CREW CENTRE OF EXPERTISE FOR WATERS

Per- and polyfluoroalkyl substances (PFAS) in drinking water supplies: A review of source, pathway, and fate for selected compounds

Carolin Vorstius, John Rowan, and Sarah Halliday

United Nations
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University of Dundee
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Glossary

1H-PFHx	1H-perfluorohexane
6:2 Cl-PFAES/F-53B	6:2 chlorinated polyfluorinated ether sulfonate
6:2 FTAB	6:2 fluorotelomer sulfonamide alkylbetaine
ADONA	Dodecafluoro-3H-4,8-dioxanonanoate
AFFF	Aqueous film forming foam
DUIN	Downstream User Import Notification
EA	Environment Agency
ECHA	The European chemicals Agency
EtFOSA/EtFOSAA/EtFOSE	N-ethyl derivatives of FOSA/FOSAA/FOSE
FASA	Perfluoroalkane sulfonamide
FASE	Perfluoroalkane sulfonamido ethanol
FBSA	Perfluorobutane sulfonamide
FHxSA	Perfluorohexan sulfonamide
FOSA	Perfluorooctane sulfonamide
FOSAA	Perfluorooctane sulfonamido acetic acid
FOSE	Perfluorooctane sulfonamido ethanol
FTAC	Fluorotelomer acrylates
FTCA	Fluorortelomer carboxylic acid
FTOH	Fluorotelomer alcohol
FTS	Fluorotelomer sulfonates
FTSA	Fluorotelomer sulfonic acid
HFC	Hydrofluorocarbon
HCFC-22	Difluoromonochloromethane
HFP	Hexafluoropropylene
HFPO-DA/GenX	Hexafluoropropylene oxide dimer acid
HFPO-TA	Hexafluoropropylene oxide trimer acid
HSE	Health and Safety Executive
LC-HRMS	Liquid chromatography-high resolution mass spectrometry
LoQ	Limit of quantification
LRAT	Long-range atmospheric transport
MeFOSA/MeFOSAA/MeFOSEE	N-methyl derivatives of FOSA/FOSAA/FOSE
PC	Principal Component
PCA	Principal Component Analysis
PFAA	Perfluoroalkyl acids
PFAS	Per- and polyfluoroalkyl substances
PFBA	Perfluorobutanoic acid
PFBE	Perfluorobutylethylene
PFBS	Perfluorobutane sulfonic acid
PFCA	Perfluoroalkane carboxylic acid
PFDA	Perfluorodecanoic acid (PFDA)
PFDoDA	Perfluorododecanoic acid
PFDoS	Perfluorododecane sulfonic acid
PFDS	Perfluorodecane sulfonate
PFECA	Perfluoroalkyl ether carboxylic acid
PFECHS	Perfluoroethylcyclohexane sulphonate
PFHpA	Perfluoroheptanoic acid

PFHpS	Perfluoroheptane sulfonate
PFHxA	Perfluorohexanoic acid_
PFHxS	Perfluorohexane sulfonic acid
PFIB	Perfluoroisobutene
PFNA	Perfluorononanoic acid
PFNS	Perfluorononane sulfonic acid
PFOA	Perfluorooctanoic acid
PFOS	Perfluorooctane sulfonate
PFOSA	Perfluorooctane sulfonamide
PFPA	Perfluoropentanoic acid
PFPS	Perfluoropentane sulfonic acid
PFPiA	Perfluoroalkyl phosphinic acids
PFSA	Perfluoroalkane sulfonic acid
PFTrDA	Perfluorotridecanoic acid
PFTriS	Perfluorotridecane sulfonic acid
PFUnDA	Perfluoroundecanoic acid
PFUnDS	Perfluoroundecane sulfonic acid
PPVE	Perfluoroethylvinyl ether
RWPP	Raw water presence potential
SSA	Sea spray aerosol
SML	Sea surface microlayer
TFE	Tetrafluoroethylene
TOP	Total Oxidizable Precursor
TrFE	Trifluoroethylene

Executive Summary

Purpose of research

This research is a follow-on from the project "Developing risk assessment approaches for PFAS and watch list parameters under the recast Drinking Water Directive – PFAS, 17ß-estradiol, nonylphenol" (Vorstius *et al.,* 2024) with a particular focus on per- and polyfluoroalkyl substances (PFAS).

Four knowledge gaps were addressed: i) assessing PFAS substances potentially occurring in the Scottish water environment from commercial import and use; ii) elucidating the relative role of atmospheric transport, and iii) the role of sea spray aerosols (SSA) as PFAS dispersal pathways, and iv) elucidating PFAS fate in different water body types.

Additionally, current knowledge on PFAS "fingerprints" – concentrations and compositions of PFAS substances characteristic to specific contamination sources – was summarised. An analysis of PFAS concentration data from 1.5 years of Scottish Water's national monitoring of raw water was also undertaken, exploring patterns of PFAS concentrations and compositions in relation to potential pollution sources.

Background

PFAS are a large group of synthetic chemicals used in a wide range of industrial and domestic applications. They have raised concern for the environment and human health, as they have been linked to cancer, impacts on child development, and immune system disorders. In Scotland, a drinking water standard of 0.1 µg/l for the sum of 20 individual PFAS compounds was introduced in January 2023. The previously conducted assessment of the relative likelihood of PFAS presence in raw water sources across Scotland (PFAS Raw Water Presence Potential (RWPP) assessment) helps inform monitoring for PFAS substances (Vorstius *et al.*, 2024).

Key findings

- The literature review provided evidence for including a risk factor based on SSA influence in coastal regions in the RWPP assessment, as well as improving pathway considerations by using considerations of groundwater vulnerability.
- The data analysis showed grouping potential for some PFAS substances and gave first indications of important explanatory risk factors.
- An update of the original PFAS RWPP assessment and individual RWPP assessments for PFAS subgroups were prepared, highlighting areas with comparatively high risk due to presence of possible PFAS sources.

Recommendations

1. Explore the feasibility of including further PFAS substances in the drinking water monitoring
2. Explore the feasibility of suspect and non-target screening to support our understanding of PFAS distribution in the Scottish freshwater environment
3. Investigate relationships of PFAS concentrations to other sampled pollutants or water quality indicators
4. Analyse for seasonal trends/patterns in the data
5. Investigate relationships of PFAS concentrations to raw water intake characteristics

1 Introduction

This report describes research commissioned by Scotland's Centre of Expertise for Waters (CREW) as a three month ‘calldown’ project that was undertaken as a follow-on to previous work carried out and described in “Developing risk assessment approaches for PFAS and watch list parameters under the recast Drinking Water Directive – PFAS, 17ß-estradiol, nonylphenol” (Vorstius *et al.,* 2024).

1.1. Background and scope

Per- and polyfluoroalkyl substances (PFAS) are a large group of synthetic chemicals produced since the 1940s with a wide range of industrial and domestic applications. Many PFAS are persistent in the environment and bioaccumulative (Scheringer *et al.,* 2022). This raises concern for the environment and human health, as they have been linked to cancer, impacts on child development, and immune system disorders (Chohan *et al.*, 2021). Over the past years, drinking water standards for PFAS substances have been introduced in many countries, including Scotland, where a standard of 0.1 µg/l for the sum of 20 named PFAS substances was set in January 2023 (The Public Water Supplies (Scotland) Amendment Regulations 2022).

To support risk assessment for PFAS substances of the Scottish public drinking water provider (Scottish Water), a research project commissioned by CREW was carried out that assessed the relative likelihood of PFAS presence in raw water sources (RWPP assessment) across Scotland (Vorstius *et al.,* 2024). The project identified several significant gaps in knowledge, including:

- sources of PFAS in the Scottish environment (e.g., which products contain PFAS, in which processes they are used, which specific PFAS compounds are used, and identifying actual vs potential sources);
- mechanisms of transport, especially the distance different compounds may travel under different conditions;
- biogeochemical behaviour of PFAS in different types of water body and in storage; and
- health implications due to compound and exposure level.

1.2. Project objectives

This study aims to address some of the knowledge gaps and improvements to the RWPP assessment suggested in Sections 3.5 and 4 of Vorstius *et al.* (2024) around the above described areas, focusing on issues identified as having the most potential to enhance the initial approach. Available evidence on four topics was examined to further enhance or modify the assessment. Additionally, although monitoring data is currently still limited, there is a growing body of PFAS monitoring data from Scotland, which has been used to draw preliminary conclusions on main contamination sources and pathway processes in Scotland. Areas addressed cover:

- Substances that may be in the Scottish water environment due to import and use, and their implications to human health
- The role of atmospheric transport and distances from emission to water source
- The role of sea spray aerosols in the overall risk from PFAS
- The fate of PFAS in the freshwater environment.

Additionally, current knowledge on PFAS “fingerprints” – concentrations and compositions of PFAS substances characteristic to specific contamination sources – is summarised to inform the analysis and interpretation of Scottish PFAS monitoring data. The data analysis then focuses on identifying patterns in PFAS concentrations and compositions in relation to potential pollution sources.

Improved insight into the above-described aspects helped to make improvements to the RWPP assessment, and to understand the frequency at which drinking water supplies need to be monitored, and whether additional PFAS substances not listed in the regulations should be considered. Due to PFAS evidence still developing, many of the identified knowledge gaps in Vorstius *et al.* (2024) cannot be fully answered at this time, but will require additional evidence from specifically designed field studies. The RWPP assessment will therefore continue to be based on the best available information and needs to stay adjustable to take account of emerging evidence. The insights gained from updating and interpreting the RWPP can then also support Scottish Water’s risk assessment process.

1.3. Structure of the report

Section 2 gives a brief overview over the methods applied in this research.

Section 3 summarises the outcomes of the literature review, for the four areas of interest: PFAS substances of interest for monitoring due to their potential presence in the UK (3.1), the role of atmospheric transport to PFAS pollution (3.2), the role of sea spray aerosols (3.3), transport to and within the freshwater environment (3.4), and approaches to fingerprinting and source tracking of PFAS in the environment (3.5).

Section 4 describes the Scottish Water raw water monitoring data for PFAS and presents the results of the statistical analysis.

Section 5 describes the update of the RWPP assessment, explaining methods used (5.1), presenting the updated assessment (5.2), and discusses its implications (5.3).

Based on the previous sections, Section 6 gives recommendations for monitoring and further research.

2 Research undertaken

The initial research project (Vorstius *et al.,* 2024) assessed potential presence of 20 substances of PFAS for drinking water supply sources in Scotland, using a source – pathway – receptor framework. The resulting RWPP assessment systematically identified areas where these substances pose a potential risk, in order to guide monitoring and sampling strategies. It also identified knowledge gaps and further research needs.

In the current project, some of these gaps were addressed and further researched through examining new literature and connecting to experts in the field. Literature was found through academic search engines, identified from relevant papers, and from recommendations from colleagues.

An increased set of monitoring data from Scottish Water's regulatory sampling, spanning 12 January 2023 to 8 July 2024, has also been examined to better understand patterns of PFAS concentrations for Scotland. The data, the approach to, and methods for data analysis are described in Section 4.1.

The insights gained from the literature review and the data analysis were used to further inform and refine the initial RWPP assessment, as per the concept outlined in Section 3.5 of Vorstius *et al.* (2024) (Figure 2.1). The approach to updating the assessment and methods is further described in Section 5.1.

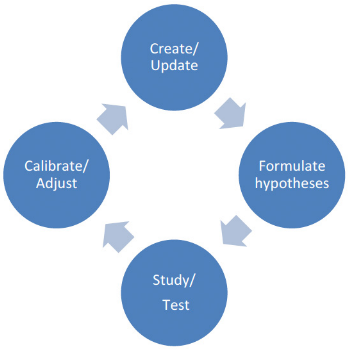

Figure 2.1: Cycle of continuous improvement of risk assessment, taken from Vorstius *et al.* (2024).

3 Summary of available evidence

This section describes the evidence on the four topics of interest: imports and uses of PFAS substances in the UK (3.1), mechanisms and distances of atmospheric PFAS transport (3.2), the role of SSA (3.3), and transport to and within the freshwater environment (3.4). It also includes literature on fingerprinting PFAS (3.5). Each section summarises the findings with a focus on main PFAS substances of interest, mechanisms and distances of travel, and concludes with describing implications of the findings for monitoring and/or the update of the RWPP assessment.

3.1. Import and use of PFAS in the UK

PFAS can enter the UK through several routes. They can be manufactured, imported in substances for use in the manufacturing of other products, or they can be contained in articles and products entering the UK market.

3.1.1 Sources of information

There is only one site in the UK (in Thorton-Cleveleys, Lancashire) where fluoropolymers are currently produced, emitting some fluorinated substances and PFAS[1] (Dalmijn *et al.,* 2024). The site is permitted to emit 5.35 tonnes per year of HFC-125 and hexafluoropropylene to the air. The Environment Agency estimates that around 250 tonnes of PFOA were released from this site between 1950 and 2012, with emissions to both surface water (the River Wyre) and to the atmosphere, and some landfilled (Dalmijn *et al.,* 2024).

Under UK REACH legislation, if a company based in Great Britain (England, Scotland and Wales) wishes to import or manufacture a substance at more than one tonne per year, a registration to Health and Safety Executive (HSE) is required. This registration includes information about the properties and uses of the substance, and an assessment of hazards and potential risks. Registration is not required for mixtures (formulations), but the individual substances comprising the mixture must be registered if the aggregated volumes reach more than one tonne per year. The UK REACH register thus holds information on PFAS substances that are manufactured or imported at volumes of more than one tonne per manufacturer/importer per year. This means that this register lacks information on PFAS that are imported at less than one tonne per year (per importer).

Chemical substances that are already regulated by other sector-specific legislations such as agricultural pesticides, medicines or radioactive substances are partially or completely exempted from REACH requirements. There are also only limited registration requirements for substances contained in other products (substances not supplied per se). For these substances, registration is only required if the substance is on the candidate list (list for substances of very high concern (SVHC)), and the substance is present in quantities above 1 tonne per producer/importer per year, and the substance is present in those articles above a concentration of 0.1% weight by weight. PFAS included on the current SVHC list are HFPO-DA/GenX, PFHxS, PFDA, PFNA, and PFOA.

The transitioning period from EU REACH to UK REACH is still in place. The obligation to register under UK REACH includes the import of substances from the EU, but existing EU REACH registration holders based in Great Britain can defer the full registration under UK REACH if they submit notifications with initial information. DUINs (downstream user import notifications) should have been submitted by 27th October 2021 by those considered downstream users or distributor in the two years prior to January 1st, 2021. Full registrations then need to be made by 27th October 2026, 2028, or 2030, depending on tonnage and hazardous properties (HSE, n.d.-a). There is also a possibility to "grandfather" an existing EU REACH registration, which requires the submission of initial information by 30th of April 2021, and full registration by the same deadlines as for DUINs (HSE, n.d.-b).

The UK REACH register is currently not openly available. It provides an overview of which PFAS substances are imported at higher volumes to be used in the manufacturing of other products. However, this does not give a complete picture of PFAS in the UK as low tonnages are excluded, and many PFAS substances contained within articles will not be captured. Alternatively, there is the database for substances registered under EU REACH, which is

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[1] Dalmijn *et al.* (2024) list these substances as tetrafluoroethylene (TFE), trifluoroethylene (TrFE), hexafluoropropylene (HFP), difluoromonochloromethane (HCFC-22), hydrofluorocarbons (HFC-23, HFC-125), Perfluoroisobutene (PFIB), 1H-perfluorohexane (1H-PFHx), perfluorobutylethylene (PFBE), and perfluoroethylvinyl ether (PPVE).

openly available (https://chem.echa.europa.eu/). It includes substances meeting the EU REACH registration obligations, manufactured and/or imported into the EU, with information on their hazardous properties, classification and labelling, and how to use them safely. It is likely to provide a similar level of information, and limitations. The European Chemicals Agency (ECHA) database could be searched under the assumption that registrations were made when the UK was still part of the EU and that registered substances were imported to the UK. The Environment Agency (EA) conducted such a search in 2019, and found 114 relevant substances (EA, 2021). HSE (2023b) also conducted a search of the UK REACH database during an HSE Analysis of the most appropriate regulatory management options (RMOA) (HSE, 2023b), identifying 36 PFAS substances from 'grandfathering' and at least 40 additional substances from DUINs in the UK. An additional 182 substances were identified from a search of the ECHA database. These substances, their uses and level of concern can be found in the Annexes to the RMOA (HSE, 2023a).

Glüge *et al.* (2020) used a variety of sources to obtain information about PFAS and their uses. They searched patents, looked at companies that manufacture and sell PFAS, and market reports.

Patents can be used to determine in which applications PFAS may be used and include information on manufacturing process or material composition. They may therefore give more specific information when it comes to characteristics of the substances produced/used, however, it is not necessarily possible to discern if the patent is actually in use, to what extent, and where, limiting its usefulness to a broader global assessment. Globally, patents can be found e.g., via Google Patents. More specifically for the UK, the UK Patents Journal (https://www.ipo.gov.uk/p-pj) contains UK patent applications that are filed, published, granted, ceased or expired, and European patents that apply to the UK. The UK Patents Design Journal (https://www.registered-design.service.gov.uk/view) contains design applications that are newly registered, notices, renewals and changes to the register. Espacenet (https://worldwide.espacenet.com/?locale=en_EP) contains details of worldwide UK patents, owners, filed documents and countries where a patent applies.

Market reports might be especially useful for understanding future trends in PFAS uses and which substances might increase in use and hence, probably in the environment. Reports used by Glüge *et al.* (2020) were the Fluorotelomer Market Report, Fluorochemicals Market Report, and the Perfluoropolyether Market Report from Global Market Insights.

Finally, material specification sheets might give information about the composition of the products and the materials contained. While Glüge *et al.* (2020) attempted to search the factsheets of major PFAS manufacturer, they concluded that the effort necessary was beyond the scope of their study. This highlights the major difficulty that there is a general lack of (easily accessible) information on PFAS contained in materials and products. Intermediate users of products may themselves be unaware of PFAS contained in them (through intentional use or contamination). While the information could be highly useful in understanding if specific applications, such as wind turbine blades, could present a source of PFAS to the environment, it is unfeasible to collect this information UK-wide.

3.1.2 Candidate PFAS substances for monitoring

The current Scottish drinking water PFAS standard comprises 20 compounds from the group of perfluoroalkane carboxylic acids (PFCAs) and perfluoroalkane sulfonic acids (PFSAs), so-called "arrowheads" that can form from more complex PFAS compounds and typically don't degrade any further[2]. These PFCAs and PFSAs are also (or have historically been) directly produced for industrial or commercial application. However, many more PFAS substances are found in the environment, including precursors and intermediate but relatively stable PFAS, and alternative PFAS substances that have been developed to replace substances that have been banned due to environmental and health concerns. Many of these alternative PFAS have shown higher mobility in the environment, leading to increasing detections in the environment, including freshwater sources and drinking water, and concerns are growing over potential impacts on health.

Substances that have been identified in the analyses of the UK and EU REACH databases (HSE, 2023b, EA, 2021) as intermediate or degradation products of imported/used PFAS, and that have potential health implications, would be candidate substances for additional monitoring (some are already included in the monitoring programme of English drinking water providers). The following section summarises groups of some of these substances that have also

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[2] The arrowhead approach is a commonly used approach to grouping PFAS for risk assessment and management, based on an "arrowhead substance of concern" (usually a PFAA) representing a group of PFAS including its salts and precursors (Cousins *et al.,* 2020).

been widely detected globally, but we refer to HSE (2023a) for the complete list of identified PFAS and degradation products.

3.1.2.1 Fluorotelomer alcohols (FTOHs)

Fluorotelomer alcohols (**FTOHs**) are volatile precursors for PFCAs. They have the generic formula $F(CF_2)_nCH_2CH_2OH$ and are commonly named after the number of fluorinated carbons and hydrocarbons, e.g., 8:2 FTOH is a molecule with 8 fluorinated carbons and a 2 carbon ethyl alcohol group (Figure 3.1). FTOHs are replacement products for PFOS and have many applications. While they have been predominantly studied in air, they have been detected in rainwater, wastewater effluent, and sludge (Chen *et al.,* 2017).

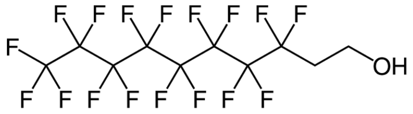

Figure 3.1: 8:2 FTOH. Taken from Wikipedia (2023)

3.1.2.2 Fluoroalkylethers (ether-PFAS)

Fluoroalkylethers are fluorinated alternative chemicals that are starting to be increasingly recorded in the environment (Munoz *et al.,* 2019).

Hexafluoropropylene oxide dimer acid (**HFPO-DA**, Figure 3.2), often called by the trade name of its ammonium salt GenX, hexafluoropropylene oxide trimer acid (**HFPO-TA**), and dodecafluoro-3H-4,8-dioxanonanoate (**ADONA**) are alternatives for PFOA in fluoropolymer production. They have increasingly come into focus over their increasing concentrations in the environment, including drinking water, and potential health implications, raising questions whether they constitute suitable alternatives to PFOA from an environmental point of view (Yang *et al.*, 2022, *Sheng et al.,* 2018).

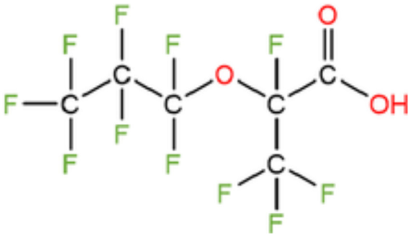

Figure 3.2: HFPO-DA. Taken from Zhao *et al.* (2022b)

6:2 chlorinated polyfluorinated ether sulfonate (**6:2 Cl-PFAES**), or called by its trade name F-53B, is an alternative to PFOS in the electroplating industry, and seems to be predominantly used in China, thus being distributed to other parts of the world through products. It has been shown to be among the most prevalent alternative PFAS in freshwater (Hamid *et al.*, 2024).

3.1.2.3 Fluorotelomer sulfonates/sulfonic acid (FTSAs)

Fluorotelomer sulfonic acids (**n:2 FTSAs**, n=4, 6, 8, 10, Figure 3.3) are intermediate degradation products of alternatives to PFOS in aqueous film forming foams (AFFF). They have been shown to degrade to 5:3 fluorotelomer carboxylic acid (**5:3 FTCA**), PFPA, and PFHxA (Zhang *et al.,* 2016). FTSAs have been found in sediment, soils, sludge and leachate.

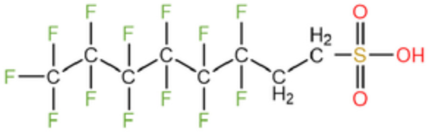

Figure 3.3: 6:2 FTSA. Taken from Zhao *et al.* (2022b)

3.1.2.4 6:2 fluorotelomer sulfonamide alkylbetaine (6:2 FTAB)

6:2 FTAB (Figure 3.4) is an alternative PFAS used as a surfactant and in AFFF. It has been detected in groundwater, surface water, soils, and sediments. It has been shown to convert to 6:2 FTSA and shorter chain PFCAs in plants (Zhao *et al.,* 2022a).

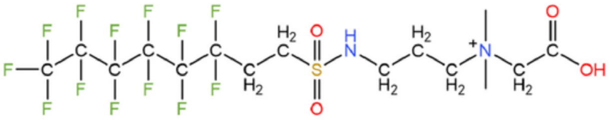

Figure 3.4: 6:2 FTAB. Taken from Zhao *et al.* (2022b)

3.1.2.5 Perfluoroalkane sulfonamides (FASAs)

Perfluoroalkane sulfonamides (FASAs), while precursors for perfluoroalkyl acids (PFAAs), are stable intermediate compounds that can persist in the environment. They have been found in high concentrations next to contaminated sites (Pickard *et al.,* 2024). FASAs such as perfluorobutane sulfonamide (**FBSA**, C4), perfluorohexan sulfona mide (**FHxSA**, C6), and perfluorooctane sulfonamide (**FOSA**, C8), have been identified in drinking water in some countries including Canada, the EU, and China (Kaboré *et al.,* 2018).

3.1.2.6 Perfluoroalkyl phosphinic acids (PFPiAs) and perfluoroalkyl phosphonic acids (PFPAs)

Other alternatives to long-chain PFAS, **PFPiAs** and **PFPA** (Figure 3.5) have been used in industrial and commercial products since the 1970s and are increasingly detected in the environment. Concerns are raised over their high bioaccumulation potential (Zhang *et al.,* 2023).

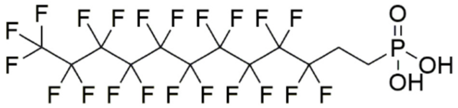

Figure 3.5: (1H,1H,2H,2H-Perfluorododecyl) phosphonic acid. Taken from Quiñones *et al.* (2017)

3.1.3 Suspect and non-target screening

The phase-out of targeted manufacturing and use of the more conventional PFAS such as longer chain PFAAs, has led to shifts towards alternative fluorinated compounds. As a consequence, increasing proportions of organofluorine components in various environmental media remain unidentified in targeted PFAS analysis, suggesting that many PFAS are present in the environment beyond the commonly analysed “arrowheads”, including their precursor substances (Zweigle *et al.*, 2023, Yeung and Mabury, 2016, Aro *et al.,* 2021).

To address this issue, Total Oxidizable Precursor (TOP) assay has been used to estimate the contribution of unidentified PFAA precursors to total PFAS in samples. During the TOP assay, PFAA precursors are chemically converted to mainly PFCAs, which are then conventionally analysed (Göckener *et al.,* 2023, Macorps *et al.,* 2023b).

Additionally, analytical methods with liquid chromatography-high resolution mass spectrometry (LC-HRMS) allow suspect and non-targeted screening that can help to identify PFAS in the environment (Faust, 2023, Krauss *et al.,* 2010, Manz *et al.,* 2023). In suspect screening, some compound-specific information such as molecular formula and structure is necessary. Lists compiling individual PFAS and their characteristics are becoming publicly available[3], supporting suspect screening methods (Joerss and Menger, 2023). In non-target screening, no a-priori information on the PFAS compound is available, and a time-consuming step involves prioritisation, i.e. the separation of the analytes of interest from the detected components. Characteristic mass differences and Kendrick mass defect analysis are for example used to separate PFAS from other hydrocarbon features in HRMS data. Techniques are being developed further to facilitate prioritisation, and PFAS identification from LC-HRMS (Zweigle *et al.,* 2024, Joerss and Menger, 2023, Bugsel *et al.,* 2023).

3.1.4 Implications for monitoring and data collection

Sampling for a selected number of PFAS compounds has the potential to miss a significant part of PFAS in the sample (Göckener *et al.*, 2023, Yeung and Mabury, 2016). The inclusion of some of the main alternative PFAS and PFAA precursors in the targeted analysis is advisable. While suspect and non-target screening at the current stage is probably unfeasible for larger scale application in drinking water monitoring, they could eventually become tools to better understand which PFAS substances are present, and thus support areas of potential high risk and with further research needs.

There are significant gaps in information about PFAS imported to the UK, especially those imported at low tonnages and contained within products. This not only challenges risk assessment, but it also hampers effective management and regulation, and reduces consumers’ ability to make informed choices. Tools to increase disclosure and facilitate PFAS identification along the supply chain could fill these gaps, but a discussion of these is beyond the scope of this work.

3.2 Atmospheric emission and transport

PFAS enter the atmosphere from emissions from manufacturing sites, landfills, wastewater treatment plants, vehicle exhausts, firefighting, and indoor products. PFAS have also been detected in SSA around the globe, which will be discussed in Section 3.3. The initial data collected by the Scottish public drinking water supplier, Scottish Water, led to speculation that there may be an influence from wind power installations in the drinking water catchments. This section therefore reviews both available evidence of atmospheric transport and deposition, with a special focus on distances of travel; and evidence on influence of wind turbines on PFAS concentrations surrounding wind power installation.

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[3] Examples for PFAS suspect lists: https://data.nist.gov/od/id/mds2-2387; https://zenodo.org/records/63488607

3.2.1 Mechanisms of transport and deposition

PFAS are transported in both the gas and the particle phase (Faust, 2023). Especially volatile fluorotelomer compounds FTOHs, polyfluoroalkylated sulfonamidoalcohols (FOSE), or FOSA are readily transported via the atmosphere, but also fluorotelomer acrylates (FTACs), other FASAs, perfluoroalkane sulfonic acids (PFSAs), and perfluoroalkane sulfonamino ethanols (FASEs). Emerging PFAS such as HFPO-DA (GenX) are also being detected in the atmosphere (Lin *et al*., 2020).

The lifetime of aerosol particles in the atmosphere is days to weeks. While longer-chain PFAS such as PFOA and PFOS have been phased out, they are still frequently detected (Gewurtz *et al.,* 2019, Saini *et al.,* 2023). It is hypothesised that the degradation of precursors (e.g., FTOHs, FOSE, FOSA) in the atmosphere can lead to elevated concentrations of arrowhead products (of which PFOA and PFOS are the most commonly studied) even if direct local emission sources are not or have ceased to be present (Lin *et al.,* 2020).

Thackray *et al.* (2020) modelled transport and deposition of PFCAs globally, showing the importance of rainfall and wet deposition for these substances. For PFOA, they estimate dry deposition to make up 35%, and variations in overall depositions rates depending on variations in PFOA formation and rates of precipitation. They show that shorter-chain deposition is highest near sources and where NOx can shorten the fluorinated chain length, leading to degradation of precursor substances into arrowheads. Shorter chain PFCAs are also more efficiently removed by wet deposition. Longer-chain PFCAs are transported further, so with increasing distance from emission sources, the ratio of short chain to long chain PFCAs decreases. An exception to this may be PFBA, as in a study of PFAS concentrations in the Arctic area, Wong *et al.* (2018) found that while most analysed PFAS compounds showed concentrations comparable to rural areas and lower than urban areas, PFBA concentrations were in a range comparable to urban and remote air. This suggests PFBA is uniformly distributed in the global atmosphere, either due to PFBA being more susceptible to long range atmospheric transport (LRAT), or due to atmospheric transformation of volatile hydrofluorocarbons and hydrofluoroethers into PFBA. Wang *et al.* (2022) also found PFBA in areas remote from emission sources, indicating high LRAT potential.

3.2.2 Travel distances

Galloway *et al.* (2020) took surface water samples around a manufacturing facility in Ohio, originally producing PFOA and later HFPO-DA. They found contamination of surface waters downwind of the facility that they were able to attribute to atmospheric deposition, and conclude the impact zone to be at least 41 km for surface water and 48 km for soils, with more than 10 ng/l up to 30 km from the plant. Schroeder *et al.* (2021) found that soils more than 200 km downwind of PFOA industrial emissions have been contaminated, as well as groundwater in 6–8 km distance from the factory.

Studies taking samples from different media (air, soil, surface water, groundwater, biota) generally see a decreasing trend of PFAS contamination with increasing distance (Galloway *et al.,* 2020, Brandsma *et al.,* 2019), but there is no conclusive evidence for setting a distance/radius of influence, as PFAS are often still detected in the furthest sampling point of the studies, and/or concentrations don't necessarily follow a predictable decreasing pattern. This is probably due to complex pathways which may involve different media, varying environmental conditions over space and time, different or variable emission rates, and differences in behaviour of different PFAS compounds. Schroeder *et al.* (2021) hypothesise that the pH of the soil influences the probability of leaching of PFAS to groundwater, with higher pH decreasing soil sorption (see Section 3.4).

Shimizu *et al.* (2021) examined several perfluoroalkyl ether carboxylic acid (PFECAs), PFAS substances developed as alternatives to PFOS and PFOA. They measured PFOS, PFOA and four PFECAs in wet and dry deposition at a site in North Carolina approximately 110 km downstream of a manufacturing site. They found that wet deposition is about a magnitude higher than dry deposition, and suggest that PFAS originate from the manufacturing facility as well as from sea spray aerosols (especially PFOS).

Analyses of rainwater in North America show that short chain PFCAS are most frequent, with PFBA and PFHxA dominating compounds. Of the PFSAs, PFOS is most commonly detected, followed by PFHxS (Pfotenhauer *et al.,* 2022). While PFOA and PFOS concentrations are higher around urbanised sites than rural sites, the distribution of shorter chain PFAS was more even (Gewurtz *et al.,* 2019).

3.2.3 Wind turbines and PFAS emissions

The rotor blades of wind turbines are manufactured from fibre-reinforced polymer composites and need to withstand harsh environmental conditions. They are subject to leading edge erosion, caused by rain, sand and flying solid particles, so blades are commonly coated to protect them against and reduce erosion (Dashtkar *et al.,* 2019). Common coatings are organic or metallic coatings containing a range of potential pollutants, such as metals or phenols. PFAS are commonly used in chrome plating, making it a possibility that PFAS substances are present in wind turbine blades.

We have not been able to find any published material that examines the emissions of PFAS substances from wind turbine blades. While evidence of leaching/emission of pollutants is generally sparse, especially under environmental conditions, the discussion is focused on microplastics (Çelik Gül and Gül, 2024), metals, or bisphenol A (commonly found in epoxy resins) (Kirchgeorg *et al.,* 2018), and on offshore windfarms. There is currently no indication that wind farms, at least in the offshore environment, lead to significant pollution of the surrounding area (Ebeling *et al.,* 2023, Wong *et al.,* 2018).

3.2.4 Conclusion for the RWPP assessment update

Evidence of atmospheric transport of PFAS from emission sources predominantly stem from studies carried out around PFAS manufacturing, and do not present a clear picture. Distances travelled and concentrations found vary depending on emission amounts, emitted substance, and environmental conditions. The initial assessment used a radius of 5 km around the emission source, based on a study by Chen *et al.* (2018). We recommend keeping this radius to represent the area of highest impact. For a national risk screening, this is a compromise between capturing likely impact and making the assessment too indiscriminate. Including longer distances only seems to be indicated if additional factors are taken into consideration such as predominant wind direction, soil properties and vegetation around the emission source, and the hydrology of potentially impacted freshwaters. This is more feasible for a local assessment rather than a national screening.

Wind farms, energy and telecommunication infrastructure were not included in the emission sources when deriving area of impact from atmospheric deposition. Based on the very limited available evidence, we recommend not changing this at this time. For better evaluation, field evidence is needed especially from soil and freshwater samples at varying distances from these potential emission sources to assess the likelihood and magnitude of PFAS contamination.

3.3 Sea spray aerosols (SSA)

From the initial RWPP assessment and the then monitoring data available (Vorstius *et al.,* 2024), the hypothesis was formulated that there may be an influence of SSA that carry PFAS and thus transport them to coastal regions of Scotland. The available evidence is reviewed to understand the mechanisms of PFAS transport on SSA and to conclude on an appropriate representation of this risk factor in the RWPP assessment.

3.3.1 Marine concentrations and emissions

SSA are formed from the sea surface microlayer (SML). SML is a layer of between 1 µm and 1 mm at the surface of the ocean that is enriched with organic matter. The hydrophobic nature of the SML enhance the surface enrichment of hydrophobic pollutants (García-Flor *et al.,* 2005). When air is entrained into seawater by the wind or breaking waves, SSA are formed from the SML via bubble bursting (Casas *et al.,* 2020). Surface active substances like PFAS can be scavenged by the air-water interface of the bubbles and thus transferred to the atmosphere via SSA (Sha *et al.,* 2021).

Studies on PFAAs in the ocean have shown concentrations to sharply decrease with increasing depth (Sha *et al.,* 2024). PFOS and PFOA have been found in significantly higher concentrations in surface water and especially the SML, compared to subsurface water, with enrichment factors of 24-109 in some near shore sampling sites in the Chinese Sea (Ju *et al.,* 2008). Work by Casas *et al.* (2020) from the Southern Ocean showed PFAS enrichment of the SML to surface water by factors of 1.2 to 5 with highest values for PFBS, PFOS and PFOA; and PFAS enrichment factors for SSA compared to SML ranged between 522 and 4690, with highest values for long-chain PFAS such as PFNA and PFDA, although highest concentrations in SSA were reported for PFBA.

A Belgian scoping study in Flanders measured PFAS concentrations in sea foam on two locations and found elevated concentrations in foam compared to sea water (between 36 and 51 µg/l for one site, sampled in May 2022, and between 1400 and 2400 µg/l for the other site, sampled in

October 2022). PFOS and PFOA (both linear and branched isomers) were predominant substances, but PFDA, PFNA, PFHxS, PFHpS, PFPS, 6:2FTS, 8:2FTS, MeFBSAA, FHxSA, FOSA, FOSAA, MeFOSAA, EtFOSAA, HFDO-PA, and PFECHS were also found (VITO, 2023). The results from the Flemish site sampled in May 2022 seem to be confirmed by a Dutch study of PFAS in 2022, measuring sea foam at 14 different locations on the Dutch coast and finding comparable concentrations (Bokkers *et al.,* 2023).

Sha *et al.* (2024) estimate that the emissions of PFOS from the oceans via SSA exceed the estimated contribution of other sources of PFOS to the atmosphere such as direct emissions and degradation of precursors, and that PFOA emissions via SSA are comparable to other atmospheric sources. They also estimate that between 15-30% of PFAA emitted via SSA is deposited to land, with the majority to coastal regions.

3.3.2 Travel distances

Analysing the results from long-term air monitoring at two Arctic sites, Sha *et al.* (2021) found that C7-C12 PFCAs were frequently detected, with significantly higher concentrations of PFNA, PFDA, and sum of PFCAs; and a higher frequency of detection for PFHxA, at the site closer to the coast (1.3 km to open water) than the site further from the coast (20 km to open water). PFOS and PFHxS were frequently detected at both sites. They found significant correlations between PFAAs and Na+ at both sites, and conclude that SSA can be an important source of atmospheric PFAS in coastal areas and beyond, as SSA can travel considerable distances (>300 km). In another study of PFAS concentrations in Artic areas, Wong *et al.* (2018) found that PFOA was found in higher concentrations at two sites closer to open water (100 m and 2 km) than a site further from the coast (~4 km) and where the water is covered by sea ice most of the year. The study authors speculated that there is additional PFOA input from SSA at the sites closer to the ocean. PFOS concentrations were comparable for all sites.

In Denmark, a study was carried out on Fanø island on the North Sea coast with no known PFAS sources, but where PFAS was found in the groundwater supply (predominantly PFOA and to a lesser extent PFHxS, and at some sites also PFBS and PFOS). Taking samples from soils, groundwater and surface water with increasing distance from the sea, the study found a pattern of exponential decrease of PFAS concentrations in groundwater and surface water, with the impact flattening off from about 2 km inland (Figure 3.6 and Figure 3.7). Predominant substances in the soils were PFOS, and to a smaller extent PFOA. They also found a decrease in PFOS/PFOA ratio with increasing distance from coast in groundwater samples (NIRAS, 2023).

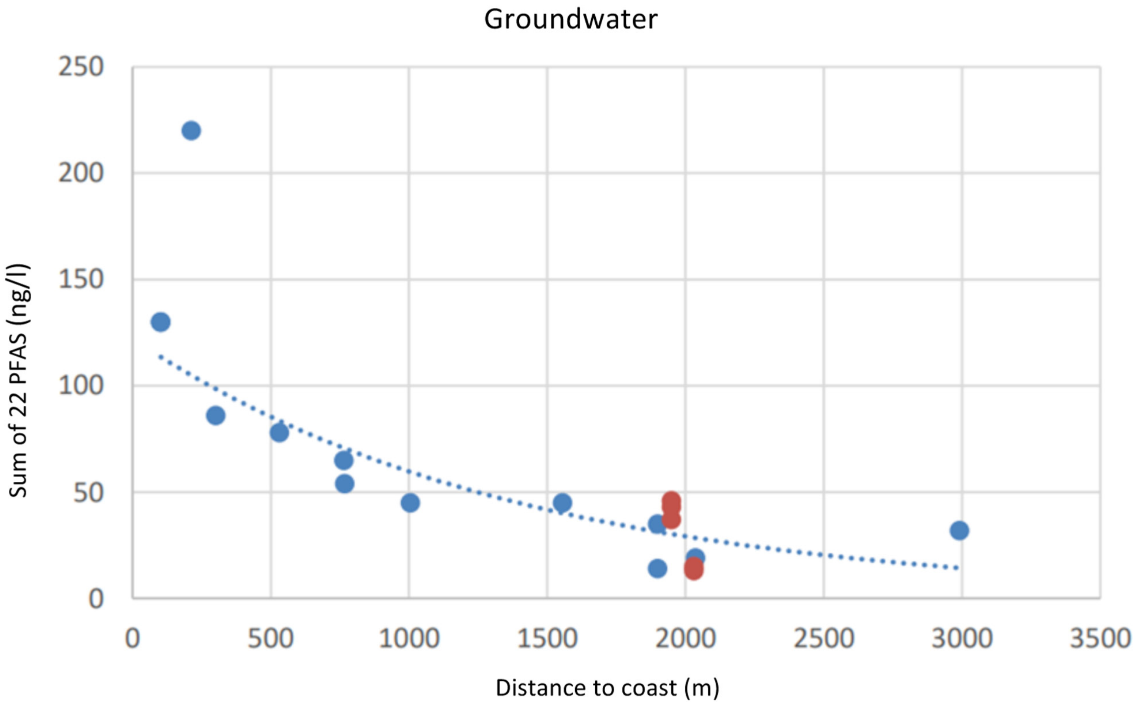


Figure 3.6: Concentration of the sum of 22 PFAS (4-13 carbon PFCAs and PFSAs, 6:2 FTS, and PFOSA) in groundwater samples measured at sites with varying distances from the coast in Fanø island in Denmark. Red and blue dots represent different sampling rounds. Adjusted from NIRAS (2023).

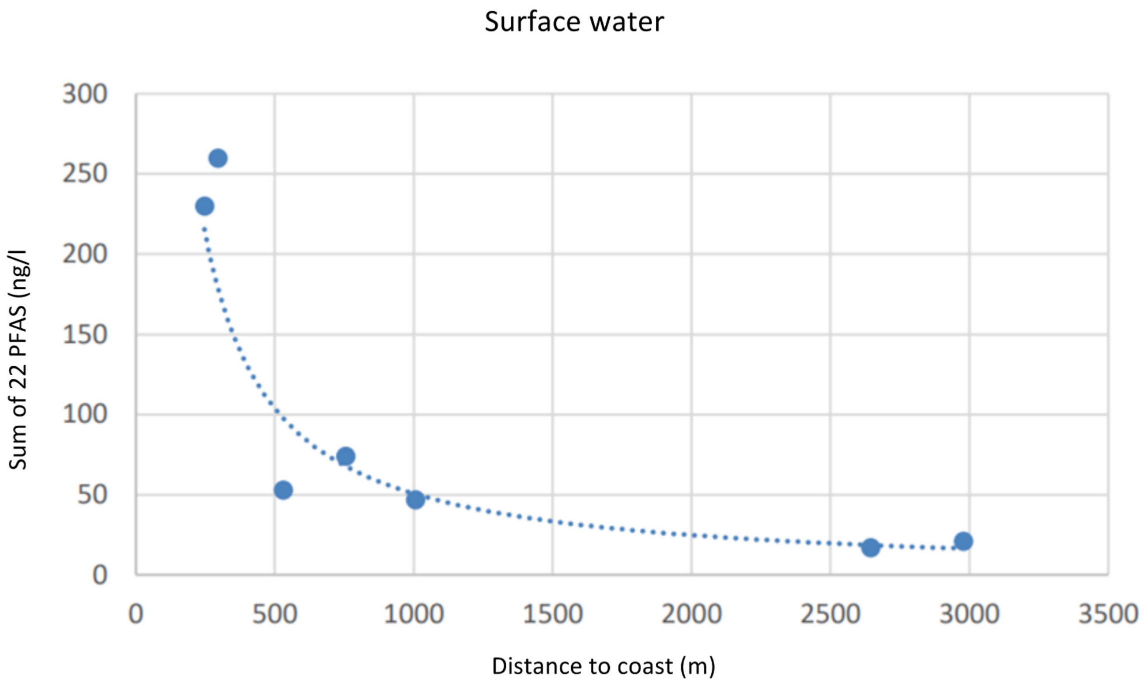


Figure 3.7: Concentration of the sum of 22 PFAS (4-13 carbon PFCAs and PFSAs, 6:2 FTS, and PFOSA) in surface water samples measured at sites with varying distances from the coast in Fanø island in Denmark.

3.3.3 Conclusion for the RWPP assessment update

There is compelling evidence that SSA are a source of PFAS especially in coastal regions. While the probable distance of the impact is difficult to determine and will likely depend on local conditions (coast characteristics, winds, PFAS concentrations and compositions in the marine environment), evidence from Europe and Arctic areas (NIRAS, 2023, Sha *et al.,* 2021, Wong *et al.,* 2018) suggest most PFAS on SSA will deposit within a distance of 2 km from the coast. We recommend including a 2 km zone from coast to inland in the assessment representing risk from SSA deposition. This is in keeping with the evidence and the approach taken for representing other risk factors.

3.4 Transport to and within the freshwater environment

The initial RWPP assessment (Vorstius *et al.*, 2024) identified some significant gaps in understanding of PFAS travel to and within freshwater, which presented challenges for the assessment. There was no distinction made in the application of pathway representation between groundwater supply sources and surface water sources. Additional evidence is reviewed to get a better understanding of possible approaches to separate between different kinds of supply sources and thus allow a more differentiated assessment.

3.4.1 Sorption behaviour

PFAS mobility in aquatic systems is influenced by partitioning to soils and sediment. This depends on chain length (higher adsorption with increasing chain length), head group (PFSAs partition more readily to soils and sediment then PFCAs), and differences in water and soil/sediment chemistry such as cation concentrations and organic carbon content. Soils and sediments can act as a sink for some PFAS compounds, especially PFSAs and linear isomers, and concentrations of PFAAs are found to be higher in sediments than in the overlying water (Balgooyen and Remucal, 2022, Mussabek *et al.,* 2020).

While PFAS characteristics are probably the most important factor in sorption potential (Milinovic *et al.,* 2015), the characteristics of the soils and sediments also play a role (Table 3.1). Variability in sorption is often attributed to organic carbon content (Balgooyen and Remucal, 2022, Tran *et al.,* 2022), but sorption cannot necessarily be predicted by organic carbon content alone (Campos Pereira *et al.,* 2018, Li *et al.,* 2018, Mussabek *et al.,* 2020). Campos Pereira *et al.* (2018) show that decreasing pH of the soil increases sorption especially for PFCAs, with an increasing strength of the relationship for increasing chain length. Increasing cation (Al^{3+}, Ca^{2+}, Na^{+}) concentrations increased sorption of medium chain length PFAS (C5-C8 PFCAs, PFHxS), but had little effect for longer chain

Table 3.1: Overview of soil characteristics influence on PFAS adsorption to soil/sediment.

Soil characteristic (increasing)	PFAS adsorption	Comment
Organic carbon content	+	Especially for shorter and medium chain PFAS
Soil pH	-	Especially for PFCAs and an increasing effect for longer chain PFAS
Cation concentration	+	For medium chain PFAS only
Clay content	+	

PFAS, or PFBS and PFBA. Campos Pereira *et al.* (2018) also found that soil organic matter was a better predictor for sorption for the shorter chain PFAS than pH, which was a better predictor for the longer chain PFAS. They hypothesise that long chain PFAS bind predominantly to the humin fraction of the soil organic matter, whereas shorter chain PFAS bind to the humic and fulvic acids. In contrast, Mussabek *et al.* (2020) found that organic matter had no significant impact on PFAS distribution in lake sediment, but sediment inorganic content had. However, Li *et al.* (2018) point out that one variable alone is a weak predictor for sorption, and multiple regression models, including at least organic content, pH and clay content, can more successfully explain sorption behaviour.

There are contradictory findings as to whether sorption to the sediment is unlikely to be reversible, or if sediments can act as a secondary source when aqueous concentrations decline (Balgooyen and Remucal, 2022). Tokranov *et al.* (2021) examined variability in PFAS concentrations between a surface water body in Massachusetts, USA, and the region where recharge of the surface water to the downgradient groundwater occurs. They show that the exchange between groundwater and surface water bodies can disperse PFAS contamination to greater distances from the source, and observe a decline of precursor substances over distance. They also observe small presence of precursors in sediments. The concentrations in porewater and groundwater varied significantly seasonally, with higher concentrations in winter. Contrary to laboratory and other field studies, they found that sorption of PFCAs is higher than PFSAs and unrelated to chain length. They hypothesise that this is due to biologically mediated sorption, and suggest that PFAAs are reversibly sequestered in summer and released in winter.

3.4.2 Distances and timescales of travel

Studying groundwater, surface water, soil and sediment samples in an AFFF impacted area in Sweden, Sörengård *et al.* (2022) showed that the concentration of PFAS substances was highest in groundwater and contamination migrated by over 10 km, explained by high hydraulic conductivity and low PFAS sorption in a coarse fluvial esker. Adamson *et al.* (2022) studied PFAS fate and transport processes at three different AFFF impacted military bases in the US. They found that PFAS groundwater contamination rapidly declined with distance (280-530 m) from contamination source at all sites, by two to three orders of magnitude, indicating non-destructive attenuation.

Gerardu *et al.* (2023) sampled soils in the vicinity (8-16 km) of a manufacturing plant in the Netherlands that has been shown to have impacted soils within a 50 km radius. They included three sampling sites on inland dunes composed of aeolian sand, and an adjacent peat site. PFOA concentrations were highest at several tens of centimetres below the surface, indicating that PFOA is mobile in these soils and that the highest atmospheric deposition has passed, whereas PFOS showed the highest content in the shallowest sample. In peat, both PFOA and PFOS content was highest in the shallowest sample, indicating less mobility. Modelling transport up to 2070, they estimate that PFOA will mostly (80-90%) migrate to groundwater in the sand soils, while PFOS will be more strongly retained (>50%).

3.4.3 Conclusion for the RWPP assessment update

Similar to the evidence from atmospheric deposition and LRAT, movement of PFAS in the soil and aquatic environment is dependent on a mixture of factors difficult to represent at national scale.

In terms of conditions that facilitate retention in soils and sediments, the assessment used soil pH and aquifer productivity as measures of retention and likelihood to reach groundwater. The groundwater vulnerability for Scotland dataset prepared by the British Geological Society (BGS, https://www.data.gov.uk/dataset/afa1a8f6-0e68-4b29-8245-8d8323eee2ec/groundwater-vulnerability-scotland-version-2) combines many characteristics that potentially play a role in mitigating movement of PFAS to groundwater sources in Scotland, such as maximum permeability of bedrock and superficial deposits, areas of thick clay, or depth to groundwater. We recommend trialling this dataset for representing pathways.

In terms of including distance of contamination source to raw water intake, while there is evidence that PFAS are retained and concentrations, as well as substances, change with distance, it is difficult to determine a clear relationship as local conditions will heavily influence movement and retention of PFAS. The effort to derive the distances and include a distance decay in the assessment seems not proportional to the benefit to the national scale assessment, but could be considered for a catchment scale assessment.

The assessment made no separation by groundwater and surface water sources. Based on the literature, contamination of an area usually leads to both surface and groundwater being affected. A general separation of the assessment therefore still seems unwarranted, but different weightings for different types of sources might be possible when considering different PFAS compounds (see Section 4.4).

3.5 Fingerprints

There is increasing interest in developing environmental forensic tools that enable source tracking for PFAS found in the environment. Due to the highly stable nature of many PFAS, this has been focused on looking at the identity, detection frequency, concentrations of and ratios between different PFAS compounds through a lens of site characteristics to define PFAS “fingerprints” for known sources.

3.5.1 Approach and methods

Approaches to identify PFAS fingerprints for different source types have included sample analysis combined with multivariate statistics and/or machine learning (Joseph *et al.,* 2023). Some “identifiers” that indicate PFAS sources can be concentration ratios, isomer ratios, or derived through e.g., principal component analysis (PCA) or clustering.

Concentration ratios between two different PFAS compounds is a relatively simple screening technique. Examples include PFHxS to PFOA, or PFOS to PFOA ratios as indicators of AFFF contamination vs. influence of manufacturing sources (Guelfo and Adamson, 2018), PFOA to PFNA ratio as indicator of atmospheric degradation of 8:2 FTOH (Ahrens *et al.,* 2023), or PFHpA to PFOA ratio to identify atmospheric deposition (Simcik and Dorweiler, 2005).

Isomers are compounds that consist of the same number of carbon atoms, fluorine atoms, and other atoms making up the PFAS substance, but that differ in their structure. The relative abundance of different isomers can be used to infer a source, or the manufacturing origin; e.g., the presence of branched isomers typically indicates production by electrochemical fluorination, whereas pre-dominantly linear isomers are produced by fluorotelomerization (Charbonnet *et al.,* 2021). Different isomers of a PFAS could also show different characteristics and behaviour, e.g., regarding mobility in the environment, bioaccumulation, or toxicity. Differences in behaviour could change the percentage of isomers further away from the contamination source.

Statistical methods like PCA or cluster analysis aim to identify underlying patterns in data that include many variables (e.g., concentrations of multiple PFAS compounds). PCA identifies composites of variables, which can support identification of similarities between sites. Because PCA leads to new, constructed variables, these can then be used in combination with correlation analysis, and be associated with contamination sources. Clustering uses other approaches to group sites together based on the similarity of data, with different techniques available. These techniques have been applied to PFAS data analysis (Nguyen *et al.,* 2017, Joseph *et al.,* 2023, Zhu *et al.*, 2022, Balgooyen and Remucal, 2022).

A limit to our current development and understanding of “fingerprints” for different sources is the variability in PFAS substances that are being investigated in different studies – e.g., a substance identified as having a high frequency and concentrations in one study might not be included in another, especially precursor substances and intermediate transformation products. Most studies are also based on one sampling event, meaning that temporal variations (e.g., trends, variability due to release-study time lapse, or seasonal differences) are significantly understudied.

Despite the limitations, some promising results have been achieved (Joseph *et al.,* 2023). Nevertheless, it needs to be remembered that there is a wide range of “profiles” of PFAS compounds even within the same source types, due to a multitude of factors e.g., use of different PFAS containing products, history of release, and transport processes. Furthermore, many commonly studied PFAS, such as PFCAs and PFSAs, are common to many sources.

It is therefore impossible to determine definitive, globally applicable ratios or values, but general tendencies can be observed and can prove useful in determining sample analysis, further investigation, and potential mitigation measures.

3.5.2 Indicative fingerprints

3.5.2.1 Ratios

In an analysis of US drinking water sampling data (UCRM3), including data on PFCA and PFSA concentrations, Guelfo and Adamson (2018) found that PFOA was found in elevated concentrations relative to other PFAS compounds at sites impacted by historical fluoropolymer manufacturing, as well as those impacted by waste water treatment works (WWTWs). In contrast, while PFOA is detected at AFFF influenced sites, it is not a major component of the foams and therefore the ratios of PFOS to PFOA and PFHxS to PFOA tend to be much higher at those sites, and higher ratios could be associated with AFFF impact scenarios in the data analysed.

Simcik and Dorweiler (2005) investigate the usefulness of the PFOA/PFHpA ratio to determine the influence of LRAT versus local contamination sources. FTOHs can be transported large distances by the atmosphere and then predominantly degrade into PFCAs (PFHpA, PFOA, PFNA). There is no known direct application of PFHpA, but PFOA has widely been applied in manufacturing, leading the authors to hypothesise that PFOA will dominate surface waters closer to local contamination sources such as manufacturing sites and WWTPs, but decrease in proportion further away from direct inputs. Data they collected from different locations in the midwestern United States seemed to confirm their hypothesis, with ratios <1 for sites impacted by local sources and ratios of >6 for more remote areas. This ratio has been used to determine dominant influence of atmospheric transport over local contamination in several academic studies since (Zhu *et al.*, 2022, Wang *et al.,* 2024, Xu *et al.,* 2024).

3.5.2.2 Predominant substances and substance compositions

Many PFAS substances, especially PFCAs and PFSAs, are common to many sources, making source differentiation based on a handful of commonly studied PFAS difficult. Table 3.2 presents a summary of the PFAS substances most commonly associated with specific types of contamination sources, collected from literature.

3.5.2.3 Compositional studies

Zhu *et al.* (2022) performed hierarchical cluster analysis and PCA on soil samples taken from the Shanghai area, testing for 18 PFCAs and PFSAs. Samples were taken from soils impacted by industry, fire stations, airports, landfills, and agriculture. Using these PFCA and PFSA compounds alone, they could distinguish industrial sites by the presence of long chain PFAS compounds, and a fire station by elevated levels of PFOS. Zhu *et al.* (2022) also found mainly positive correlations between PFAS compounds at most sites, but the agricultural sites showed more negative than positive correlations. Biological factors may affect PFAS compositions due to biodegradation or plant uptake.

Joseph *et al.* (2023), using PCA and cluster analysis, clearly distinguished three groups of sources by PFAS composition: WWTPs, biosolid and landfill leachate, and AFFF impacted groundwater, with a trend in concentrations. WWTPs showed lower relative concentrations, moderate concentrations were found in biosolid and landfill leachate, and high concentrations were found in AFFF-impacted groundwater.

Guelfo and Adamson (2018) performed a hierarchical cluster analysis on UCMR3 data, finding that PFOA and PFHpA cluster together as well as PFOS and PFHxS, while PFBS and PFNA did not cluster with either of these groups. They hypothesise that PFNA is not associated with other PFAS in the primary known use (manufacturing of polyvinylidene fluoride), partly explaining this dissimilarity, while separate clustering of PFBS might be the result of several factors, including manufacturing away from long-chain PFAS to shorter chain PFAS like PFBS. They also discuss the weaker sorption characteristics of PFBS, potentially leading to higher concentrations in surface waters. They also find a clear shift towards higher percentages of PFSAs compared to PFCAs in groundwater relative to surface water.

Paige *et al.* (2024) took samples from surface water and sediment from 65 sites around the area of Melbourne, analysing 33 PFAS compounds and comparing the sites according to dominant land use. Apart from one of the sites, which had a known AFFF contamination incident, there were no identified major point sources in the area, and Australia has no manufacturing plants for PFAS. From the comparison of surface water mean concentration values per group (rural, residential, industrial and wastewater influenced), rural areas showed the lowest concentrations (Table 3.3). While throughout the whole dataset, PFOS had the highest

Table 3.2: Dominating PFAS compounds from different sources.

Source of contamination	Dominating (detected at high frequencies and at high concentrations) or diagnostic (often associated with a specific source) PFAS substances	Reference and scope	Comments
Airports	PFOS, PFAA ≤ 8 Cs (PFPA, PFHxA, PFBA, PFHpA, PFOA), PFBS, PFPS, 6:2 FTS	Anderson et al. (2023): The study provides a review of information and data on PFAS contamination around airports with a focus on the USA.	The study author's observations show that PFCAs are the dominant group in PFAS contamination associated with airports. PFAS other than PFCAs and PFSAs constituted 17% which is a higher percentage than at other sites. The ratio of PFHxS to PFOS and PFOA were high and distinguishable from other sources.
AFFF impacted sites	PFHxA, FHxSA, PFPS, PFHxS, FBSA	Joseph et al. (2023): The study conducted target and suspect screening on samples taken from six different PFAS source types, including landfill leachate, wastewater effluent, biosolid leachate, and AFFF-impacted groundwater throughout the US.	(Joseph *et al.,* 2023) found consistent PFAS patterns for AFFF impacted sites, suggesting that source attribution to AFFF can be done with a few diagnostic PFAS.
	PFOS, PFHxS, 6:2 FTSA, with changes further downstream through degradation of 6:2 FTSA to PFHxA and PFPA	Ahrens et al. (2023): This study looks at PFAS in Svalbard, trying to distinguish local sources and LRAT by taking samples from different environmental media at different locations in the region.	PFAS composition in soil and water changed with increasing distance, with 6:2 FTSA the major compound close to a firefighting training site but not detected in soils a few hundred metres away, suggesting that 6:2 FTSA was degraded and/or retained in the soil. PFHxA and PFPA were amongst the predominant compounds in drainage water downhill of the site.
Chrome plating	PFOS, PFHxA, 6:2 FTS	Anderson *et al.* (2023)	PFAS profile similar to airports but with a higher percentage of PFSAs.
Industrial sites	PFOA, PFHxA, PFOS, PFHxS, PFHpA, PFBS, PFBA, PFPA, PFDS.	Anderson *et al.* (2023)	While PFAAs with chain lengths of 8 or less were dominant, there are more longer chain compounds present than compared to other sites. PFCAs show clear dominance over PFSAs and 6:2 FTS is less frequently detected.
Industrial areas	FTSAs, PFOS precursors (N-MeFOSAA, N-EtFOSAA)	Macorps *et al.* (2023a): The study examined PFAS in surface sediment at a nation-wide scale in France.	Precursor substances were dominated by PFOS precursors and FTSAs at industrial areas in Macorps *et al*. (2023b).

Table 3.2: Dominating PFAS compounds from different sources.

Source of contamination	Dominating (detected at high frequencies and at high concentrations) or diagnostic (often associated with a specific source) PFAS substances	Reference and scope	Comments
Wastewater treatment works	PFHxA, PFPA 3:3 FTCA, 5:3 FTCA	Anderson *et al*. (2023)	Anderson *et al*. (2023) found that WWTP and landfill profiles are similar to each other. Lower detection frequencies and concentrations than other point sources, with main contributions from PFCAs with 6 or less carbons, less representation of PFSAs (hence a much lower ratio of PFHxS to PFOA). FTSs are relatively lacking, probably due to further transformation, but transformation intermediates such as 3:3 FTCA and 5:3 FTCA were detected at higher frequencies.
	PFBA, PFOA, PFNA, Fipronil (organofluorine-containing insecticide)	Joseph *et al.* (2023)	Landfill leachate and wastewater could be better distinguished in Joseph *et al*. (2023), with fipronil being identified as diagnostic.
Landfills	6:2 FTCA, 5:3 FTCA	Joseph *et al.* (2023) Charbonnet *et al.* (2021): This review discusses techniques for environmental source tracking of PFAS.	5:3 FTCA is primarily associated with landfill leachate.
	PFOA, PFBA, PFPA	Xu *et al.* (2021): This study analysed PFAS in different environmental media around a Chinese landfill.	Landfills may contribute to PFBA and PFPA in nearby surface waters.
Biosolid	PFOA, PFOS, 3:3 FTCA, 5:5 FTCA, 6:2 FTCA, 7:3 FTCA, EtFOSAA, MeFOSAA	Joseph *et al.* (2023)	Joseph *et al*. (2023) found similar profiles of biosolid and landfill leachate, but identified PFAS compositions that could distinguish those sources (not described).
Paper and pulp industry/ renewable energy industry	PFOA, PFCAs (4-6 carbons), PFOS, 6:2 FTS	Joseph *et al.* (2023)	Wastewater effluent from paper and pulp and renewable energy generation showed similar PFAS profiles, with PFAS being generally detected more frequently and at higher concentrations for paper and pulp effluent.
Meltwater	PFBA	Ahrens *et al.* (2023)	Ahrens *et al*. (2023) found a dominance of PFBA in meltwater despite longer chain PFAS (PFNA, PFOA, PFDA, PFUnDA) being dominant in surface snow, which is in agreement with previous studies.
		Casal *et al.* (2017): This study examined the relevance of atmospheric snow deposition as a source of PFASs to Maritime Antarctica, taking samples from plankton, seawater, surface snow, and snow melt	Comparisons between fresh snow, surface snow and snowmelt concentrations of PFBA by Casal *et al*. (2017) suggest early removal of the shortest-chain PFCAs from melting surface snow.

Table 3.3: Mean concentrations in ng/l for PFAS substances from surface water sites in the area of Melbourne, split by sites receiveing wastewater (WWTP, n=4), sites dominated by rural (n=8), residential (n=37) or industrial (n=15) land uses. Reproduced from Paige *et al.* (2024).

Congener	Rural	Residential	Industrial	WWTP
PFBA	5.7	14.4	16.5	8.2
PFPeA	0.8	10.1	14.8	6.8
PFHxA	0.9	12.4	15.6	8.5
PFHpA	0.2	4.7	5.9	2.6
PFOA	0.6	13	9.5	6.8
PFNA	0	1.5	1.7	0.6
PFDA	0	3.6	1.8	0.8
PFUdA	0	0.2	0.1	0
PFDoA	0	0.3	0	0
PFTeDA	0	0	0	0
PFBS	0.2	3.5	2.9	1.2
PFPeS	0	0.2	0.8	0.2
PFHxS	0.9	3.2	8.8	3.8
PFHpS	0	0	0.3	0
PFOS	1.2	8.8	37.2	5.4
PFOSA	0	0.1	0.1	0
N-MeFOOSA	0	0.2	0.2	0
N-EtFOSAA	0	0.1	0	0
6:2 FTS	0	0	13.4	0
8:2 FTS	0	0	0.1	0
Σ33 PFAS	10.4	76.2	129.8	44.8

mean concentration (14.3 ng/l). PFBA showed the highest median concentration (11.3 ng/l), similar to the mean concentration, showing an even distribution throughout the study area. Looking at ratios between PFCAs and PFSAs, they found that industrial areas showed relatively more PFSAs (5:4) than the other areas (4:1), with PFOS and PFHxS making major contributions to PFAS in industrial areas. In rural areas, PFBA made up more than half of the PFAS, but was found in concentrations still below the sites in the other groups.

3.5.3 Implications for monitoring and data collection

The challenges associated with defining “fingerprints” for specific types of PFAS contamination sources are apparent by the many often subtly different, but sometimes even contradictory findings between different studies. Differences in concentrations and compositions of PFAS compounds associated with the same type of source can have various reasons. Among those are the sampled media (air, soils, water, biota, etc.), plus differences in sampling and analysis, distance to contamination source, or pathways (Charbonnet *et al.,* 2021). In freshwaters, PFAS contamination may be a mix of multiple diffuse and/or point sources, blurring the distinction.

Due to many of the sampled PFAS, such as PFSAs and PFCAs, being common to many sites, fingerprinting based on these compounds may be of limited success (e.g., Zhu *et al.* (2022)). Including more compounds such as FTSA, FTCAs, FOSAs can help to distinguish more clearly between sources, and Joseph *et al.* (2023) suggest to include even more, rarely tested PFAS compounds. While the analysis of a large group of compounds over a large number of sites may be infeasible, the identification of “sentinel” substances that are indicative of specific sources (Charbonnet *et al.,* 2021) may enable targeted sampling and analysis at sites where source tracking is important for mitigation purposes.

4 Raw water monitoring data analysis

Scottish Water commenced regular PFAS sampling in January 2023. This includes sampling for the 20 PFAS substances included in the drinking water standard (0.1 µg/l for the sum of PFAS) for raw and treated water. As the focus of this research lies on identifying potential presence of PFAS in raw water, the data available from the raw water sampling was examined and analysed.

Building on the findings of "fingerprints" - typical substances, concentrations, and ratios between PFAS substances depending on pollution source – from the literature (Section 3.5), the Scottish Water raw water monitoring data can be explored to see if comparable patterns are observable, and if these patterns can be interpreted to better understand sources of contamination, pathways, and consequent concentrations in drinking water supply sources.

4.1 Data and methods

The water quality dataset comprises Scottish Water raw water quality monitoring data for 20 PFAS compounds for the sampling period 12/1/2023 to 8/7/2024. Samples were taken from 316 points at drinking water treatment works, sampling either the raw water (which may be blended or pre-treated) or source water that can be allocated to a specific supply catchment. Sites were sampled at varying frequencies, with 1-17 samples per site included in the dataset. Data describing "risk factors" within the supply catchments were compiled during the previous project (Vorstius *et al.*, 2024).

The water quality data were summarised to give an indication of which substances are found in what kind of concentrations throughout Scotland, and mapped to understand any spatial patterns. To support further analysis, below detection limits were set to 0. While it is acknowledged that a sample with a result below limit of quantification (<LoQ) may contain the PFAS substance in very small concentration, it was chosen to use 0 as a representation for <LoQ over other commonly used approaches (such as LoQ value, or 50% of LoQ) to avoid potentially misleading comparisons (as LoQs differ for individual PFAS).

Correlations between PFAS substances were derived from the complete dataset. Mean concentrations as well as percentages detections per substance per catchment were derived for further analysis. Boxplots were created to visualise differences in mean concentrations between catchments of different water source types, and Wilcoxon tests undertaken to test for statistically significant means. Principle Component Analysis (PCA) was performed that included individual samples where at least one PFAS substance was detected. Additionally, PCA was performed on mean catchment concentrations per substance, and on percentages of detections per substance, both with catchment risk factors included as supplementary variables (these included area in km^2, density (number per km^2) of airports, electrical infrastructure, fire stations, industrial estates, landfills, metal recyclers, ore mines, renewable energy, ski areas, telecommunication infrastructure, water discharges, septic tanks, and road length, percentage of arable area, area of potential biosolid application, area within 2 km of the coast, and area of atmospheric deposition, and mean slope and long-term mean annual precipitation).

4.2 Results

4.2.1 Substances and concentrations found

The sampled substances comprise PFAAs of varying chain lengths (4-13 carbon atoms). In the current dataset, all of these substances have been detected, although some are much more common than others (Table 4.1, Figure A.18A). Generally, the shorter chain PFCAs (PFBA, PFPA, PFHpA) are most commonly found (in 48.5%, 23.1%, 22.8% of samples, respectively), as well as PFOA (29.5%), PFOS (8.5%) and PFBS (8.5%). PFBA has detection rates of 100 % at 93 sites, and the highest maximum concentration in a single sample was for PFBA. PFOA is also consistently found at 64 sites, while PFOS is consistently found at only 10 sites (at 8 of which PFOA also has a detection rate of 100%). PFHxS has only been found at 7 sampling sites, but is found at one site at relatively high concentrations (>0.01 µg/l). Other PFSAs, and longer chain PFCAs, are found much less frequently and at fewer sites.

Table 4.1: Summary of Scottish Water PFAS sampling data. No. of samples per substance = 1512, no. of sites = 316. Drinking water standard = 0.1 µg/l for the sum of all 20 PFAS substances.

Substance	No. of detections (%)	No. of sites with detections (%)	Maximum concentration (µg/l)	Limit of quantification (LoQ) (µg/l)
Perfluorobutanoic acid (PFBA)	734 (48.5)	217 (68.7)	0.01633	0.00026
Perfluorooctanoic acid (PFOA)	446 (29.5)	139 (43.9)	0.0036	0.00026
Perfluoropentanoic acid (PFPA)	349 (23.1)	170 (53.8)	0.00892	0.00015
Perfluoroheptanoic acid (PFHpA)	345 (22.8)	133 (42.1)	0.00131	0.00016
Perfluorooctane sulfonate (PFOS)	129 (8.5)	74 (23.4)	0.0064	0.00023
Perfluorobutane sulfonic acid (PFBS)	128 (8.5)	73 (23.1)	0.00219	0.00016
Perfluorononanoic acid (PFNA)	92 (6.1)	54 (17.1)	0.0009	0.00018
Perfluorohexanoic acid (PFHxA)	62 (4.1)	32 (10.1)	0.001	0.00016
Perfluorotridecane sulfonic acid (PFTriS)	33 (2.2)	30 (9.5)	0.00323	0.00028
Perfluorohexane sulfonic acid (PFHxS)	32 (2.1)	7 (2.2)	0.01343	0.00031
Perfluoroundecane sulfonic acid (PFUnDS)	32 (2.1)	30 (9.5)	0.00241	0.00024
Perfluorododecane sulfonic acid (PFDoS)	16 (1.1)	16 (5.1)	0.00141	0.00031
Perfluorododecanoic acid (PFDoDA)	12 (0.8)	12 (3.8)	0.00178	0.0003
Perfluorodecane sulfonate (PFDS)	12 (0.8)	12 (3.8)	0.00112	0.00022
Perfluorotridecanoic acid (PFTrDA)	9 (0.6)	9 (2.8)	0.00278	0.00052
Perfluoropentane sulfonic acid (PFPS)	9 (0.6)	2 (0.6)	0.0018	0.00021
Perfluoroundecanoic acid (PFUnDA)	7 (0.5)	7 (2.2)	0.00059	0.00031
Perfluoroheptane sulfonate (PFHpS)	5 (0.3)	2 (0.6)	0.00031	0.00015
Perfluorononane sulfonic acid (PFNS)	4 (0.3)	4 (1.2)	0.00023	0.00016
Perfluorodecanoic acid (PFDA)	2 (0.1)	2 (0.6)	0.0003	0.00014

The sum of PFAS value has exceeded the value of 0.01 µg/l 16 times at 9 sites throughout Scotland (Figure 4.1 – please note where there is not a one to one relationship between a sample point and a catchment, for example, if more than one raw water source supplies the sample point, these samples are not included on the map).

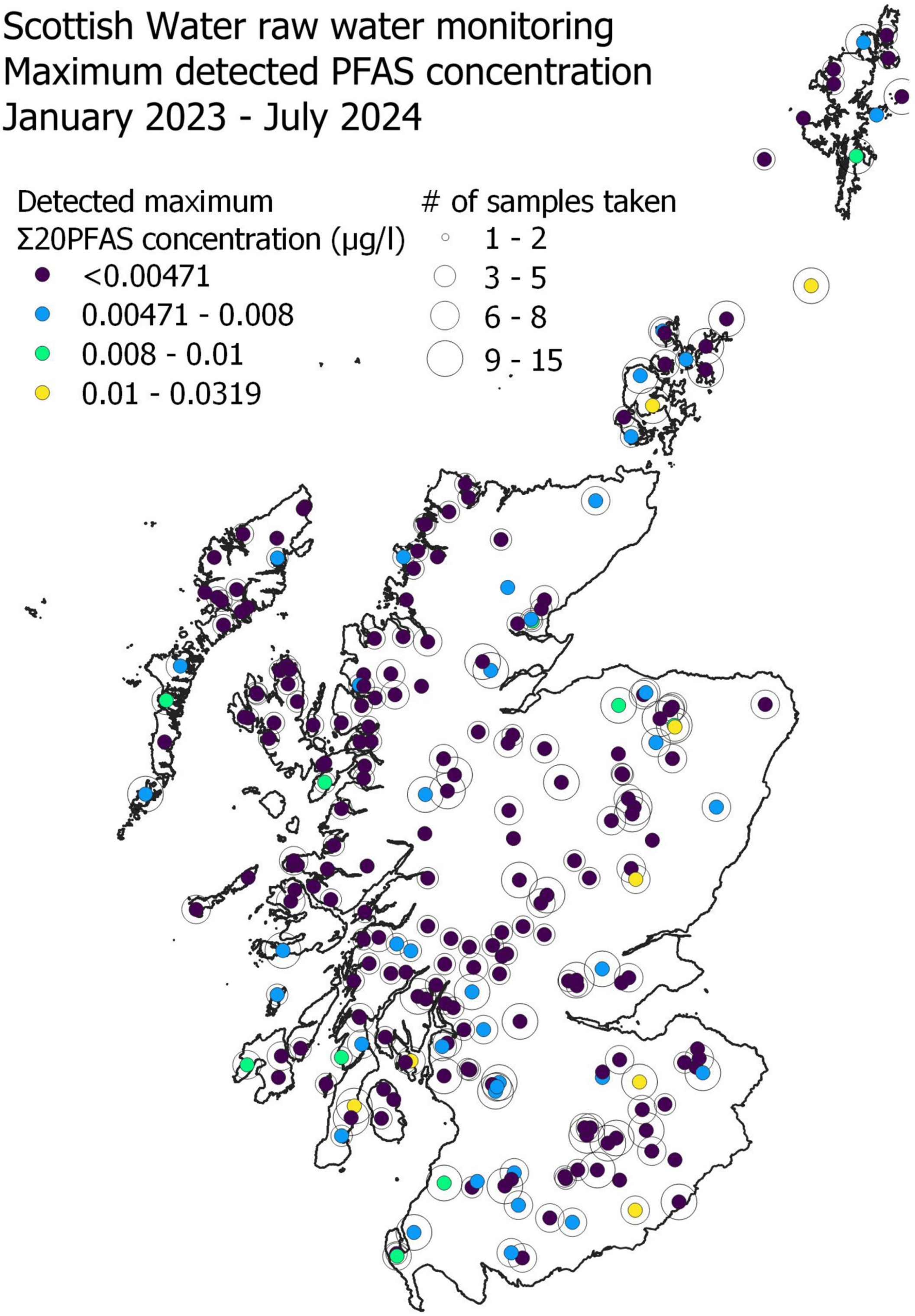


Figure 4.1: Scottish Water supply catchment centroids, mapped with sampling results for Sum of PFAS values from Scottish Water raw water monitoring from 12/01/23 to 08/07/24, with detected maximum concentrations (in µg/l) and number of samples taken. Regulatory limit for drinking water in Scotland: 0.1 µg/l. 252 of 316 sites included.

4.2.2 Ratios

There are two raw water supplies that have a ratio of PFHxS/PFOA of around 1, and one site where samples have a ratio of between 8 and 14.5. This site has an airport in the catchment. This site, as well as one sample from another site, also show PFHxS/PFOS ratios higher than 1. Only nine sites show PFOS/PFOA ratios of above 1. Three sites (two of which serve the same treatment works) have PFNA/PFOA ratios above 1. Where both PFHpA and PFOA were detected, more than 78.8% of samples had ratio of <1 (indicating local contamination sources), while no sample had a ratio >6 (highest ratio was 3.97). The fact that no samples show a higher ratio of PFHpA to PFOA also indicates that local sources play a predominant role in Scotland, rather than LRAT.

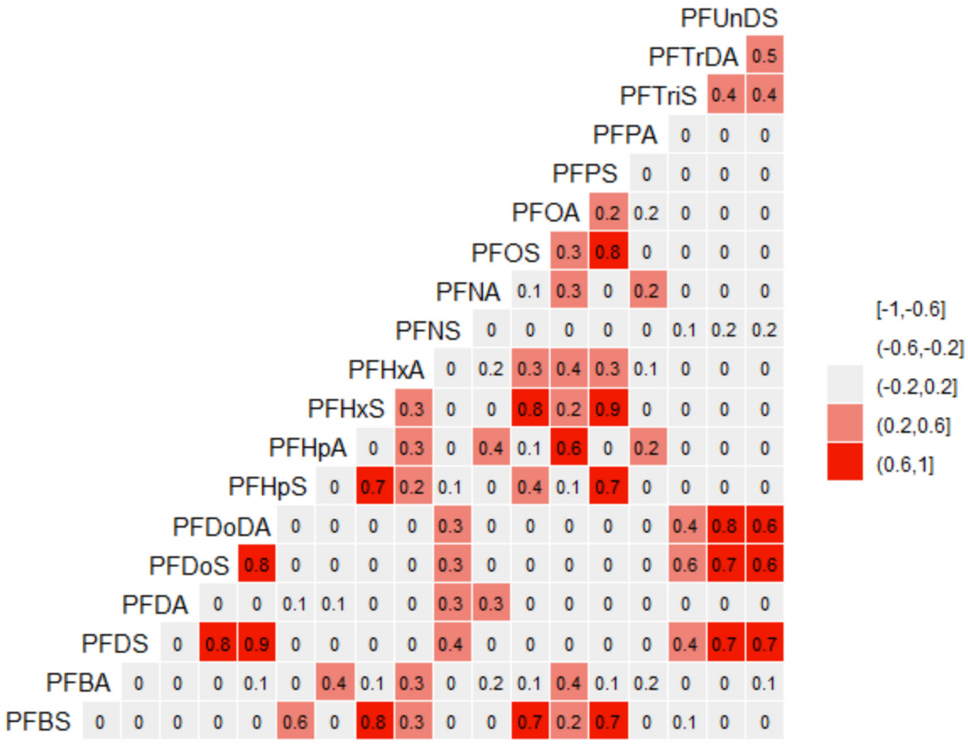
4.2.3 Substance compositions and relationships to sources of pollution

PFAS variables (concentrations of individual PFAS substances) are highly skewed and include many zeros (note that below limit of quantification values were set to 0 for the purpose of the analysis). This makes meaningful statistical analysis more challenging.

Correlations between PFAS variables over all samples show that longer chain PFAAs generally correlate. Correlations are also observable between PFSAs with chain lengths of 4-8 carbons. Short and medium chain PFCAs are less well correlated, except PFOA and PFHpA (Figure 4.2). These trends can also be observed when looking at spatial distribution of mean concentrations for longer chain PFAS (Figure A.1).

The PCA on individual samples that have at least one PFAS detection explains 23.2% of variance with Principal Component (PC) 1, 20.2% with PC 2, and 11.1 % with PC 3, so these three PCs together explain just over 54% of variance. The biplot for PC 1 and PC 2 (Figure 4.3) shows that the longer chain PFSAs (PFDS, PFDoDS, PFNS, PFUnDS, PFTriS) and the longest chain PFCAs (PFDoDA, PFUnDA, PFTrDA) correlate with PC 1, whereas the short and medium chain PFSAs (PFBS, PHHpS, PFHxS, PFOS, PFPS), as well as (to a lesser degree) PFOA and PFHxA, correlate with PC 2. PFBA, PFHpA, PFHxA, PFNA, and PFOA correlate with PC 3 (Table A.1 and Figure A.2).

Individual samples concentrate around the origin of the biplot, but there is an outstanding cluster in the upper left part of the plot that is strongly related to PC 2. These samples are all taken from the same site. There are also individual sample scattered along PC 1, with an outstanding one on the far right of the plot; these are mainly from different sites.



	PFBA	PFDS	PFDA	PFDoS	PFDoDA	PFHpS	PFHpA	PFHxS	PFHxA	PFNS	PFNA	PFOS	PFOA	PFPS	PFPA	PFTriS	PFTrDA	PFUnDS
PFTrDA																		0.5
PFTriS																	0.4	0.4
PFPA																0	0	0
PFPS															0	0	0	0
PFOA														0.2	0.2	0	0	0
PFOS													0.3	0.8	0	0	0	0
PFNA												0.1	0.3	0	0.2	0	0	0
PFNS											0	0	0	0	0	0.1	0.2	0.2
PFHxA										0	0.2	0.3	0.4	0.3	0.1	0	0	0
PFHxS									0.3	0	0	0.8	0.2	0.9	0	0	0	0
PFHpA								0	0.3	0	0.4	0.1	0.6	0	0.2	0	0	0
PFHpS							0	0.7	0.2	0.1	0	0.4	0.1	0.7	0	0	0	0
PFDoDA						0	0	0	0	0.3	0	0	0	0	0	0.4	0.8	0.6
PFDoS					0.8	0	0	0	0	0.3	0	0	0	0	0	0.6	0.7	0.6
PFDA				0	0	0.1	0.1	0	0	0.3	0.3	0	0	0	0	0	0	0
PFDS			0	0.8	0.9	0	0	0	0	0.4	0	0	0	0	0	0.4	0.7	0.7
PFBA		0	0	0	0.1	0	0.4	0.1	0.3	0	0.2	0.1	0.4	0.1	0.2	0	0	0.1
PFBS	0	0	0	0	0	0.6	0	0.8	0.3	0	0	0.7	0.2	0.7	0	0.1	0	0

Figure 4.2: Correlations matrix between different PFAS substances.

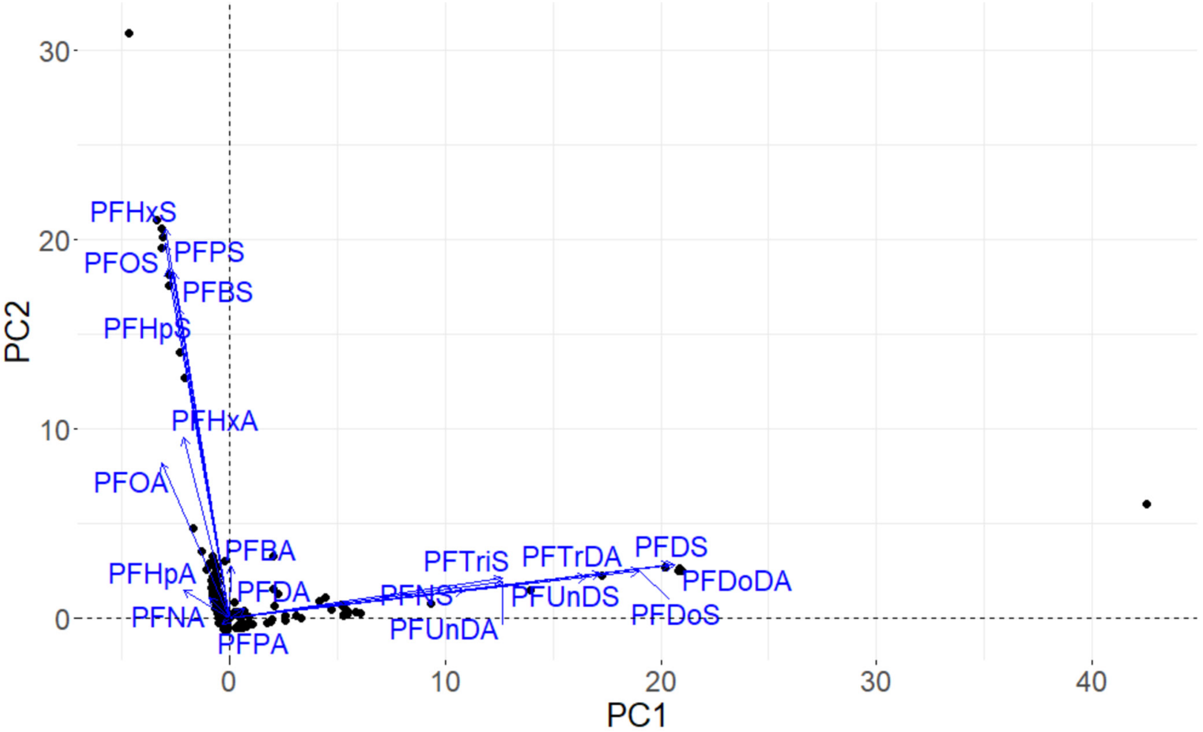


Figure 4.3: Biplot of the first two PCs from a PCA on concentrations for each PFAS substance in individual samples where at least one PFAS substance was above the limit of quantification (n=1003). Black dots are individual samples, blue arrows are PFAS substance. Points/variables in the right of the plot associate positively with PC1, to the left – negatively. Points/variables in the top associate positively with PC2, in the bottom – negatively. The longer the arrow/further from the plot origin the point, the stronger the association.

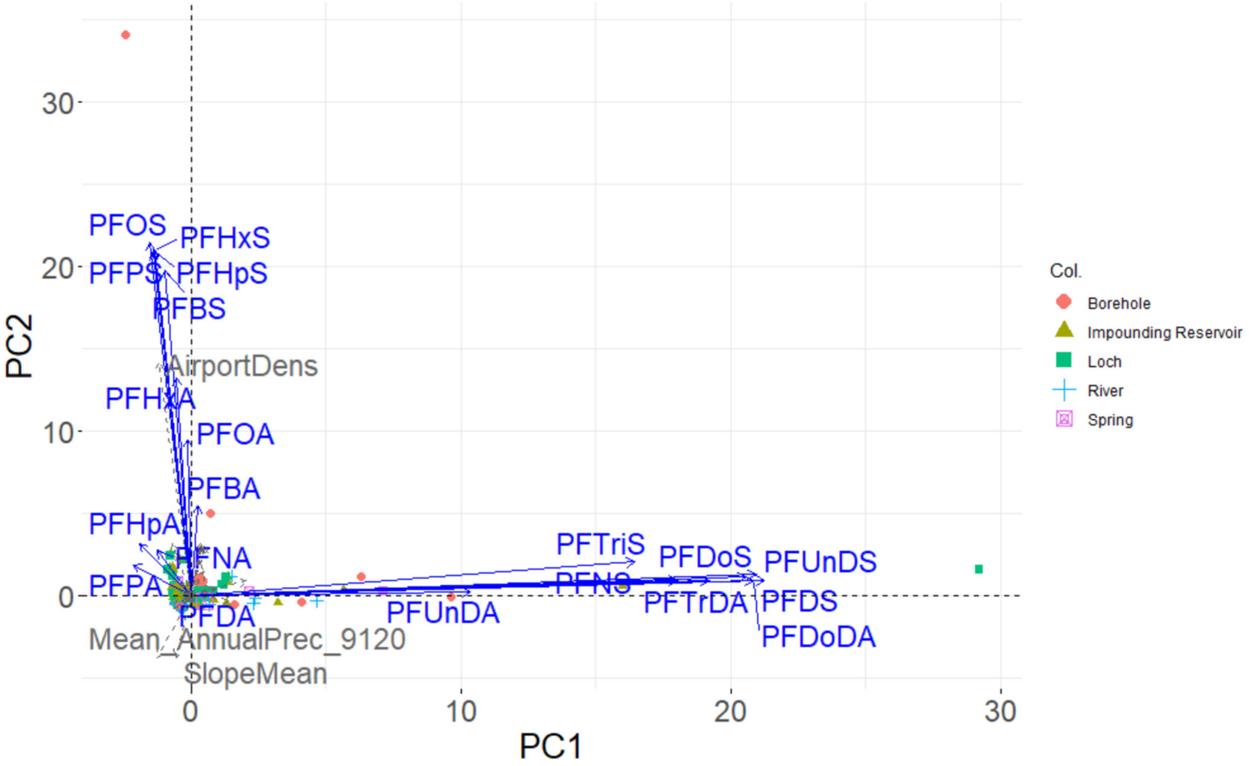


Figure 4.4: Biplot of the first two PCs from a PCA on mean concentrations per PFAS substance per catchment (n=317), with source risk factors superimposed as supplementary variables. Dots are catchments, symbolised by type of source, blue arrows are PFAS substance, grey dashed arrows are risk factors. Points/variables in the right of the plot associate positively with PC1, to the left – negatively. Points/variables in the top associate positively with PC2, in the bottom – negatively. The longer the arrow/further from the plot origin the point, the stronger the association.

Using the mean concentrations per substance per site allows the use of risk factors as described in Vorstius et al. (2024) as supplementary variables (this correlates the variables to the PCs but does not influence the PCA). This PCA explains 30.7% of variance with PC 1, 27% of variance with PC 2, and another 14.6% with PC 3, explaining 72.4% of variance with the first three PCs together. PC 1 and PC 2 show very similar patterns to the PCA run on individual samples, with PC 1 correlating to the longer chain PFSAs and longest chain PFCAs, and PC 2 correlated to the short and medium chain PFSAs, PFHxA, and PFOA (Figure 4.4). PC 3 again correlates to some PFCAs (PFBA, PFHpA, PFNA, and PFPA), but also negatively with some PFSAs (PFBS, PFPS, PFHpS, PFHxS). None of the supplementary variables correlates strongly with PC 1, and only airport density shows a good correlation to PC 2. Mean annual precipitation and mean slope are negatively correlated with PC 3 (Table A.2 and Figure A.3).

The PCA results for concentrations of all samples individually, and for mean concentrations, strongly resemble each other. As PC 2 is possibly strongly influenced by outlying concentrations from one particular site, the PCA on mean concentrations was rerun without this site (Figure A.4, Figure A.5 and Table A.3). This results in the same overall picture for PC 1 (explaining 32.4% of variation), with correlation to long chain PFAS and no significant correlations to source risk factors. For PC 2 (explaining 18.8% of variation), the short to medium chain (4-9 carbons) PFCAs and PFOS now correlate. There are also weaker correlations to a number of source risk factors, including industrial estates, metal recyclers, discharges to water, and negative correlations to mean slope and mean annual precipitation. This confirms that the PCAs using all data points are strongly influenced by one site. PC 3 (explaining 9.7% of variation) of the PCA with this outlier removed also shows a decoupling of most PFCAs and PFSAs, with positive correlations to PFBS, PFHxS, PFOS, PFTriS, and PFHxA, and negative correlations to PFBA, PFHpA, and PFPA. Strong correlations are with industrial estates, metal recyclers, discharges to water, septic tanks, arable area, area of potential biosolid application, and area of potential for paper waste discharge spreading.

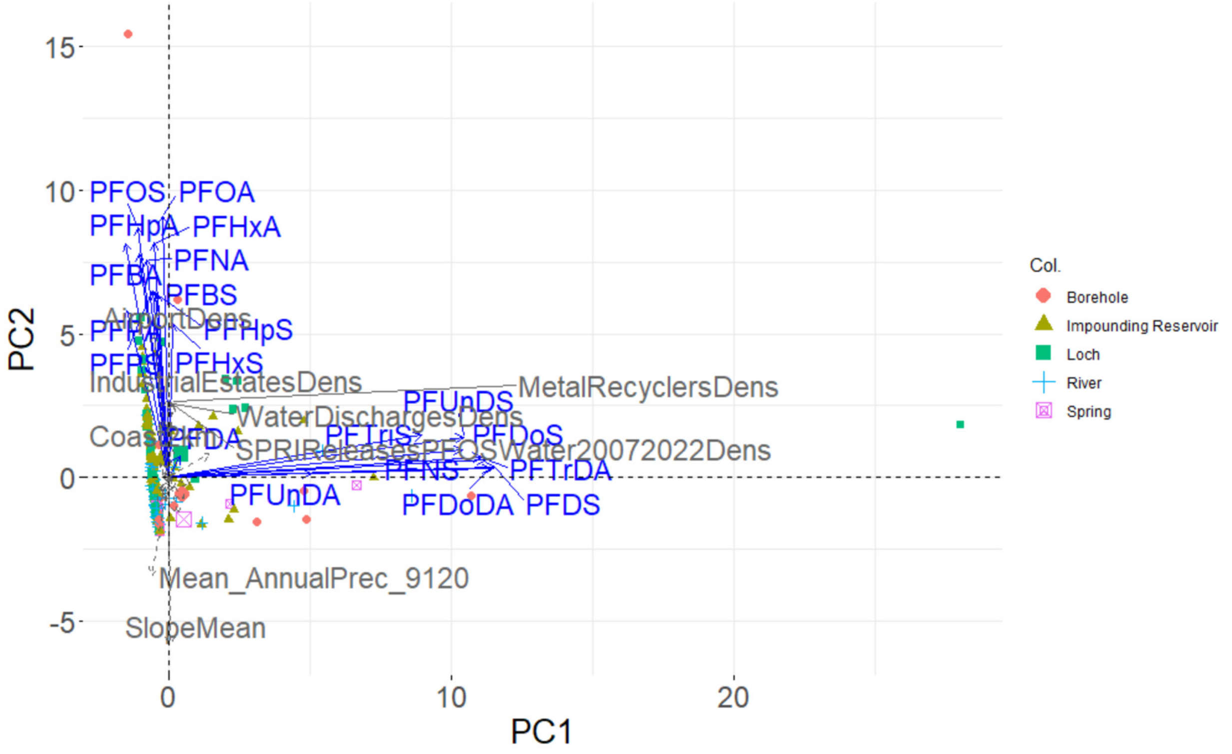


Figure 4.5: Biplot of the first two PCs from a PCA on percentage detection per PFAS substance per catchment (n=317), with source risk factors superimposed as supplementary variables. Dots are catchments, symbolised by type of source, blue arrows are PFAS substance, grey dashed arrows are risk factors. Points/variables in the right of the plot associate positively with PC1, to the left – negatively. Points/variables in the top associate positively with PC2, in the bottom – negatively. The longer the arrow/further from the plot origin the point, the stronger the association.

A PCA using percentage detection for each substance per site rather than mean concentrations explains 58% of variance (26.2%, 18.8%, and 12.9% for PC 1, PC 2, and PC 3, respectively). While the first PC still correlates to the longer chain PFSAs and longest chain PFCAs, the second PC correlates to all short and medium chain PFAAs (Figure 4.5), resembling the PCA on mean concentrations with the outlying catchment removed. There are also more, though not very strongly, correlated supplementary variables: airports, industrial estates, metal recyclers, wastewater discharges, and a negative correlation to mean annual precipitation and mean slope. The third PC shows positive correlations to shorter chain PFSAs (PFBS, PFHpS, PFHxS, PFPS) and negative correlations to short and medium chain PFCAs (PFBA, PHHpA, PFNA, PFOA), again resembling the other PCAs. Correlated supplementary variables are airports, industrial estates, metal recyclers, discharges to water, septic tanks, arable area, and a negative correlation to mean slope (Table A.4 and Figure A.6).

4.2.4 Influence of water source type

Looking at mean concentrations per PFAS substance per site, it can be seen that mean concentrations tend to be higher in surface waters, especially lochs and reservoirs, for the short and medium chain PFCAs (PFBA, PFPA, PFHpA, PFOA), except PFHxA (see Figure 4.6 and Figure 4.7 as examples).

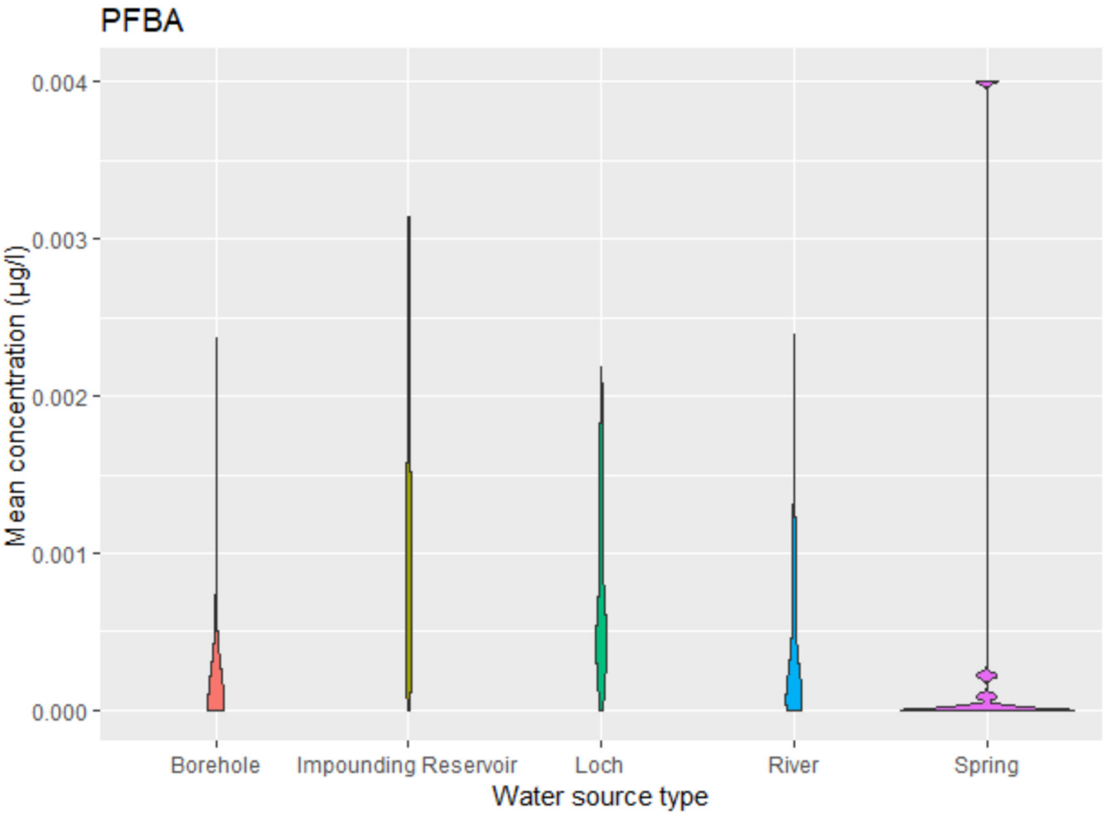


Figure 4.6: Violin plot of mean catchment PFBA concentrations by type of source. Means are statistically significantly different ($p<0.05$) between all types except reservoirs and lochs, and boreholes and springs.

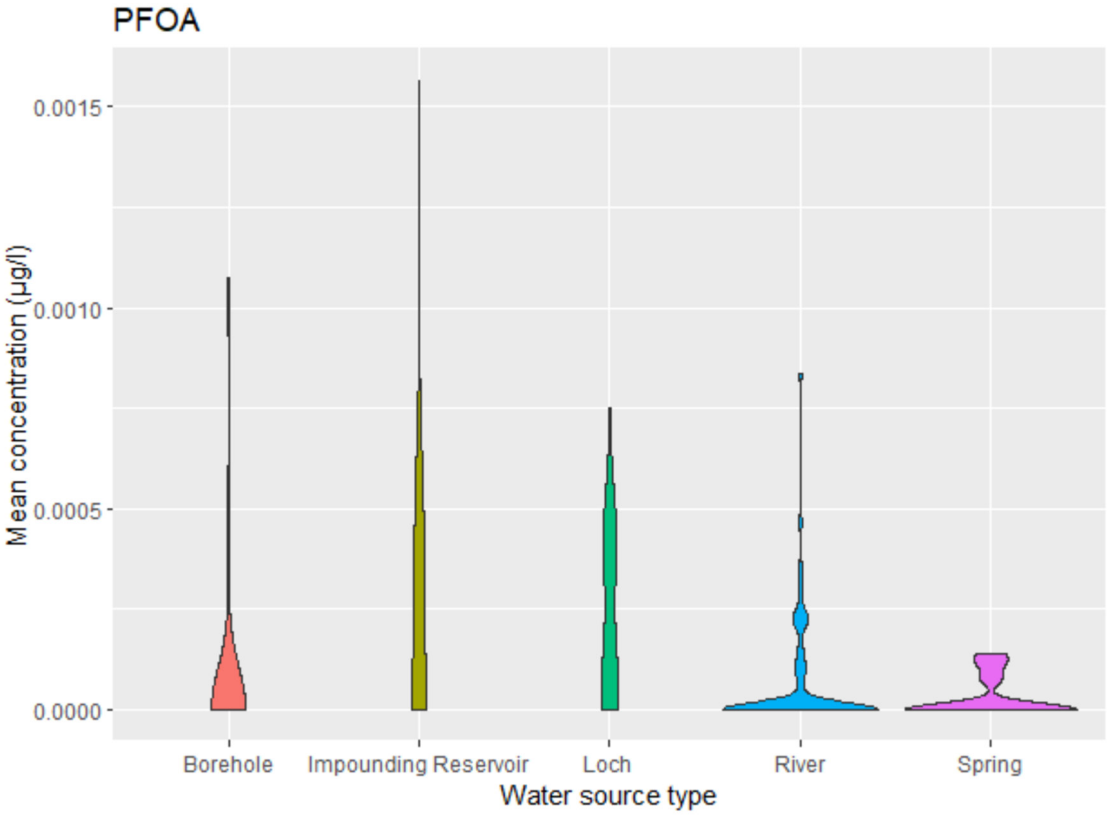


Figure 4.7: Violin plot of mean catchment PFOA concentrations by type of source. Means are statistically significantly different ($p<0.05$) between reservoirs and boreholes, river, and springs, and between lochs and rivers, boreholes and springs.

4.3 Discussion

The greater presence of shorter chain PFCAs, PFOA and PFOS, and sporadic high concentrations or detection frequencies of PFSAs, especially PFHxS, are consistent with the more mobile behaviour of shorter chain PFCAs, particularly PFBA, legacy contamination of PFOA and PFOS, and potential high influence of local contamination sources. Correlations between the less frequently detected longer chain PFAAs, as well as between PFSAs, could indicate common sources for PFSAs and for long-chain PFAAs. These patterns are confirmed by the PCAs. The PCAs run on concentrations show independent signals for these two groups of PFAS, although none of the risk factors identified could explain the presence or concentration of longer chain PFAS. Literature suggests industrial activity leads to a higher presence of long chain PFAAs (Anderson *et al.,* 2023). This is not observable in this dataset, although the low number of samples with detection of these substances could hinder any meaningful statistical connection.

Airport density especially seems to be connected to higher concentrations and detections of PFSAs, however this may be strongly influenced by one site showing particular high concentrations/detections of PFBS, PFHxS and PFOS, and having an airport in the catchment. The less well-defined signal of the grouping of short to medium chain PFCAs (on concentrations) or on short and medium chain PFAAs (on percentage detection) could be interpreted through several factors, making it likely that many sites are influenced by a mix of sources. This is supported by the correlation of a number of supplementary variables to the second PC of the PCA using mean PFAS concentrations. The fact that no samples show a higher ratio of PFHpA to PFOA also indicates that local sources play a predominant role in Scotland, rather than LRAT. The negative correlation of mean annual precipitation in both PCAs, is indicative of precipitation not being a suitable proxy for wet deposition, although inferences from this relationship are difficult as the timescales for the annual precipitation mean and the sampling do not match.

Differences between groundwater and surface water are manifest mainly in the higher concentrations of short to medium chain PFCAs in surface water as compared to groundwater. On the other hand, highest concentrations of PFSAs have been found in groundwater sources, although there is no clear trend observable. This may again reflect a higher mobility of short chain PFCAs as compared to longer chains and PFSAs, and a quicker transport of these substances through soils and to surface water sources. Groundwater may be impacted for longer by PFSAs due to a longer retention time in soils, leading to a slower release to groundwater.

4.4 Implication for the RWPP assessment update

The analysis of the data showed no clear predominant sources, suggesting rather that many sites experience diffuse pollution. This could explain why many risk factors did not or only weakly correlated to the PCs.

The assessment included a rather large number of risk factors, which would mirror the hypothesis that freshwater in Scotland is impacted by a mixture of sources, and which led to a pattern of identified risk reflecting population density and industrial activity in Scotland. However, locally some sites may be heavily influenced by a specific source, leading to the highest observed concentrations. This gets “lost” amid all other considered risk factors in the assessment. Weighting the risk factors could alleviate this effect. Candidate risk factors for more weight based on the data analysis would be airport density, industrial estate density, density of metal recyclers, and waste water discharge density.

Alternatively or additionally, separate assessments could be prepared by groups of PFAS. As the data shows highest contamination from PFBA and other short chain PFCAs, and locally from PFHxS, these would be candidate groups for separate assessments. These assessments could be based on fewer risk factors, or differentially weighted risk factors, that are considered to be of predominant importance. The results could also inform monitoring for additional substances; e.g., FTSAs could be important constituents of PFAS mixtures especially at sites potentially influenced by industry and AFFF, where there is also higher likelihood of elevated PFSA concentrations.

5 PFAS RWPP assessment update

Following the literature review on the individual topics and the data analysis, the following updates to the RWPP assessment have been carried out:

- Including risk of PFAS pollution from sea spray aerosols (SSA) in the assessment, represented by a 2 km coastal “risk” zone of SSA influence (see 3.3.3);
- Replacing the pathways used (moderate to high aquifer productivity and topsoil pH above 6) by a pathway based on groundwater vulnerability (see 3.4.3);
- Removing mean annual precipitation as a risk factor until better understood as a potential risk factor/proxy (see 4.4);
- Splitting the assessment by PFAS groups, and prepare PFAS subgroup assessments for short and medium carbon chain length PFCAs, short to medium chain PFSAs, and long chain PFAS (see 4.4).

These changes were included in the updated PFAS RWPP assessment and are presented and discussed in this section.

5.1 Methods

The rationale for, approach to, and putting together of the RWPP assessment has not changed from the previous project and is described in section 2.2 of Vorstius *et al.* (2024). The following section describes methods where additional resources were used or changes made.

5.1.1 Preparing new and additional risk factor layers

Potential PFAS pollution from SSA was represented by a zone of influence spanning coast to 2 km inland. To prepare this layer, a polygon shapefile of this zone was created and the percentage area lying in this zone was calculated for each Scottish Water supply catchment.

The groundwater vulnerability layer from the Hydrogeology of Scotland maps created by the British Geological Survey (BGS) was used (Ó Dochartaigh *et al.,* 2015) to create a new pathway layer. Areas identified with groundwater vulnerability class 5 (defined as “vulnerable to most pollutants, with rapid impact in many scenarios”) were chosen as representing areas more likely to facilitate movement of PFAS substances through the soil and subsoil and reach freshwater sources, and were extracted from the layer. It was considered to also include areas classed as 4 for groundwater vulnerability (defined as "vulnerable to those pollutants not readily adsorbed or transformed") as part of this pathway, to reflect the reduced adsorption tendencies of shorter chain PFAS. However, this would have covered the totality of the area of many catchments, making the assessment too indiscriminate.

Topsoil organic carbon content was included as a pathway consideration in the grouped assessments (5.1.2.2). To this end, areas with less than 5% of organic carbon in the topsoil were extracted from the topsoil organic carbon content map prepared by the James Hutton Institute (JHI, 2014). Five percent organic carbon content was used as a threshold based on Tran *et al.* (2022), showing increasing concentrations of PFOA and PFOS in soils with app. 5% organic carbon content and more.

5.1.2 Risk factor inclusion and weightings in the assessments

5.1.2.1 Update of the RWPP assessment for the 20 PFAS compounds

The PFAS RWPP assessment was created for all 20 PFAS compounds together in two versions: without pathway considerations, and all risk factors weighted equally; and with a pathway consideration. The initial RWPP assessment with pathways used areas on top of moderate to highly productive aquifers and areas with a median topsoil pH above 6 as two pathways, identifying these as characteristics likely to facilitate movement to freshwater sources. In this updated assessment, these two pathways were replaced by a single pathway based on areas identified as having groundwater vulnerability class 5, and risk factors on these areas were double weighted in the assessment. As soil pH is not considered in the groundwater vulnerability classification, it was considered to keep this pathway and combine the two pathway layers as previously. However, to better understand the impact of including groundwater vulnerability considerations, and keeping the assessment initially simple, it was decided to trial groundwater vulnerability as the only pathway.

5.1.2.2 Creating RWPP assessment for subgroups

Apart from updating the overall assessment for the 20 compounds included in the drinking water standard, the assessment was split to create three separate assessments for subgroups of PFAS. PFAS were grouped according to pollution sources and environmental behaviour, i.e. grouped together if they have common sources and share characteristics that facilitate their movement through the environment. Within these assessments, groundwater supplies and surface water supplies were treated differently due to using different pathway options: groundwater vulnerability for groundwater supplies, and either topsoil organic carbon content or soil pH for surface water supplies.

A) Short to medium chain (4-9 carbons) PFAAs:

Short chain PFCAs are shown to move more easily through the environment, leading to a more even distribution through the environment. They are also potential degradation products from longer chain PFCAs and PFSAs. All identified potential PFAS sources are important for these substances, but dominance of short to medium chain PFCAs is usually associated with influence of waste products, e.g. wastewater discharges, landfills, waste application to land. These source risk factors were therefore weighted higher for this group.

Campos Pereira *et al.* (2018) found organic carbon content to be a predictor for sorption to soil for shorter chain PFAS, so the organic carbon content pathway layer was used for surface water supplies in this group.

B) Short to medium chain (4-8 carbons) PFAAs:

Effluents from and environmental media influenced by AFFF impacted sites, industrial sites, and chrome plating, show a higher proportion of PFSAs compared to other PFAS pollution sources, and were therefore attributed a higher weight for the RWPP assessment for this group. Topsoil organic carbon content was also used as pathway consideration for surface water supplies in this group.

C) Long chain (10-13 carbons) PFASs:

Compared to other pollution sources, areas influenced by industrial activity show correspondence to a higher proportion of long chain PFAS. These sites were therefore weighted higher for the assessment. As long chain PFAS have been phased out over the past decades and tend to be detected less frequently in Scottish freshwater sources, most source risk factors were weighted less strongly compared to the other groups.

Campos Pereira *et al.* (2018) showed that, contrary to shorter chain PFAS, soil pH is a better predictor for sorption to soil for long chain PFAS, so for this group, soil pH was chosen as a pathway factor for surface water supply sources.

Specific weights applied to each source risk factor (per pathway) for each group are shown in Table A.5.

5.1.2.3 Mapping raw water monitoring data

Mean concentrations per sample site of individual PFAS were mapped using the raw water monitoring data for the 20 PFAS compounds from Scottish Water for the sampling period 12/1/2023 to 8/7/2024. For statistical analysis and mapping of concentration means (Figure A.1), results below the limit of quantification (LoQ) were set to 0 (see 4.1).

5.2 Results

5.2.1 Sum of 20 PFAS RWPP assessment

Overall, the RWPP assessment for the 20 PFAS especially identifies catchments in the Northeast, the Central Belt, and the South of Scotland as of higher risk for raw water PFAS pollution (Figure 5.1). This can be explained by higher pressures through population and industry presence. Including the new pathway of groundwater vulnerability increases catchment risk scores especially along the West coast of Scotland.

In comparison to the initial RWPP assessment, the assessment without a pathway consideration was changed by including an additional risk factor (SSA) and removing a risk factor (long range atmospheric transport and wet deposition represented by mean annual precipitation). Due to the large number of risk factors included in the assessment, this changes the risk scores only slightly for most catchments.

Replacing the two pathways of topsoil pH and aquifer productivity with groundwater vulnerability leads to a more marked difference. The initial two pathways tended to exacerbate the difference between the high and low scoring catchments, as the pathways overlapped the areas of higher risk factor densities. The groundwater vulnerability pathway however leads to increases mainly in areas of lower risk catchments (West coast and Highlands). This seems to be a better match for actual observations.

5.2.2 PFAS Subgroups RWPP assessment

The overall patterns of the subgroup RWPP assessments are similar and resemble the RWPP assessment for the 20 substances, with highest risk catchments in the Northeast, the Central Belt and the South of Scotland. However, the differences in weightings for risk factors lead to some subtle changes in the assessments (Figure 5.2).

The assessment for short and medium chain PFCAs lead to slightly higher scores for many catchments. This could be due to higher weight given to risk factors that tend to be widespread, e.g., septic tanks. In comparison, the assessment for the longer chain PFAS highlights fewer catchments as high risk, due to many risk factors given a lower weight. This matches with observations that longer chain PFAS are less frequently detected and usually in comparatively low concentrations. The difference in pathway layers used for surface water supply sources does not lead to many differences, probably due to much spatial overlap between the two layers.

5.2.3 Scottish Water raw water monitoring results

Mean raw water concentrations for each PFAS substance were mapped and are presented in Figure A.1. It is observable that relatively high concentrations are widespread for PFBA and PFPA, followed by PFOA, PFOS and PFBS. Concentrations of PFHpA tend to be lower, although detections are also widespread. While patterns of concentrations between PFBA, PFPA, and PFHpA appear similar, higher concentrations of PFOA occur in the South, especially the Southwest. Based on this observation, there may be a possible atmospheric influence of the fluoropolymer production site in Lancashire in England (see 3.1.1), as it is known that historic PFOA emissions occurred. This would be especially significant as the site now emits shorter chain PFAS, with possible impacts on areas in Southwestern Scotland.

With exceptions of a few catchments showing relatively high concentrations of PFSAs, mean concentrations tend to be lower for this group. If looking at the maps for PFBS, PFHxS, and PFOS, higher mean concentrations stand out for a few catchments, including the catchment consistently identified as of highest risk in all RWPP assessments in the South of Scotland. Mean concentration patterns for PFHxA also bear some resemblance to the PFSA group. PFHxA is associated with AFFF contamination alongside PFSAs, making this connection unsurprising. Mean concentrations of longer chain PFAS are also lower and less widespread compared to short and medium chain PFCAs.

Using Spearman's correlation analysis between risk scores and mean concentrations, none of the correlations between the mean concentrations and risk scores of the subgroup assessment in the short and medium chain PFCAs are significant, although it is near to significance for PFBA (p=0.07). For the short to medium chain PFSA group, both PFHxS and PFOS mean concentrations show significant correlations to catchment risk scores (p=0.0002 and p=0.013, respectively). From the long chain PFAS group, PFTriS mean concentrations are significantly correlated to catchment risk score (p=0.018).

5.3 Discussion

The current update of the RWPP assessment made several changes to the initial assessment. The replacement of the pathway layer results in an RWPP assessment that highlights intrinsic vulnerability of catchments in the West and the highlands of Scotland. A further refinement could involve the application of different pathway layers depending on water supply source type, as applied in the subgroup assessments, with a combination of two pathways (topsoil pH and topsoil organic carbon content) for surface water supplies. However, a caveat of this approach is that it disregards the reciprocal influence of groundwater and surface water that has been evidenced from the literature (see 3.4). Alternatively, all these pathway layers could be combined. With more data becoming available in future, better conclusions can be drawn about the appropriate pathways.

There are only slight differences in the RWPP assessments prepared for different groups of PFAS substances, with overall similar spatial patterns of areas of higher and lower comparative risk. Looking at mean concentrations of the individual PFAS substances, there are however different spatial patterns observable. Changes to weightings could be made in the subgroup assessments to further reflect these differences, however as with the pathway approach, evidence for making judgments on appropriate weightings is lacking at this stage.

Importantly, high presence of risk factors is not always associated with observations of increased PFAS concentrations. This could be due to potential of risk factor presence being used in the assessment rather than actual, meaning that a risk factor is assumed to be present but is not. Risk factors may differ in character, influencing a possible release of pollutants. For example, landfills differ in size, age, engineering, and/or types of waste received, and have thus different pollution potential. Local conditions also influence transport from pollution source to water body. Both cases challenge a screening type assessment, and trying to improve on these limitations would often be time and resources consuming with small overall benefit to the assessment.

On the other hand, there are still catchments showing comparatively high PFAS concentrations but are not being picked up in the RWPP assessments. Due to the many possible sources of PFAS, it is possible that sources have been overlooked. However, some risk factors have been represented by proxies that reflect many pollution sources associated with human activities, e.g., road density or industrial estate density. Movement through the environment is therefore probably important to explain the mismatches between assessment scores and observed PFAS concentrations. As discussed above, these will largely depend on local conditions difficult to represent in a national-scale risk screening.

Looking at PFAS substances individually and as subgroups allows a better interpretation of possible pollution sources. Correlations between some of the PFSA mean concentrations and assessment scores are encouraging, indicating that sources for some PFSAs are well identified. This group is slightly less mobile in the environment, making the assessment less challenging compared to the shorter chain PFCAs.

To further increase understanding of PFAS pollution sources, and fate along the transport to freshwater sources, a wider array of monitored PFAS substances would be helpful, especially FTOHs, FTSAs, FTABs, FTCAs, FASAs, or ether-PFAS. Similarly, suspect or non-target screening methods could support better source allocation, as well as provide a clearer picture of PFAS pollution in the Scottish environment, especially as it has been shown that total PFAS is not always correlated to the sum of a PFAS subgroup (Göckener *et al.,* 2023).

Aspects to still be explored in the monitoring data are seasonal aspects, as well as relationships between concentrations and short term dynamic environmental conditions, especially weather-related factors. This would help to understand if PFAS substances are subject to mobilisation or dilution, and if there are differences in processes between substances and sites. For example, it is observable that PFPA is inconsistently detected at several sites. However, if it is detected, it is often

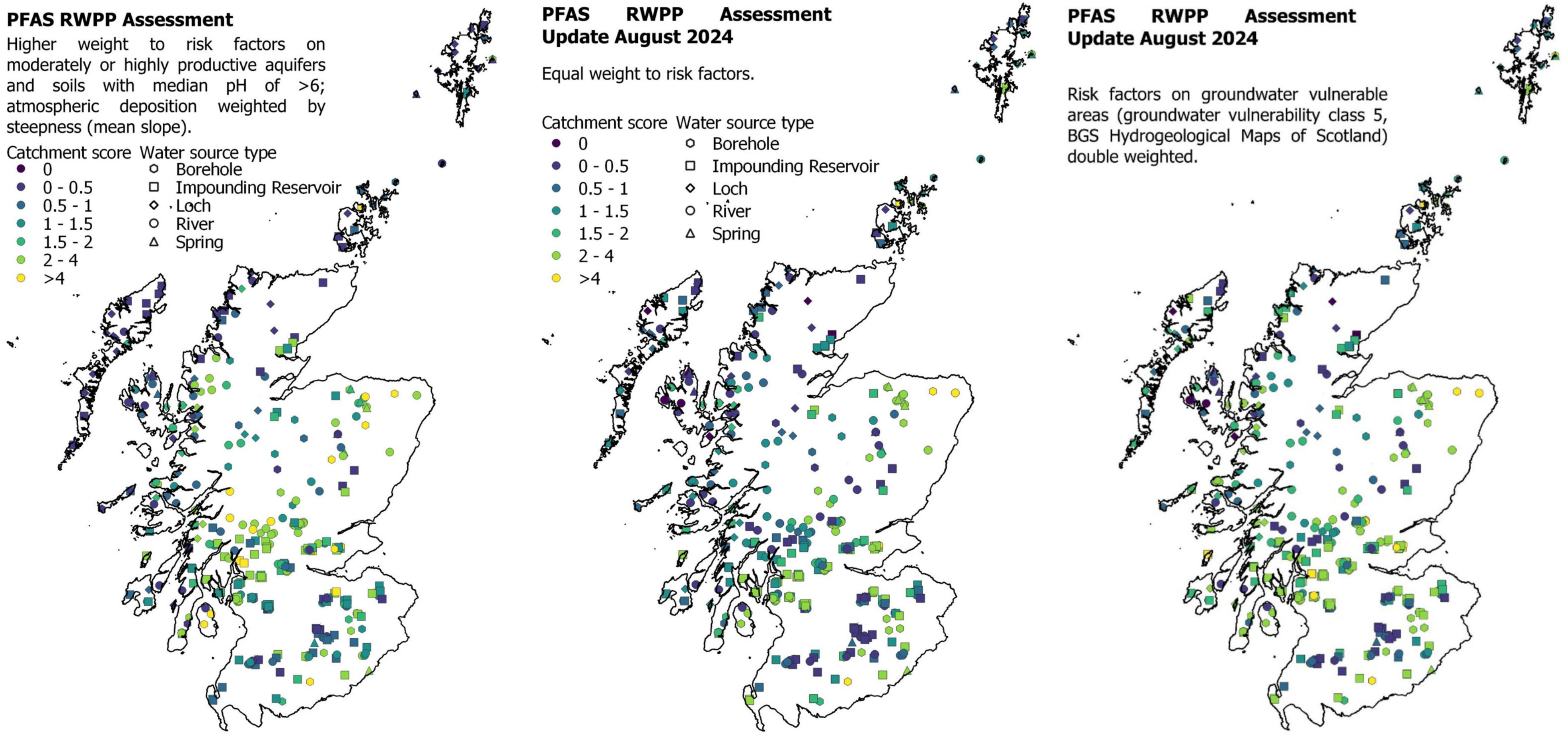


Figure 5.1: RWPP assessment for all 20 PFAS substances included in the Scottish drinking water standard. Left: Previous assessment with pathway considerations, risk factors on moderately to highly productive aquifers receiving higher weight. Middle: Updated assessment, without pathway considerations; equal weight to all risk factors. Right: Updated assessment with pathway considerations; risk factors on groundwater vulnerable areas (class 5) double weighted.

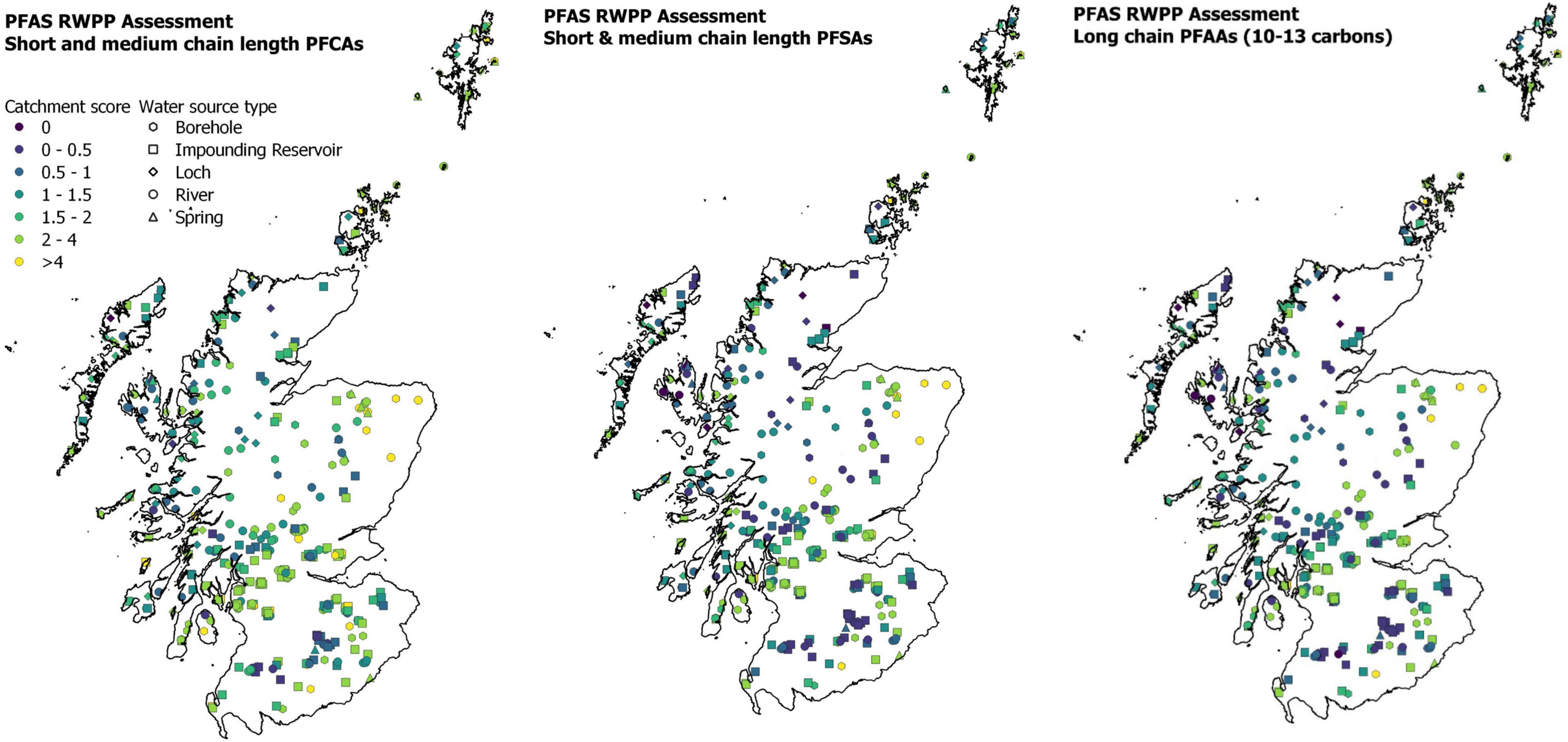


Figure 5.2: RWPP assessment for PFAS subgroups. Left: Assessment for short to medium chain (4-9 carbons) PFCAs. Middle: Assessment for short to medium chain PFSAs. Right: Assessment for long chain (10-13 carbons) PFAS.

the major component of the sum of PFAS, while other PFAS substances show less variation in concentration at the same site.

Relationships to climate, seasonality, and weather are especially important as climate change may alter catchment conditions and change processes. Mobilisation and dilution effects will directly be influenced by changes in climate, potentially leading to changes in risk patterns. Climate change may also have more indirect effects. For example, changing soil conditions or reservoir dynamics may influence retention of, or release from, PFAS in soils and sediment.

5.4 Conclusion

The update of the PFAS RWPP constitutes the first iteration of a continuous improvement cycle as outlined in Vorstius *et al.* (2024). There are some changes that can be observed compared to the first RWPP assessment, most notable a slight increase in risk score for some coastal Western catchments, and a reduction in risk score for catchments in the Western Central Belt. This can be attributed to the inclusion SSA as a risk factor, which impacts risk scores for coastal catchments; the replacement of the pathway layers that highlights different areas as potentially vulnerable, especially in the West and in the highlands; and the removal of mean annual precipitation, which removes a widespread risk factor.

The RWPP assessments for a reduced group of PFAS substances were a first trial of using the recognised patterns to inform a more targeted assessment. The resulting assessments showed some differences in risk scoring for individual catchments but had strongly resembling overall patterns. This is due to almost all risk factors being included in all RWPP assessments, albeit with different weighting, as the observed groups of PFAS (PFCAs and PFSAs) have many common sources. Inclusion of further PFAS, especially intermediate PFAS substances, could help further differentiation.

While the Scottish Water raw water monitoring now yields a substantial number of samples, due to the spatially widespread sampling and the inclusion of 20 substances, numbers of samples at individual sites can be very low. The data therefore needs to be interpreted with caution as it is not possible to identify if sampling results are representative for the individual sites, or whether there are any temporal patterns or trends. Verification of the risk assessment by monitoring data is therefore still premature.

The biggest challenges in creating meaningful outputs for an overall assessment, as well as subgroup assessments, remain our limited knowledge on relative importance of pollution sources, as well as the great variety of potential impact within one category of pollution sources. For example, industrial activities will have very different magnitudes of PFAS emissions and pollution potential; wastewater treatment works are known to produce effluents with varying concentrations of PFAS, etc. Another factor is the complex influence of static and dynamic local characteristics, such as weather components. Monitoring guided by and interpreted with the help of the RWPP assessment, can yield further insight into these crucial questions.

6 Recommendations

Based on the findings of the literature review, the data analysis, and the PFAS RWPP assessment update, the authors put forward the following recommendations:

1. Explore the feasibility of including further PFAS substances in the drinking water monitoring

While comparatively high concentrations of the sum of the 20 PFAS currently included in the sampling can indicate where total PFAS concentration is high, this may overlook sites with high concentrations of alternative or precursor PFAS. Candidate substances for additional monitoring to address this issue include FTOHs, ether-PFAS, FTSAs, 5:3 FTCA, FTABs, FASAs and related compounds, PFPiAs and PFPAs. These substances can furthermore help to characterise pollution sources, e.g., 5:3 FTCA is commonly associated with landfills.

2. Explore the feasibility of suspect and non-target screening to support our understanding of PFAS distribution in the Scottish freshwater environment

Health implications of many new or understudied PFAS compounds are currently unclear. Limitations in our analytical abilities to determine many PFAS substances, and uncertainties regarding their health impacts, challenges a management approach based on total PFAS. Non-target or suspect screening methods could support a better understanding of the number (and eventually also identity) of PFAS substances present in water. This would develop a better understanding of PFAS in the environment and identify priority areas for further research. While these methods may currently be unfeasible for large scale application, developments in this area should be observed to understand their potential for managing PFAS in drinking water.

3. Investigate relationships of PFAS concentrations to other sampled pollutants or water quality indicators

Some of the risk factors included in the RWPP assessment are represented by proxies, and/or build on potential for pollution rather than actual pollution. Investigating linkages to other water quality parameters could provide some evidence for the impact of the risk factors. For example, concentrations of pesticides or nutrients could provide better insight into the likelihood of agricultural practices influencing the supply source; presence of other observed industrial chemicals could indicate general pressure from industry in an area; etc. These relationships may not be identifiable when investigating the complete dataset but might be observed in a subset.

4. Analyse for seasonal trends/patterns in the data

The current dataset provides limited opportunity for this aspect as it still covers a limited timespan; however, testing for seasonal differences in concentrations of the different PFAS, or relating concentrations to weather parameters would be beneficial to understand PFAS mobilisation processes. This will be of particular importance to understand potential climate change impacts on PFAS concentrations.

5. Investigate relationships of PFAS concentrations to raw water intake characteristics

There are still many knowledge gaps concerning fate of PFAS in freshwater. For example, distribution characteristics and dynamics of PFAS within reservoir and lakes is largely unknown. Therefore, it is not clear if the mechanics of how water is abstracted from a supply impact PFAS concentrations; e.g., the depth of the water abstraction intake; residence time of abstracted water in the water body; exchange with sediments; etc.

These recommendations aim at improving the basis for understanding PFAS pollution in the environment in Scotland, with a focus on supporting the regulation and management of public drinking water supply. They outline next steps to be taken that will generate more insights to continue the cycle of improvement for risk assessment, and thus further improve guidance for monitoring and regulation.

Previous recommendations made in Section 4 of Vorstius *et al.* (2024) have a wider scope, intending to capture the challenge of PFAS pollution more holistically, and are still considered valid.

7 Conclusion

This research, commissioned as a three month 'call-down' project by Scotland's Centre of Expertise for Waters (CREW), is a follow-on study addressing some of the knowledge gaps identified in the project "Developing risk assessment approaches for PFAS and watch list parameters under the recast Drinking Water Directive – PFAS, 17ß-estradiol, nonylphenol" (Vorstius *et al.,* 2024). It focused on four topics that provided scope for improving the original PFAS RWPP assessment and used the available data to gain first insights into PFAS substances composition and occurrence in association with identified risk factors.

The literature review provided evidence for including a risk factor based on SSA influence in coastal regions, as well as improving the pathway considerations by using considerations of groundwater vulnerability. The data analysis showed grouping potential for some PFAS substances and gave first indications of important explanatory risk factors. This led to the removal of mean annual rainfall as a risk factor proxy from the updated assessment, until better understanding of its effect on PFAS concentrations in Scotland is achieved. Furthermore, it supported the preparation of individual RWPP assessments for PFAS subgroups that utilised different weightings for risk factors and differentiation of pathway considerations dependent on water supply type.

With more monitoring data becoming continuously available, this will eventually lead to a more reliable picture of PFAS concentration pattern across Scotland. Comparison to the assessments will allow a better understanding of sources of pollution, and catchment characteristics that influence PFAS mobility in the environment. The assessments can also serve to guide study designs for testing specific hypotheses around PFAS pollution sources and transport. This will ultimately improve our ability to manage PFAS in Scotland.

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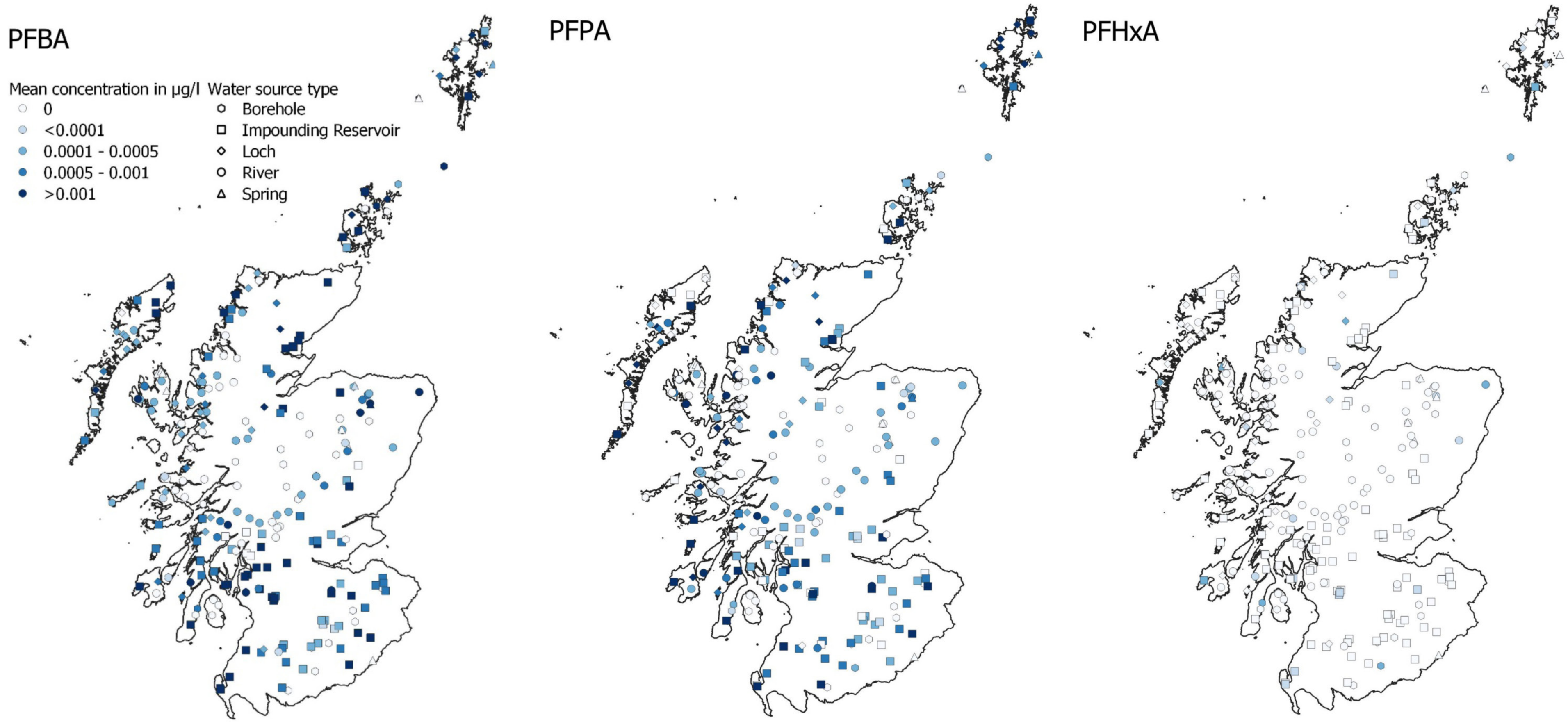
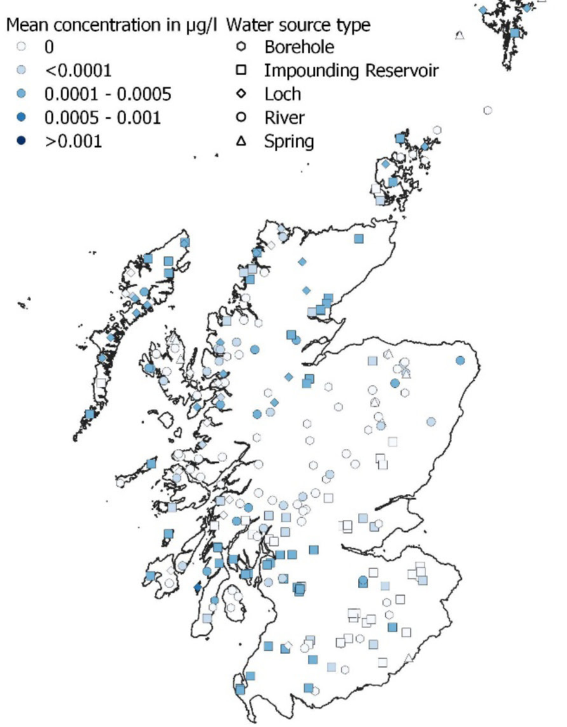
Appendices

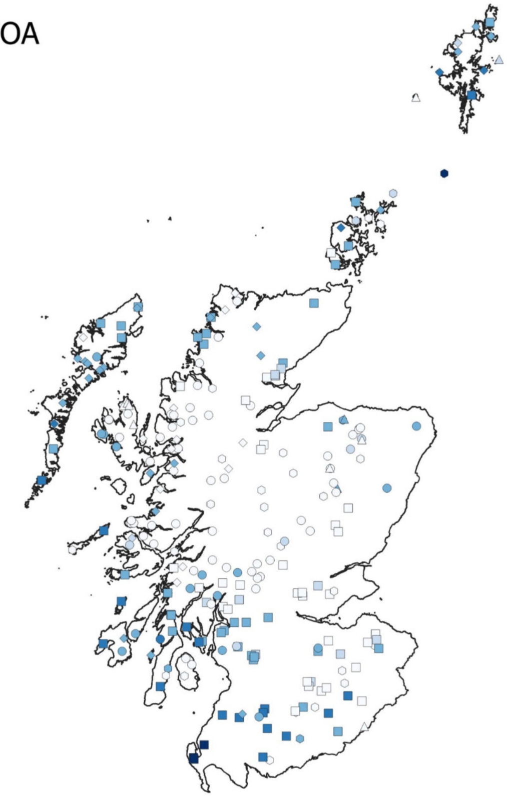
Appendix A. PFAS substances mean concentration maps


PFBA
PFPA
PFHxA
Mean concentration in µg/l
○ 0
● <0.0001
● 0.0001 - 0.0005
● 0.0005 - 0.001
● >0.001
Water source type
○ Borehole
☐ Impounding Reservoir
◇ Loch
○ River
△ Spring

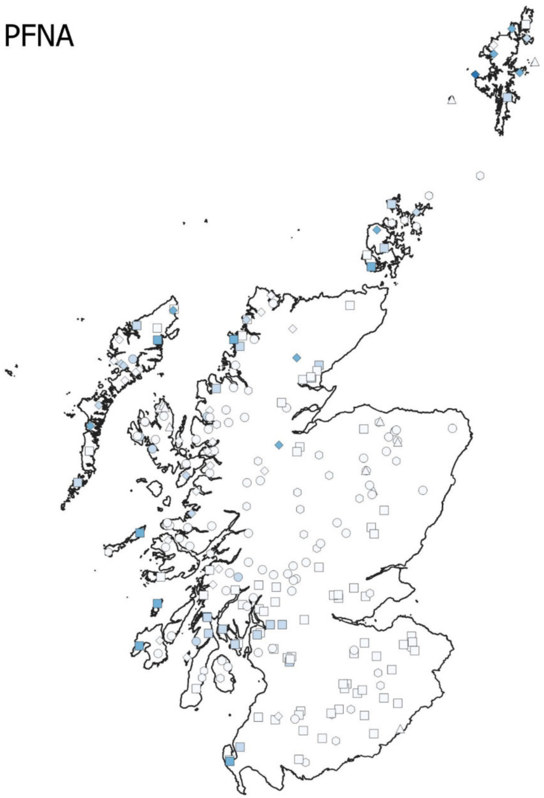
PFHpA



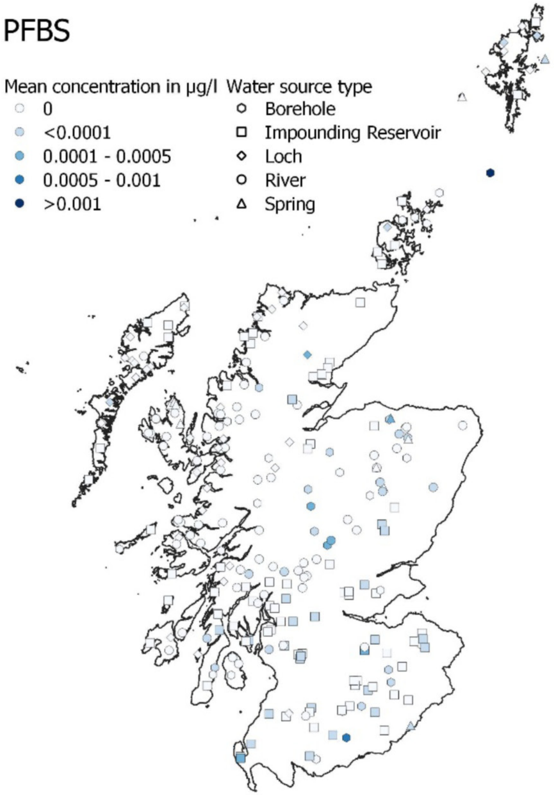
PFOA



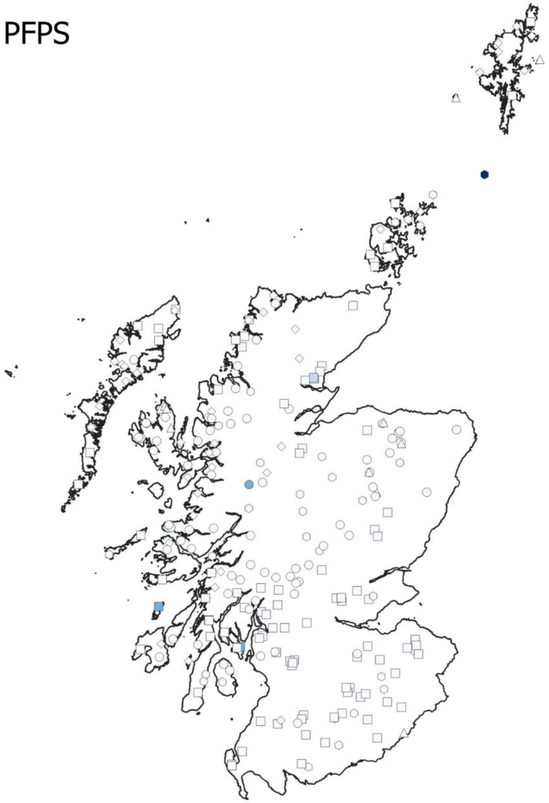
PFNA



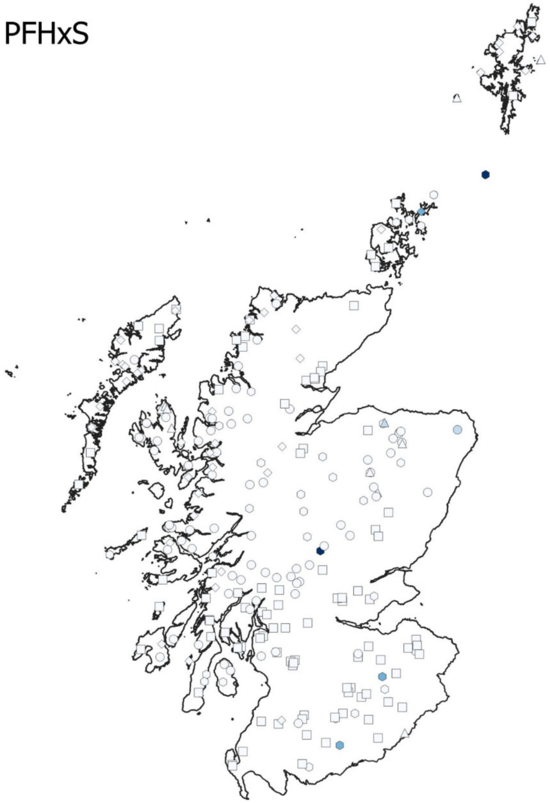
PFBS

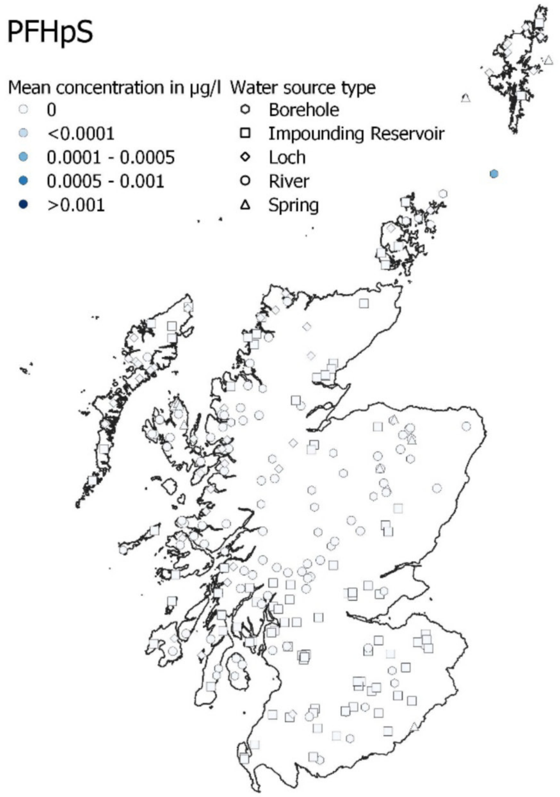


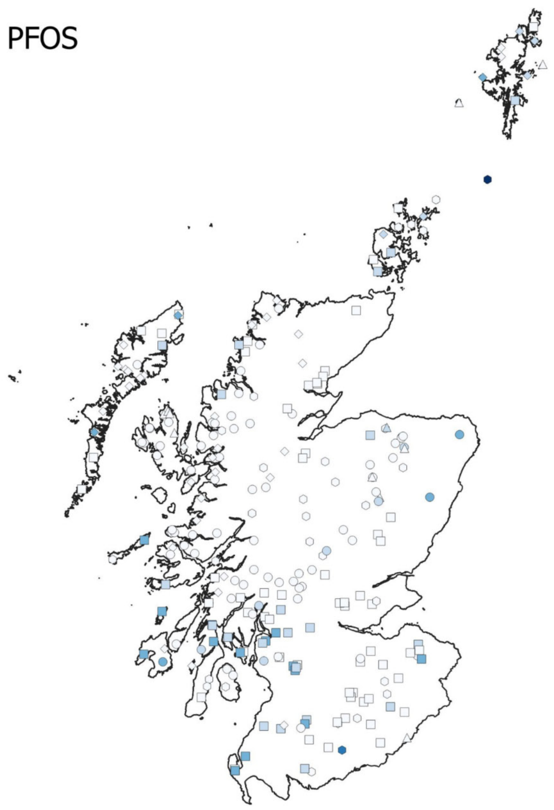
PFPS

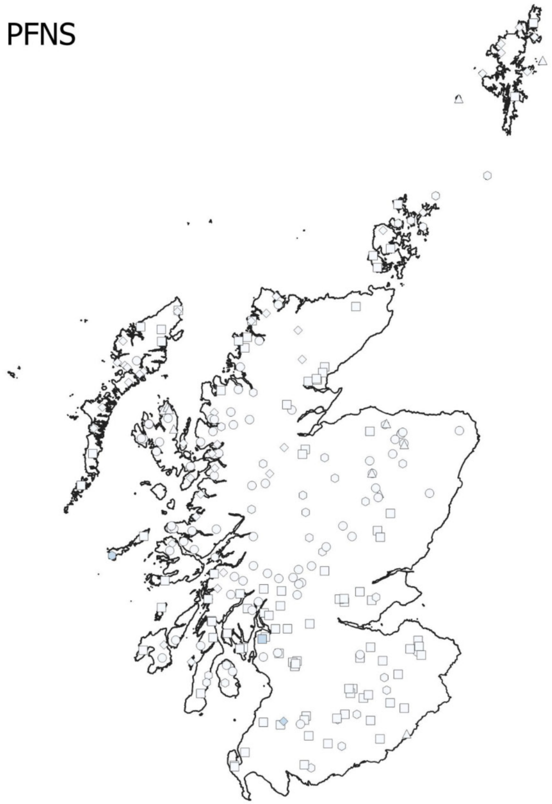


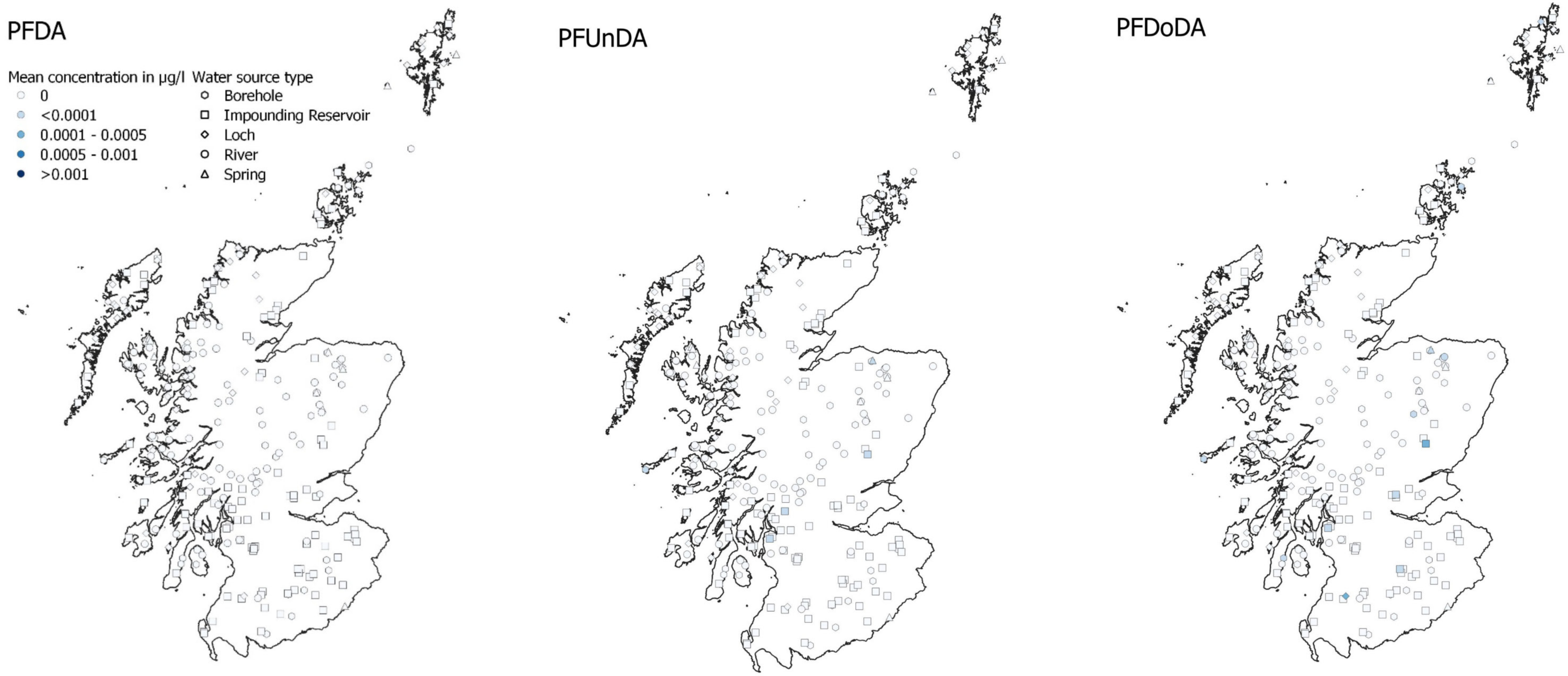
PFHxS

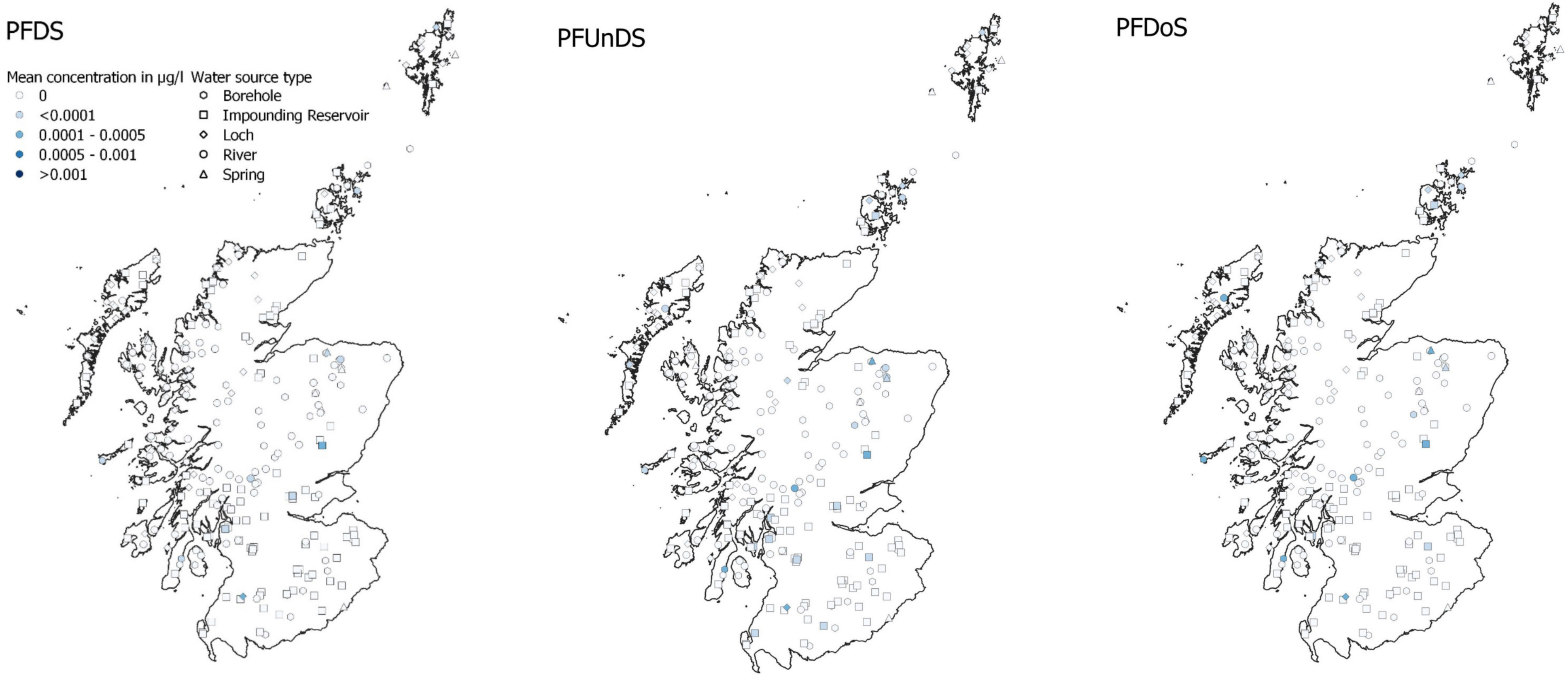



PFHpS
Mean concentration in µg/l
0
<0.0001
0.0001 - 0.0005
0.0005 - 0.001
>0.001
Water source type
Borehole
Impounding Reservoir
Loch
River
Spring


PFOS


PFNS


PFDA
Mean concentration in µg/l
○ 0
● <0.0001
● 0.0001 - 0.0005
● 0.0005 - 0.001
● >0.001
Water source type
○ Borehole
□ Impounding Reservoir
◇ Loch
○ River
△ Spring
PFUnDA
PFDoDA


PFDS
Mean concentration in µg/l
0
<0.0001
0.0001 - 0.0005
0.0005 - 0.001
>0.001
Water source type
Borehole
Impounding Reservoir
Loch
River
Spring
PFUnDS
PFDoS

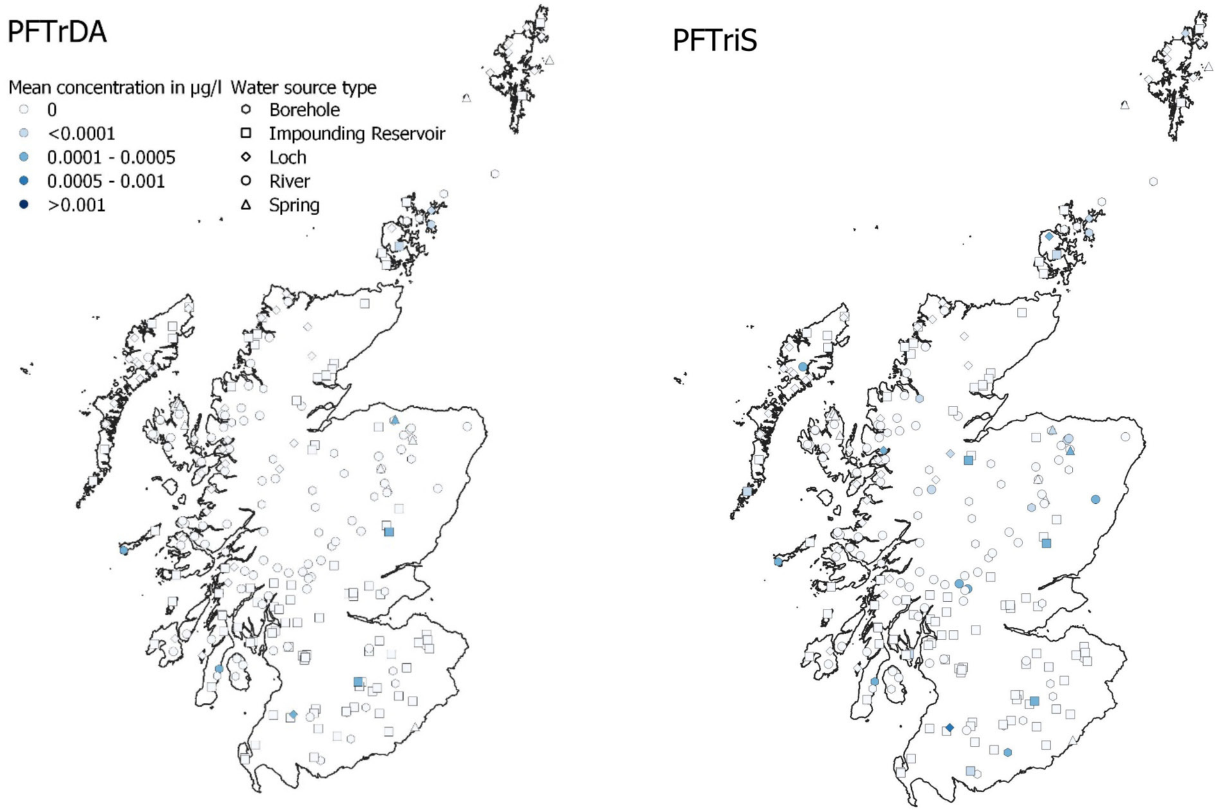


Figure A.1: Mean concentrations per PFAS substance in raw water sampling from 1 January 2023 to 8 July 2024 across Scottish drinking water supply catchments. For deriving means, <LoQ was set to 0 to facilitate mapping and comparison between PFAS substances with different LoQs. Values of 0 and close to 0 therefore reflect no or limited numbers of detections, rather than confirmed absence of the PFAs substance. Compare Table 4.1 for summary of the data.

Appendix B. PCAs

Table A.1: Variable coordinates for PCA on concentrations for each PFAS substance in individual samples where at least one PFAS substance was above the limit of quantification. *marks significant correlations. Positive and negative scores of >0.2 are highlighted.

PFAS	PC 1	PC 2	PC 3
Variation explained	*23.2%*	*20.2%*	*11.1%*
PFBS	-0.11645*	0.82669*	-0.19031*
PFBA	0.00216	0.12459*	0.53759*
PFDS	0.92369*	0.12806*	0.03499
PFDA	0.02991	0.02420	0.21138*
PFDoS	0.85971*	0.11378*	0.00020
PFDoDA	0.93456*	0.12886*	0.03104
PFHpS	-0.10652*	0.74182*	-0.17351*
PFHpA	-0.09708*	0.06701*	0.82423*
PFHxS	-0.13575*	0.93099*	-0.17976*
PFHxA	-0.09639*	0.43202*	0.44140*
PFNS	0.49191*	0.06729*	0.03235
PFNA	-0.04223	0.05087	0.61417*
PFOS	-0.13093*	0.83555*	-0.02445
PFOA	-0.14307*	0.37161*	0.67197*
PFPS	-0.13398*	0.89571*	-0.16012*
PFPA	-0.01519	-0.01410	0.22982*
PFTriS	0.57367*	0.09589	-0.00943
PFTrDA	0.77995*	0.10720*	0.00887
PFUnDS	0.74982*	0.09867*	0.02194
PFUnDA	0.57422*	0.08891	0.09674*

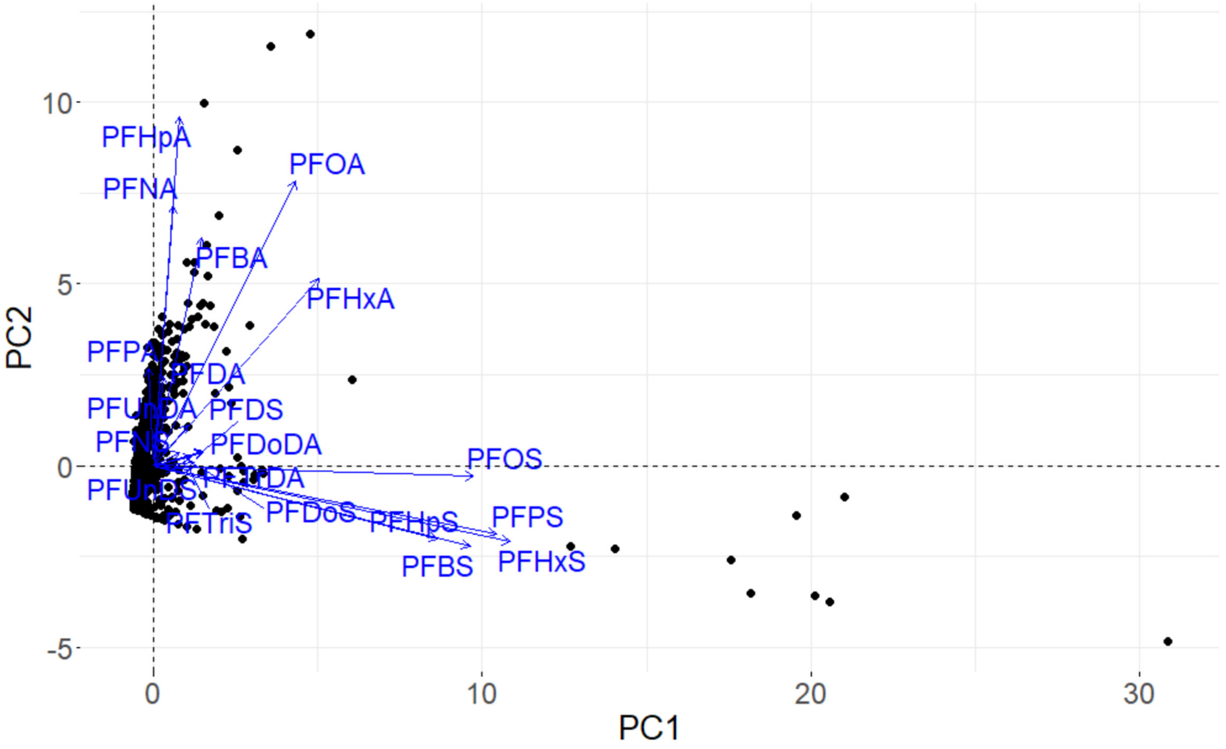


Figure A.2: Biplot of PC 2 and PC 3 of the PCA on concentrations for each PFAS substance in individual samples where at least one PFAS substance was above the limit of quantification (n=1003). Dots are catchments, symbolised by type of source, blue arrows are PFAS substance. Points/variables in the top associate positively with PC2, in the bottom – negatively. The longer the arrow/further from the plot origin the point, the stronger the association.

Table A.2: Variable coordinates for PCA mean concentration per PFAS substance per catchment. *marks significant correlations. Positive and negative scores of >0.2 are highlighted.

PFAS	PC 1	PC 2	PC 3
Variation explained	*30.7%*	*27%*	*14.6%*
PFBS	-0.04385	0.89711*	-0.19416*
PFBA	0.01179	0.24752*	0.72429*
PFDS	0.96316*	0.04380	0.00040
PFDA	0.00000	0.00000	0.00000
PFDoS	0.93321*	0.05155	0.02367
PFDoDA	0.94521*	0.04070	-0.00895
PFHpS	-0.06372	0.95091*	-0.23432*
PFHpA	-0.08756	0.14346*	0.84918*
PFHxS	-0.06141	0.95331*	-0.23921*
PFHxA	-0.02458	0.59999*	0.28059*
PFNS	0.81747*	0.04281	0.02071
PFNA	-0.05811	0.12472*	0.65594*
PFOS	-0.07027	0.97400*	-0.08923
PFOA	-0.00611	0.43043*	0.68675*
PFPS	-0.06407	0.94400	-0.20652*
PFPA	-0.09761	0.08571	0.58955*
PFTriS	0.74580*	0.09303	0.04782
PFTrDA	0.87034*	0.04191	-0.00434
PFUnDS	0.94962*	0.05840	0.06194
PFUnDA	0.46869*	0.01052	-0.01437

Table A.2: Variable coordinates for PCA mean concentration per PFAS substance per catchment. *marks significant correlations. Positive and negative scores of >0.2 are highlighted.

Supplementary variables	PC 1	PC 2	PC 3
	30.7%	*27%*	*14.6%*
Area	-0.01309	0.01374	-0.07427
Airports	-0.05478	0.64165*	-0.09263
Electrical Infrastructure	0.04279	0.02063	-0.01910
Fire stations	-0.01026	-0.01068	-0.05963
Industrial estates	0.02342	0.14081*	-0.01941
Landfills	-0.01148	-0.02043	-0.06313
Metal recyclers	0.01560	0.14250*	-0.03253
Ore Mines	-0.01214	-0.01662	-0.05469
Renewable energy	-0.03018	-0.03479	-0.06782
Ski infrastructure	-0.01342	-0.00789	-0.03136
Telecommunication infrastructure	-0.01954	-0.00273	-0.04592
Discharges to water	0.01905	0.14068*	-0.03862
PFOS wastewater releases to water	0.01812	0.13942*	-0.03777
Waste management discharges to water	-0.01532	-0.0044	-0.08848
Septic tanks	0.01482	0.093838	-0.11876
Roads	0.03441	0.02361	-0.10837
Biosolid application potential	-0.01486	0.03655	-0.07359
Paper waste discharge spreading potential	0.00809	0.05832	-0.06797
Arable area	-0.01522	0.03645	-0.07240
Area of atmospheric deposition from point sources	-0.00175	-0.02831	0.03576
Area of atmospheric deposition from arable areas	0.00144	-0.02521	-0.03469
Area of PFC releases to air	0.06927	-0.01120	-0.01681
2 km coastal zone	-0.03229	0.14344*	0.14758*
Mean slope	-0.02587	-0.17145*	-0.47919*
Mean annual precipitation 1991-2020	-0.05631	-0.16959*	-0.23833*
Area with groundwater vulnerability class 5	0.00277	0.01104	-0.14971*
Area with groundwater vulnerability class 4	-0.00297	-0.02690	0.06375
Area with topsoil total carbon content <5%	0.02063	0.06527	-0.12983*
Area with topsoil median pH >6	0.09365	0.04351	0.04187

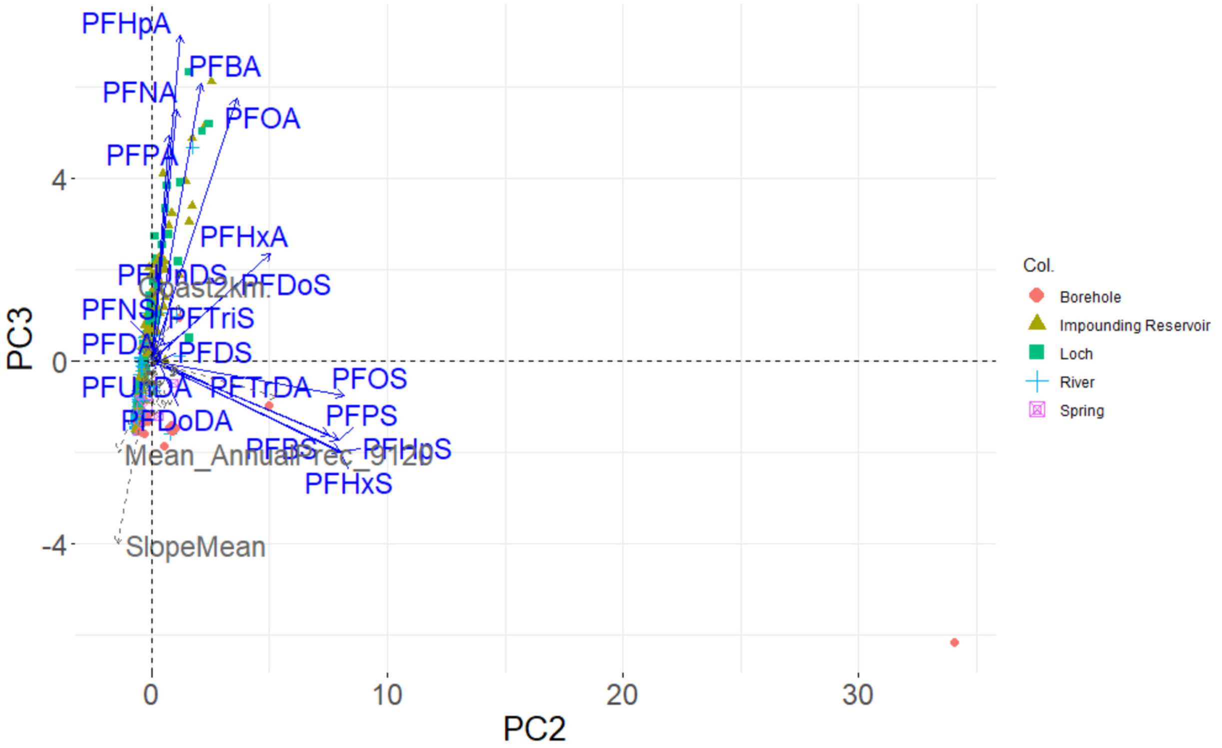


Figure A.3: Biplot of PC 2 and PC 3 from a PCA on mean concentration per PFAS substance per catchment (n=317), with source risk factors superimposed as supplementary variables. Dots are catchments, symbolised by type of source, blue arrows are PFAS substance, grey dashed arrows are risk factors. Points/variables in the right of the plot associate positively with PC1, to the left – negatively. Points/variables in the top associate positively with PC2, in the bottom – negatively. The longer the arrow/further from the plot origin the point, the stronger the association.

Table A.3: Variable coordinates for PCA on mean concentrations per PFAS substance per catchment with the outlier for comparatively high PFSA concentrations removed. *marks significant correlations. Positive and negative scores of >0.2 are highlighted.

PFAS	PC 1	PC 2	PC 3
Variation explained	*32.4%*	*18.8%*	*9.7%*
PFBS	0.03407	0.26627*	0.81932*
PFBA	0.02377	0.69401*	-0.27412*
PFDS	0.96383*	-0.01051	-0.05269
PFDA	0.00000	0.00000	0.00000
PFDoS	0.93446*	0.01530	-0.02879
PFDoDA	0.94579*	-0.01980	-0.05410
PFHpS	0.00000	0.00000	0.00000
PFHpA	-0.08314	0.81037*	-0.22664*
PFHxS	0.02943	0.06714	0.66090*
PFHxA	0.01563	0.53749*	0.27314*
PFNS	0.81811*	0.00955	-0.05803
PFNA	-0.05471	0.66982*	-0.14340*
PFOS	-0.01994	0.68738*	0.44068*
PFOA	0.01808	0.80234*	-0.10147
PFPS	-0.00656	0.17639*	-0.14194*
PFPA	-0.09516	0.50958*	-0.33322*
PFTriS	0.75060*	0.13194*	0.25726*
PFTrDA	0.87125*	-0.01275	-0.03341
PFUnDS	0.95108*	0.05000	-0.05375
PFUnDA	0.46851*	-0.02523	-0.06484

Table A.3: Variable coordinates for PCA on mean concentrations per PFAS substance per catchment with the outlier for comparatively high PFSA concentrations removed. *marks significant correlations. Positive and negative scores of >0.2 are highlighted.

Supplementary variables	PC 1	PC 2	PC 3
	32.4%	18.8%	9.7%
Area	-0.01062	-0.02126	0.20656*
Airports	-0.01604	0.07553	-0.02713
Electrical Infrastructure	0.04453	0.03868	0.17791*
Fire stations	-0.01050	-0.04484	0.05518
Industrial estates	0.03455	0.22969*	0.71497*
Landfills	-0.01254	-0.06074	0.01621
Metal recyclers	0.02748	0.21900*	0.74029*
Ore Mines	-0.01297	-0.05115	0.01817
Renewable energy	-0.03203	-0.07419	0.00294
Ski infrastructure	-0.01362	-0.02468	0.02927
Telecommunication infrastructure	-0.01926	-0.02653	0.07499
Discharges to water	0.03084	0.21349*	0.74287*
PFOS wastewater releases to water	0.02981	0.21159*	0.73504*
Waste management discharges to water	-0.01480	-0.04463	0.14235*
Septic tanks	0.02386	0.09990	0.68228*
Roads	0.03710	-0.06188	0.15254*
Biosolid application potential	-0.01098	0.02776	0.32875*
Paper waste discharge spreading potential	0.01323	0.07039	0.40750*
Arable area	-0.01136	0.02877	0.32795*
Area of atmospheric deposition from point sources	-0.00337	0.05750	0.08829
Area of atmospheric deposition from arable areas	0.00032	0.01702	0.14555*
Area of PFC releases to air	0.06835	-0.02451	-0.03361
2 km coastal zone	-0.02429	0.14387*	-0.15193*
Mean slope	-0.03507	-0.50533*	0.01470
Mean annual precipitation 1991-2020	-0.06704	-0.27893*	-0.04818
Area with groundwater vulnerability class 5	0.00347	-0.17508*	-0.12597*
Area with groundwater vulnerability class 4	-0.00438	0.08057	0.10563
Area with topsoil total carbon content <5%	0.02743	-0.03969	0.32872*
Area with topsoil median pH >6	0.09707	0.09813	0.18849*

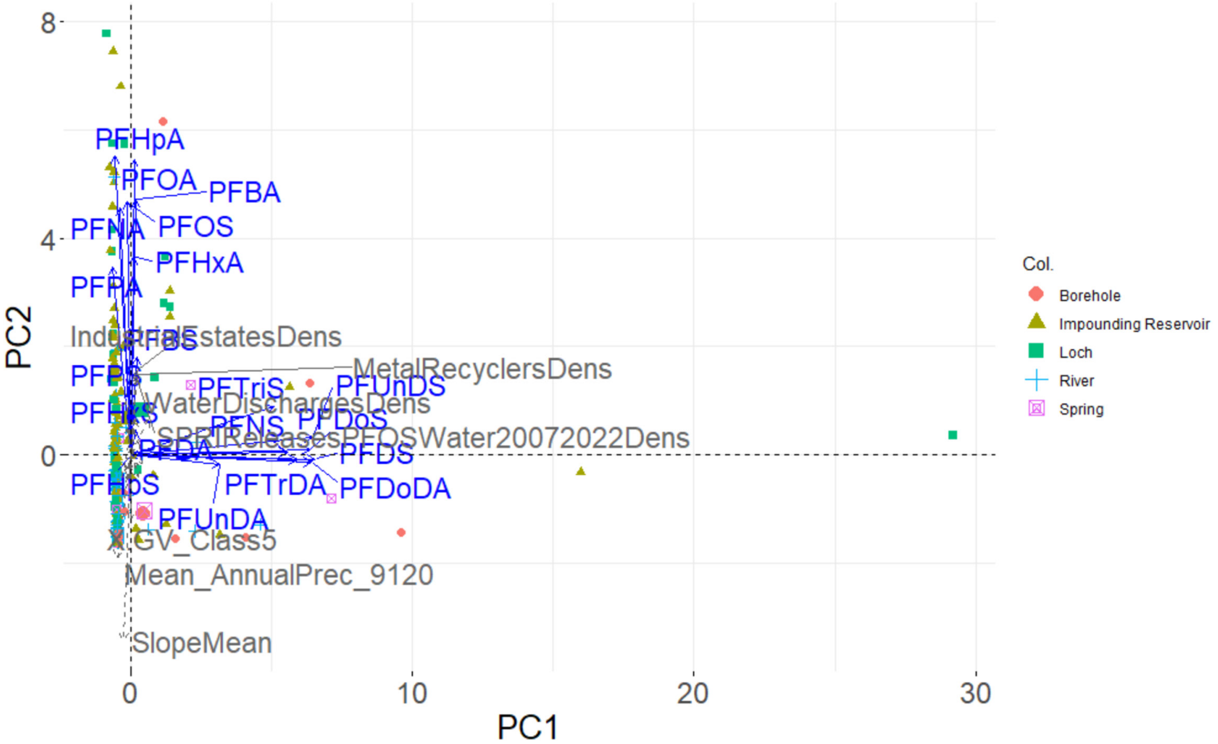


Figure A.4: Biplot of the first two PCs from a PCA on mean concentration per PFAS substance per catchment with the outlier for comparatively high concentrations of PFSAs removed (n=316), with source risk factors superimposed as supplementary variables. Dots are catchments, symbolised by type of source, blue arrows are PFAS substance, grey dashed arrows are risk factors. Points/variables in the right of the plot associate positively with PC1, to the left – negatively. Points/variables in the top associate positively with PC2, in the bottom – negatively. The longer the arrow/further from the plot origin the point, the stronger the association.

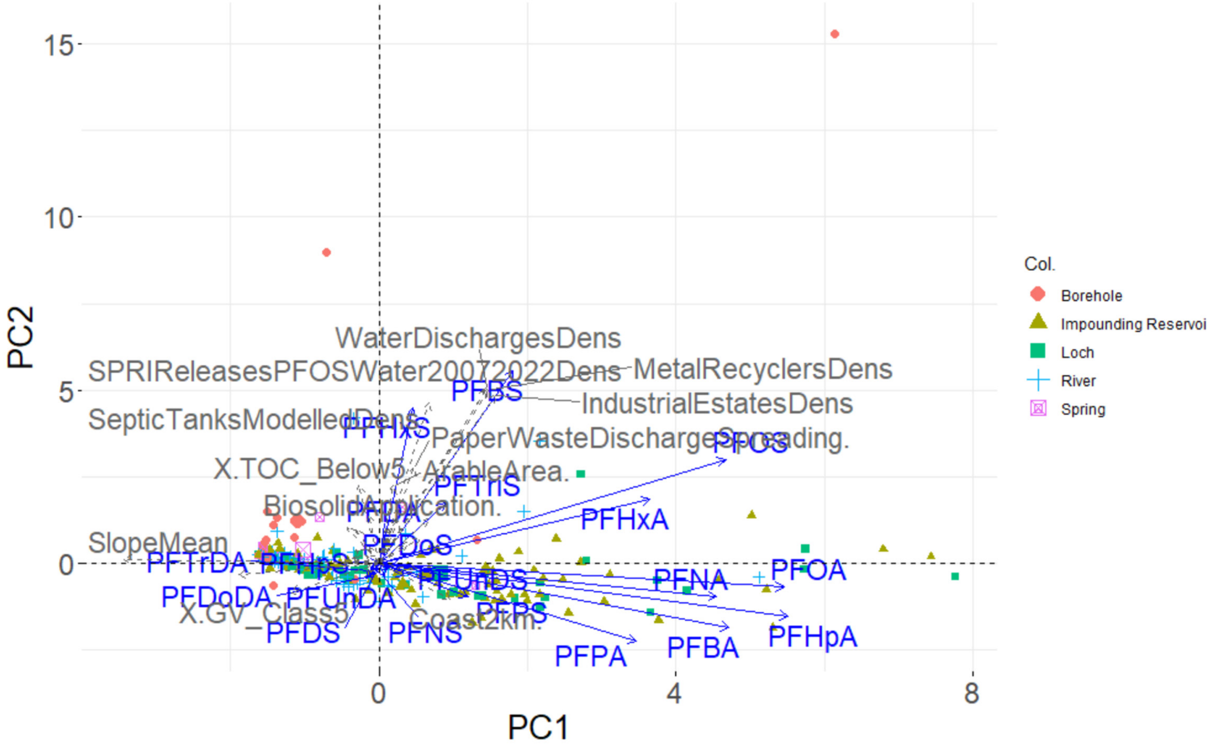


Figure A.5: Biplot of PC 2 and PC 3 from a PCA on mean concentration per PFAS substance per catchment with the outlier for comparatively high concentrations of PFSAs removed (n=316), with source risk factors superimposed as supplementary variables. Dots are catchments, symbolised by type of source, blue arrows are PFAS substance, grey dashed arrows are risk factors. Points/variables in the right of the plot associate positively with PC1, to the left – negatively. Points/variables in the top associate positively with PC2, in the bottom – negatively. The longer the arrow/further from the plot origin the point, the stronger the association.

Table A.4: Variable coordinates for PCA on percentages detection per PFAS substance per catchment. *marks significant correlations. Positive and negative scores of >0.2 are highlighted.

PFAS	PC 1	PC 2	PC 3
Variation explained	*26.2%*	*18.8%*	*12.9%*
PFBS	-0.04075	0.50877*	0.48803*
PFBA	-0.08585	0.61483*	-0.44898*
PFDS	0.91058*	0.02956	-0.01894
PFDA	0.02908	0.05809	0.15824*
PFDoS	0.87162*	0.05686	-0.01790
PFDoDA	0.89851*	0.02395	-0.01627
PFHpS	-0.03252	0.50255*	0.71978*
PFHpA	-0.12243	0.64404*	-0.57126*
PFHxS	0.00810	0.42413*	0.65167*
PFHxA	-0.04271	0.64490*	0.18735*
PFNS	0.81872*	0.07653	0.00046
PFNA	-0.06269	0.59853*	-0.46121*
PFOS	-0.08963	0.68791*	-0.06471
PFOA	-0.01883	0.71902*	-0.38596*
PFPS	-0.04691	0.51280*	0.63772*
PFPA	-0.11838	0.45996*	-0.19867*
PFTriS	0.70701*	0.11632	-0.00328
PFTrDA	0.88496*	0.04978	0.00665
PFUnDS	0.82441*	0.11118	-0.07277
PFUnDA	0.39705*	0.00933	0.00214

Table A.4: Variable coordinates for PCA on percentages detection per PFAS substance per catchment. *marks significant correlations. Positive and negative scores of >0.2 are highlighted.

Supplementary variables	PC 1	PC 2	PC 3
	26.2%	18.8%	12.9%
Area	-0.02349	0.00000	0.07851
Airports	-0.04398	0.38972*	0.44449*
Electrical Infrastructure	0.01954	0.00938	0.04777
Fire stations	-0.00987	-0.04561	0.05265
Industrial estates	-0.00491	0.21582*	0.19126*
Landfills	-0.01097	-0.06676	0.03728
Metal recyclers	0.00497	0.20989*	0.21319*
Ore Mines	-0.01133	-0.05616	0.03496
Renewable energy	-0.02755	-0.08008	0.01714
Ski infrastructure	-0.01484	-0.01926	0.02071
Telecommunication infrastructure	-0.02036	-0.05040	0.06498
Discharges to water	0.00740	0.20355*	0.21434*
PFOS wastewater releases to water	0.00778	0.20194*	0.21266*
Waste management discharges to water	-0.01788	-0.04921	0.07946
Septic tanks	0.00363	0.08659	0.25535*
Roads	0.04098	-0.06036	0.14336*
Biosolid application potential	-0.02545	0.03754	0.16569*
Paper waste discharge spreading potential	-0.00767	0.05718	0.13742*
Arable area	-0.02619	0.03805	0.16427*
Area of atmospheric deposition from point sources	-0.01850	0.02187	0.00300
Area of atmospheric deposition from arable areas	0.00312	0.00627	0.08053
Area of PFC releases to air	0.06159	-0.01826	-0.00990
2 km coastal zone	-0.02027	0.15989*	-0.05011
Mean slope	0.00197	-0.46130*	0.21522*
Mean annual precipitation 1991-2020	-0.04869	-0.27087*	-0.01238
Area with groundwater vulnerability class 5	0.02543	-0.10283	0.03128
Area with groundwater vulnerability class 4	-0.02026	0.01910	0.02011
Area with topsoil total carbon content <5%	0.01198	-0.03460	0.26955*
Area with topsoil median pH >6	0.11251	0.06506	0.07428

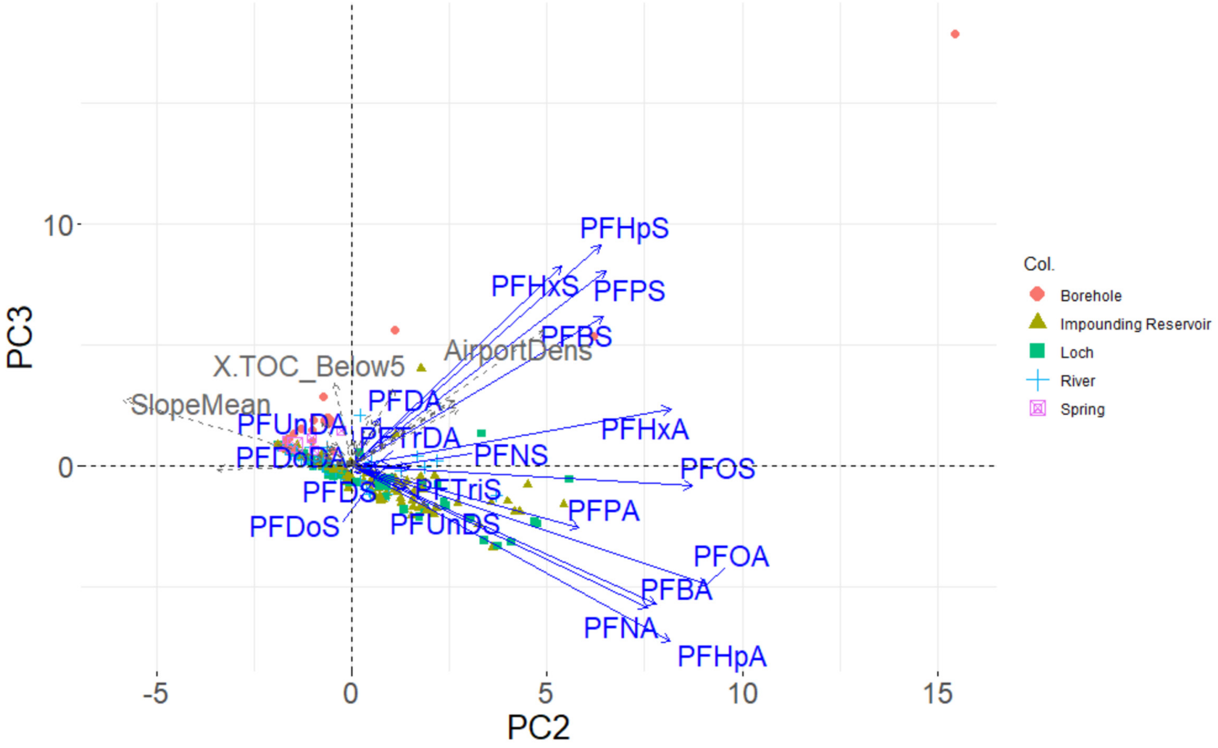


Figure A.6: Biplot of PC 2 and PC 3 from a PCA on percentage detections per PFAS substance per catchment (n=317), with source risk factors superimposed as supplementary variables. Dots are catchments, symbolised by type of source, blue arrows are PFAS substance, grey dashed arrows are risk factors. Points/variables in the right of the plot associate positively with PC1, to the left – negatively. Points/variables in the top associate positively with PC2, in the bottom – negatively. The longer the arrow/further from the plot origin the point, the stronger the association.

Appendix C. RWPP PFAS subgroup assessments

Table A.5: Weighting applied to risk factors in PFAS subgroup assessments – risk score for the particular risk factor (normalised density or percentage) multiplied as below. S/M-C PFCAs = Short and medium chain (4-9 carbons) PFCAs; S/M-C PFSAs = Short and medium chain PFSAs; L-C PFAS = Long chain (10-13 carbons) PFAS. No pathway = Risk factors outwith the identified pathway condition facilitating transport to water source; GV = Class 5 of the BGS Hydrogeological Maps of Scotland, groundwater vulnerability layer, used for groundwater supply sources; Soil pH = Topsoil with pH of 6 or above, JHI Topsoil pH map, used for surface water supply sources in the long chain PFAS assessment; OC = Topsoil with organic carbon content of below 5%, JHI Topsoil Organic carbon content map, used for surface water supply sources on the short and medium chain PFAS assessments.

Risk factor	Pathway	Weighting			Comment
		S/M-C PFCAs	S/M-C PFSAs	L-C PFAS	
Airports	No pathway	2	3	1	Airports have been reported as major sources of PFAS pollution, especially short to medium chain PFAAs, and there's usually an association with higher proportions of PFSAs, especially PFOS and PFHxS
	GV	3	4	2	
	Soil pH/OC	3	4	2	
Electrical Infrastructure	No pathway	1	1	1	No new evidence to underpin this as a source, and no evidence that this has a major impact locally, so given less weight.
	GV				
	Soil pH/OC				
Fire Stations	No pathway	2	3	1	Same rationale as airports due to potential impact of AFFF storage.
	GV	3	4	2	
	Soil pH/OC	3	4	2	
Industrial Estates	No pathway	1	2	2	Literature shows higher proportions of longer chain PFAAs. Conflicting literature on importance of PFSAs, but indications that PFSA precursors are released from industrial sites. Industrial estates are also a proxy for potential use of AFFF.
	GV	2	3	3	
	Soil pH/OC	2	3	3	
Landfills	No pathway	2	1	1	No pathway used as landfills usually bypass superficial layers. Landfills usually associated with short chain PFCAs.
	GV				
	Soil pH/OC				
Metal Recyclers	No pathway	1	2	1	Metal recyclers correlated a little to the PFSA grouping in the PCA - chrome plating is usually associated with higher proportions of PFSAs.
	GV	2	3	2	
	Soil pH/OC	2	3	2	
Renewable Energy Infrastructure	No pathway	1	1	1	As electrical infrastructure
	GV				
	Soil pH/OC				
Ski Infrastructure	No pathway	1	1	1	No occurrence on areas with median topsoil pH above 6
	GV	2	2	2	
	Soil pH/OC	2	2	n/a	
Telecomunication Infrastructure	No pathway	1	1	1	As electrical infrastructure
	GV				
	Soil pH/OC				

Table A.5: Weighting applied to risk factors in PFAS subgroup assessments – risk score for the particular risk factor (normalised density or percentage) multiplied as below. S/M-C PFCAs = Short and medium chain (4-9 carbons) PFCAs; S/M-C PFSAs = Short and medium chain PFSAs; L-C PFAS = Long chain (10-13 carbons) PFAS. No pathway = Risk factors outwith the identified pathway condition facilitating transport to water source; GV = Class 5 of the BGS Hydrogeological Maps of Scotland, groundwater vulnerability layer, used for groundwater supply sources; Soil pH = Topsoil with pH of 6 or above, JHI Topsoil pH map, used for surface water supply sources in the long chain PFAS assessment; OC = Topsoil with organic carbon content of below 5%, JHI Topsoil Organic carbon content map, used for surface water supply sources on the short and medium chain PFAS assessments.

Risk factor	Pathway	Weighting			Comment
		S/M-C PFCAs	S/M-C PFSAs	L-C PFAS	
Discharges to water	No pathway	2	2	2	Direct route to water
	GV				
	Soil pH/OC				
Registered PFOS releases to water	No pathway	1	2	1	These are identical to some of the Water discharges, effectively counting known sites that release PFOS twice, giving them more weight.
	GV				
	Soil pH/OC				
Discharges to water from waste management	No pathway	2	1	1	Direct route to water. See landfills
	GV				
	Soil pH/OC				
Septic Tanks (modelled)	No pathway	2	1	1	See landfills. Shallower depths, so pathways included.
	GV	3	2	2	
	Soil pH/OC	3	2	2	
Roads	No pathway	1	1	1	Roads could be a proxy for a number of risk factors, including potential use of AFFF, higher population density, or atmospheric pollution.
	GV	2	2	2	
	Soil pH/OC	2	2	2	
Biosolid application potential	No pathway	2	1	1	Wastewater product Soil pH is not used in combination with biosolid application potential as soil pH was used in the creation of the layer.
	GV	3	2	2	
	Soil pH/OC	3	2	n/a	
Paper waste discharge spreading potential	No pathway	1	1		Literature indicating that this has more relevance for shorter chain PFCAs and PFOS.
	GV	2	2		
	Soil pH/OC	2	2		
Arable area	No pathway	1	1	1	Pesticides are known to include PFAS, although substances and impact to freshwater unclear.
	GV	2	2	2	
	Soil pH/OC	2	2	2	

Table A.5: Weighting applied to risk factors in PFAS subgroup assessments – risk score for the particular risk factor (normalised density or percentage) multiplied as below. S/M-C PFCAs = Short and medium chain (4-9 carbons) PFCAs; S/M-C PFSAs = Short and medium chain PFSAs; L-C PFAS = Long chain (10-13 carbons) PFAS. No pathway = Risk factors outwith the identified pathway condition facilitating transport to water source; GV = Class 5 of the BGS Hydrogeological Maps of Scotland, groundwater vulnerability layer, used for groundwater supply sources; Soil pH = Topsoil with pH of 6 or above, JHI Topsoil pH map, used for surface water supply sources in the long chain PFAS assessment; OC = Topsoil with organic carbon content of below 5%, JHI Topsoil Organic carbon content map, used for surface water supply sources on the short and medium chain PFAS assessments.

Risk factor	Pathway	Weighting			Comment
		S/M-C PFCAs	S/M-C PFSAs	L-C PFAS	
Area of atmospheric deposition around point sources (5 km)	No pathway	1	1	1	Atmospheric sources weighted by topography before, but this was removed in the update.
	GV	2	2	2	
	Soil pH/OC	2	2	2	
Registered releases of PFCs to air (5 km)	No pathway	1	1	1	These are known sites releasing perfluorocarbons (5 sites). Not always matching points used for the atmospheric deposition layer, but area overlaps the above layer, effectively double counting these sites.
	GV	2	2	2	
	Soil pH/OC	2	2	2	
Area of atmospheric deposition around arable agricultural areas (5 km)	No pathway	1	1	1	
	GV	2	2	2	
	Soil pH/OC	2	2	2	
Sea spray aerosol deposition	No pathway	2	2	2	Assumed to be a locally important factor
	GV	3	3	3	
	Soil pH/OC	3	3	3	
Mean annual precipitation	Mean slope				Used as a proxy for long range atmospheric transport and wet deposition. Removed for this update as statistical analysis indicated a reverse effect.
Groundwater vulnerability class 4	*Ground-water sources*	3			Used in the short and medium chain PFCAs assessment as an additional risk factor for groundwater sources, reflecting better mobility of these PFAS

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