.5 and 6.7 ng/g ww (Bustnes et al., 2022); which are comparable to the average and median PFOS concentrations in tawny owl eggs from Oslo area of 14.4 and 9.7 ng/g ww. The mean sumPFCA (2.9 ng/g ww) and sumPFAS (13.8 ng/g ww) concentrations in the temporal trend study were lower than those detected in our Oslo study of 9.8 and 25.5 ng/g ww, respectively.

In a Swedish study with ten eggs of tawny owl collected in 2014, the median total PFOS concentration was 7.9 ng/g ww (linear PFOS was 7.6 ng/g ww); Eriksson et al. (2016). In the same study, PFTrDA dominated the carboxylates with a median value of 1.4 ng/g ww, which is in agreement with the median value of PFTrDA in our study of tawny owl in 2020 with 1.3 ng/g ww. The Swedish study also included the species common kestrel (*Falco tinnunculus*) and osprey (*Pandion haliaetus*) where PFUnDA had highest concentrations among the carboxylates.

For comparison to herring gull egg from inner Oslofjord as part of monitoring in 2020 (Grung et al., 2021), PFOS was 19 ng/g ww followed by PFTrDA and PFDoDA of 1 ng/g ww.

Wu et al. (2020) investigated PFAS in 22 bald eagle (*Haliaeetus leucocephalus*) eggs collected between year 2000 and 2012 in Michigan, USA. PFOS was detected in all egg samples and dominated with a median concentration value of 106 ng/g ww (28.5–1338 ng/g ww). Among the carboxylates, PFUnDA dominated with a median of 10.3 ng/g ww (2.26–107 ng/g ww).

3.1.4. Terrestrial mammals

Of the mammals, liver samples from brown rat revealed higher PFOS and PFAS concentration than red fox liver samples, but with lower DF for some of the compounds. The average, median and highest PFOS concentration were twice the concentration of the red fox liver samples. Brown rat liver samples and tawny owl eggs had in general lower DF than the other biota samples. PFAS were measured in the livers of 40 red foxes from central Hesse, Germany, shot routinely or found dead in 2013 (Riebe et al., 2016). The median and average PFOS concentration was 28.6 and 46.6 ng/g ww, respectively, with a maximum concentration of 320 ng/g ww. The median and mean concentration of PFOS of the 66 red foxes from the area around Oslo city were 10.0 and 16.1 ng/g ww, respectively, with a maximum concentration of 144 ng/g ww, approximately half of the concentrations measured in German red foxes from 2013. The detection frequencies of the other PFAS in the German foxes were in general lower, and the detected concentrations for PFNA, PFDA PFDoDA were in agreement or slightly higher when compared to foxes from Oslo.

Herzke et al., reported recently comparable data for PFOS in wild mink and otter liver in Norway, with a mean of 135 and 130 ng/g ww respectively. In contrast, PFCA concentrations were up to 10 times higher in otter and mink (Herzke et al., 2023) than in the red fox liver samples from Oslo.

For PFAS in brown rat, one study from Japan investigated PFAS concentrations in 216 blood samples collected in the period 2004–2009 (Taniyasu et al., 2013). PFOS concentration ranged from <0.05 to 148 ng/mL (Taniyasu et al., 2013). PFOS accounted for 45 % of total PFAS concentration, whereas PFUnDA and PFNA, accounted for 20 and 10 % of total PFAS, respectively. To the best of our knowledge, this is the only study in addition to the Oslo study reporting PFAS occurrence data in free living brown rats from urban areas.

3.2. Other PFAS across species

6:2 FTS in earthworm had highest DF across the species with a maximum of 115 ng/g ww. The highest concentrations were at locations known to be popular for outdoor and skiing activities and at Alna. 8:2 FTS was found in the highest number of samples of fieldfare eggs, sparrowhawk eggs and earthworm, where the eggs had maximum concentrations of 55.1 and 47.6 ng/g ww. The highest concentrations of 8:2 FTS in fieldfare eggs were detected at the locations Alna and at Grønmo. Both 6:2 FTS and 8:2 FTS are known as precursors to PFOA and other PFCA compounds through oxidation processes (Al Amin et al., 2021; Houtz and Sedlak, 2012). FOSA was detected in many samples of the species with highest concentrations in red fox and rat liver of 3.3 and 5.5 ng/g ww. FOSA is known to be metabolised and form PFOS and PFHxS by microorganisms and plants in soil, and to some extent in earthworm (Zhang et al., 2021). One study of PFAS in North Sea top predators found that carnivora species had a much higher capacity of transforming FOSA to PFOS than cetacean species (Galatius et al., 2013).

3.3. Terrestrial species in an urban environment and their potential use as markers of PFAS contamination

Passerines such as fieldfares may roam over larger areas, but in our study, one location near a former landfill (Grønmo) was characterised by relatively high PFAS concentrations in soil and earthworms and revealed the highest PFOS concentrations (70–601 ng/g) in fieldfare egg over the years 2015–2020. During the years 2018–2020 the concentrations in fieldfare egg were still elevated and with low variability varying from 231 to 276 ng/g ww. This former landfill is nowadays used for recreational activities. Our findings support the potential of fieldfare eggs as bioindicators for urban PFAS contamination, despite spending the wintering further south. This is in line with the view that small passerines are so-called income breeders and form their eggs after spring arrival from nutrients acquired at the breeding grounds (Morganti et al., 2021). It is also shown that PFAS concentrations in snow bunting eggs (*Plectrophenax nivalis*) reflect local PFAS exposure after spring arrival despite short time (few days) from spring arrival to egg laying in the Arctic (Warner et al., 2019).

Samples of earthworms, fieldfare and sparrowhawk eggs were also collected at non-urban (background) stations (years 2013–2014), see Figs. S1–S5. The average and median sumPFAS values were approximately 4 times lower at the reference site compared to the Oslo area (Table 2, Fig. 3).

PFOS concentrations were significantly higher in urban areas compared to background in all species ($F_{1,\ 267} = 207$, $P < 0.0005$, Fig. 3). The difference, however, varied among species. It means that there was a statistically significant interaction between species and location ($F_{3,\ 267} = 4.3$, $P = 0.005$, Fig. 3). Urban and background differed the most in earthworm, fieldfare and red fox and the least in sparrowhawk (Fig. 3). SumPFAS concentrations were also higher in urban areas compared to background ($F_{1,\ 267} = 148$, $P < 0.0005$), and the difference varied among the species in the same way as for PFOS ($F_{3,\ 267} = 4.7$, $P = 0.003$). The lower difference between urban and background found for sparrowhawks may indicate that the urban sparrowhawks also forage in areas outside the city centre and rely on prey with a mix of urban and non-urban footprint in PFAS contamination. Fieldfare eggs and earthworm are suitable bioindicators as they integrate PFAS contamination within their biotope. They are easily accessible in their environment and breeding locations, and since they have a wide distribution, they also can be compared across locations on a large geographical scale.

3.4. Bioaccumulation and magnification

Soil and earthworm were sampled at the exact same locations from 2015 to 2020. BSAF values between earthworm and soil were well above 1 for many PFAS at various location in Oslo city area, see Table S3, revealing accumulation from soil to earthworm for many PFAS. For PFOS, the average BSAF values varied from 11 to 41 across 5 years. The PFCA compounds also revealed BSAF >1, see Table S3. In addition to the PFAS shown in Table S3, PFBS, PFPeS and PFHpS (only detected one year) also had BSAF well above 1. The location near the former airport Fornebu in 2017 had the highest BSAF values of PFDoDA, PFTrDA and PFTeDA of 148–330.

Since earthworm is the main diet of fieldfare, magnification ratios between fieldfare egg and earthworm collected the same year at the same locations, were calculated for detected concentrations. The results revealed a higher variation in ratio values than for the BSAF calculations, see Table S4, but it was clearly higher magnification from earthworm to egg of the LC- PFCAs, PFUnDA to PFTeDA at some locations.

3.5. Trophic levels and trophic magnification of PFAS in a terrestrial food chain

Fig. 4 shows the isotopic niche space illustrated with ellipses covering 95% of the $\delta^{15}N$ and $\delta^{13}C$ data (year 2015–2019) for each of the three species and demonstrates both separation and some overlap among the species. The mean $\delta^{15}N$ values for the three species were 4.7, 7.1 and 7.4 (‰) for earthworm, fieldfare egg and sparrowhawk egg, respectively. The means for fieldfare and sparrowhawk were significantly higher than that of earthworm ($p < 0.001$), while sparrowhawk was not statistically higher than fieldfare ($p = 0.54$). With respect to $\delta^{13}C$, all species means differed significantly ($p < 0.05$), with means of −26.9, −26.4 and − 25.4 (‰) for fieldfare, earthworm and sparrowhawk, respectively.

As an example, for strongly bioaccumulating compounds, significant higher PCB and PBDE lipid normalised concentrations were found in sparrowhawk compared to fieldfare, indicating sparrowhawk residing at a higher trophic level than fieldfare.

The overlap in $\delta^{15}N$ and isotopic niche space (Fig. 4) may indicate that the species do not follow a strict 3-species predator:prey relationship. Earthworm is known to be a major part of the diet for fieldfare, but it is more uncertain how large portion of the diet fieldfare is for sparrowhawk. The prey of sparrowhawk may also include other passerines. In addition, earthworm and fieldfare eggs were available for sampling in the area around the city centre of Oslo, while several sparrowhawk eggs were sampled further away from the city centre, which may indicate that the sparrowhawks may also feed on less urban prey.

Table 2
Comparison of PFOS (linear PFOS isomer) and sumPFAS concentrations (ng/g ww) for urban and background locations. Background locations in southern part of Norway and the urban Oslo area with data from 2013 to 2020. Average and median concentrations with minimum to maximum concentrations in parenthesis. n: number of samples (included LOD values). LOD values were substituted with 0.5*LOD. SumPFAS is the sum of FOSA, PFBS, PFPS, PFHxS, PFHpS, PFOS, PFNS, PFDS, PFUnDS, PFDoDS, PFTrDS, PFTeDS, PFHxA, PFHpA, PFOA, PFNA, PFDA, PFUnDA, PFDoDA, PFTrDA, PFTeDA, PFHxDA, 6:2 FTS and 8:2FTS.

	PFOS Background	PFOS Oslo	sumPFAS Background	sumPFAS Oslo
Earthworm (2013–2020)	0.77; 0.38 (0.05–3.31) n = 13	42.2; 8.53 (0.05–955) n = 55	5.31; 4.31 (0.42–12.6) n = 13	71.4 18.0 (1.59–1087) n = 55
Fieldfare (2015–2020)	1.87; 1.33 (0.72–5.67) n = 10	56.3; 23.3 (0.007–601) n = 57	9.06; 7.40 (4.51–17.3) n = 10	97.2; 58.6 (3.25–691) n = 57
Sparrowhawk (2014–2019)	6.65; 6.36 (2.09–13.6) n = 10	53.5; 31.8 (3.18–367) n = 50	15.6; 13.5 (8.29–34.0) n = 10	91.5; 63.7 (6.03–480) n = 50
Red fox (2014–2020)	0.41; 0.37 (0.002–1.33) n = 14	16.1; 10.0 (1.35–144) n = 66	0.94; 0.86 (0.26–2.38) n = 14	23.7; 16.3 (2.26–181) n = 66

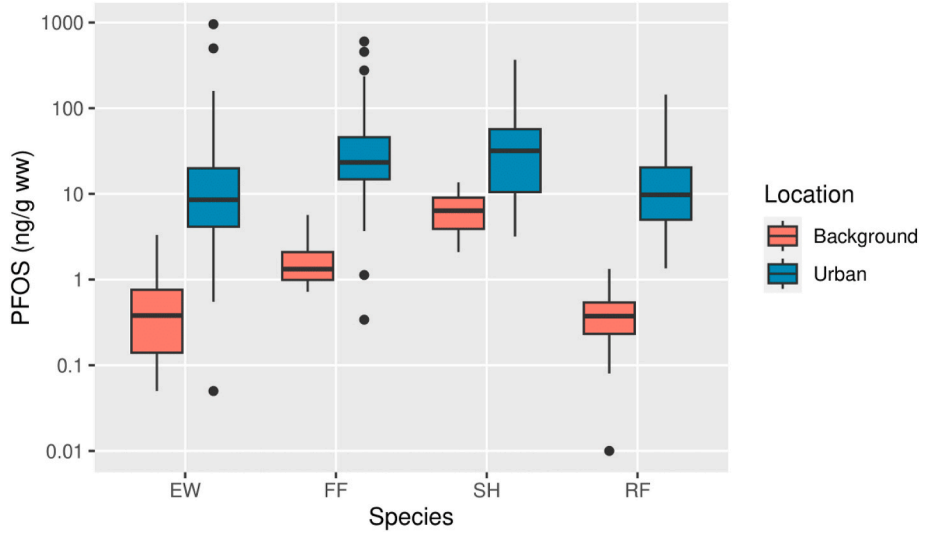

Fig. 3. Box plot of PFOS concentrations (ng/g ww) in species (earthworm, EW; fieldfare, FF; sparrowhawk, SH; red fox, RF) with both background and urban data.

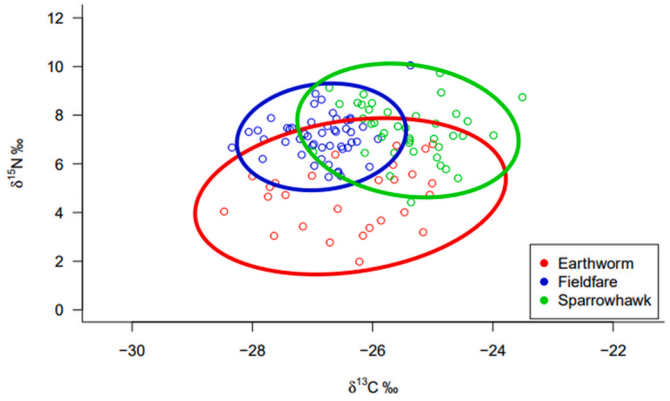

Fig. 4. $\delta^{15}N$ and $\delta^{13}C$ values from years 2015–2019 of earthworm (whole animal samples), n = 27, fieldfare (eggs), n = 49 and sparrowhawk (eggs), n = 40. Isotopic niche space is illustrated with ellipses covering 95% of the stable isotope data for each species, calculated with the Stable isotope Bayesian ellipses (SIBER) package in R (Jackson et al., 2011; R Core Team, 2020).

Although the difference in stable isotope $\delta^{15}N$ data (year 2015–2019) between sparrowhawk and fieldfare egg did not point to two different trophic levels, we have chosen to calculate an indicative TMF. The average PFAS concentrations and stable $\delta^{15}N$ values of sparrowhawk eggs during this period (2015–2019) were slightly higher than in fieldfare eggs. A relationship between the mean of $\delta^{15}N$ data and the mean of log concentration of PFOS for the years 2015–2019 where all three species were sampled show that the difference in $\delta^{15}N$ data and PFOS data between earthworm and fieldfare egg is significantly higher than between fieldfare and sparrowhawk egg, Fig. S8, and with large st. dev values of both parameters. Fig. S9 shows the same relationship when high concentrations from point sources (former and present industry) for earthworm (Alna, Grønmo and Fornebu) and fieldfare egg (Grønmo) were removed, which gave lower st.dev of the mean log PFOS concentration and slightly higher slope. Calculations without the point sources data confirmed a 40 % increase of TMF for PFOS.

Pollution point sources with very high contaminant concentrations may have large effect on the TMF results. However, we decided to include all data from 2015 to 2019 for the three species since the very high concentrations at the point sources were first and foremost for PFOS.

With the requirement of at least 60 % detection frequencies, food chain magnification factors could be obtained for PFHxS, PFOS, 8:2 FTS, PFOA, PFNA, PFDA, PFUnDA, PFDoDA, PFTrDA, PFTeDA, see Table 3.

Indicative TMF above 1 was found for 8 PFAS with increasing carbon chain length up to a chain length of 13 carbons; PFDoDA > PFTrDA > PFUnDA > PFDÃ PFTeDA>8:2 FTS > PFOS ~ PFNA. PFHxS and PFOA showed TMFs <1, indicating trophic dilution (Table 3). Log concentration of some of the compounds (PFOS, PFUnDA and PFDoDA) plotted against TL are given in Figs. S10–S12.

In the recent study of Fremlin et al., (2023) of a Canadian terrestrial food web consisting of invertebrates including earthworm, and several bird species, the calculated TMFs revealed that PFNA, PFDA, PFUnDA, PFDoDA, PFTrDA, PFTeDA, PFOS, and PFDS biomagnified in the food web; while PFOA, PFHxDA, and PFHxS did not appear to biomagnify; and PFBS biodiluted. These calculated TMFs were based on a chemical activity-based approach, which involved normalizing concentrations of PFAS in biota to their relative biochemical composition (Fremlin et al., 2023). Although our study from Oslo used concentration data on a wet weight basis, the results agree largely with the results from Fremlin et al. (2023), for which PFAS compounds have the potential to magnify or not.

Table 3
Calculated indicative TMF values of PFAS based on the 2015–2019 data for earthworm, fieldfare egg and sparrowhawk egg, along with Pearson correlation coefficient (R) and coefficient of determination (R^2). b is the slope estimate for the regression of log compound on trophic level. Significant P-values mean that b is different from zero. Note, positive values of b indicate magnification, while negative values indicate degradation.

Compound	TMF	R^2	R	P	Slope (b)	Significance
PFHxS	0.5	0.07	0.26	<0.005	−0.35	Significant
PFOS	1.7	0.05	0.22	0.01	0.22	Significant
8:2 FTS[a]	1.8	0.04	0.20	0.03	0.25	Significant
PFOA	0.7	0.02	0.15	0.06	−0.15	Nearly sign.
PFNA	1.6	0.06	0.25	<0.005	0.21	Significant
PFDA	2.0	0.11	0.33	<0.005	0.29	Significant
PFUnDA	2.3	0.13	0.36	<0.005	0.36	Significant
PFDoDA	2.5	0.15	0.38	<0.005	0.40	Significant
PFTrDA	2.3	0.13	0.37	<0.005	0.37	Significant
PFTeDA	1.9	0.09	0.30	<0.005	0.29	Significant
PFHxDA[b]	1.2	0.003	0.058	0.64	0.07	Not sign. different from 0

[a] only analysed in the years 2016–2019.
[b] only analysed in the years 2017–2019.

3.6. Threshold values for PFOS

The results were compared to known threshold values of PFOS for soil and predators to indicate the environmental harm potentially caused by the PFAS found in Oslo. Canada developed a dietary intake threshold for avian wildlife of 8.2 ng/g ww for PFOS, while the European Commission has set a 'specific quality standard for concentration in prey biota tissue protective of secondary poisoning in predators' (QSbiota) of 33 ng/g ww for PFOS (Ankley et al., 2021), which also was given tentatively in EU EQS PFOS dossier from 2011 (European Commission, 2011). In addition to these thresholds in feed of birds and predators, a predicted-no-effect-concentration (PNEC) value of 1000 ng/g ww of PFOS in eggs has been proposed (Ankley et al., 2021; Newsted et al., 2005; Van der Schyff et al., 2020) and a Canadian whole egg protective value of PFOS value of 1900 ng/g ww (Ankley et al., 2021).

None of the eggs exceeded the proposed PNEC of PFOS for bird eggs, but earthworm as prey items for fieldfare, and fieldfare as prey for sparrowhawk, showed average concentrations above the Canadian and European thresholds in food for avian wildlife and predators, respectively. For the earthworm samples, 18 % of the samples exceeded the European threshold of PFOS in diet for predators of 33 ng/g ww. For fieldfare (eggs) as diet for predators, 35 % of the samples were above the same threshold.

None of the soil samples exceeded a proposed PNEC for soil living organisms of 373 ng/g dw, which was based on experimental data for worm toxicity (Brooke et al., 2004).

4. Conclusion

The presented terrestrial study on an urban diffuse pollution scenario, is one of very few studies of its kind covering a high number of samples of various species collected over numerous years. Both invertebrates, birds and mammals were included in the study.

Our study confirmed that PFAS can be detected in all species and soil during all years. PFOS and the LC-PFCAs were the most dominating compounds in all samples, proving their ubiquitous distribution. Several point sources from PFAS emissions could be identified, connected to urban related activities as airports, professional skiing activities, recycling and waste disposal sites together with industrial activities. Furthermore, additional diffuse urban PFAS emissions cause general increased PFAS concentrations compared to less populated reference cites. Bioaccumulation (from soil to earthworm), magnification ratio (from earthworm to fieldfare egg) and an indicative TMF with a food chain approach revealed that several PFAS compounds have the potential to biomagnify.

None of the avian species exceeded the proposed predicted no-effect concentration (PNEC) for bird eggs of 1000 ng/g for PFOS, but earthworm as prey for fieldfare, and fieldfare eggs as proxy for whole birds as prey for sparrowhawk, had average concentrations above the Canadian and European thresholds in diet for avian wildlife and predators (Ankley et al., 2021).

PFOS concentrations were significantly higher in the urban area compared to background areas of earthworm, fieldfare egg, sparrowhawk egg and red fox liver. We find that fieldfare and earthworm are suitable bioindicators due to them habituating a large geographically area on a global scale, making them easily accessible and comparable between different locations, while at the same time able to integrate PFAS contamination within their biotope.

To the best of our knowledge, liver from urban rats were analysed for PFAS for the first time, evidencing a high potential for PFAS exposure and subsequently the high abundance of PFAS sources throughout the urban terrestrial environment. Efforts to identify, reduce and remediate urban PFAS emissions need to be intensified, to both protect the environmental and human health in urban settings.

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CRediT authorship contribution statement

Eldbjørg S. Heimstad: Writing – review & editing, Writing – original draft, Visualization, Validation, Software, Resources, Project administration, Methodology, Investigation, Funding acquisition, Conceptualization. **Torgeir Nygård:** Writing – original draft, Visualization, Validation, Software, Methodology, Investigation, Data curation, Conceptualization. **Børge Moe:** Writing – review & editing, Writing – original draft, Visualization, Validation, Software, Methodology, Investigation, Formal analysis, Data curation. **Dorte Herzke:** Writing – review & editing, Writing – original draft, Visualization, Validation, Software, Project administration, Methodology, Investigation, Funding acquisition, Formal analysis, Data curation, Conceptualization.

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.

Data availability

I have uploaded an excel file with the urban data

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.envpol.2024.123735.

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