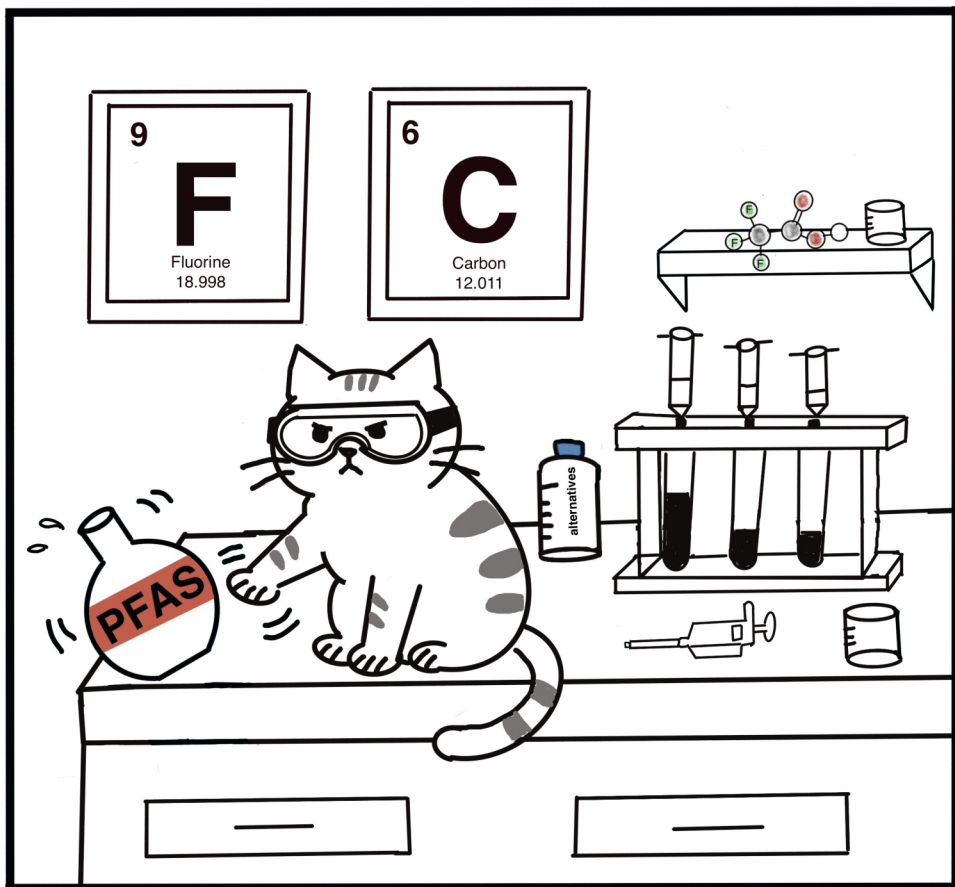


Tracking fluorinated chemicals and identifying alternatives in contemporary consumer products and technologies

Eleni Savvidou



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Abstract

Per- and polyfluoroalkyl substances (PFAS) are a large group of synthetic chemicals with unique physicochemical properties that have led to their widespread use, ranging from everyday consumer products to critical technologies. Their extreme persistence, combined with other hazardous properties, raises significant concerns for human health and the environment. Limited understanding of PFAS uses across different applications, together with gaps in analytical capabilities, hampers progress towards their phase out and replacement with safer alternatives.

In this work, analytical investigations were combined with a search of potential PFAS alternatives. Fluorine-based analytical approaches were applied to assess PFAS use across different product categories (**Papers I and II**), and pyrolysis-gas chromatography-mass spectrometry (Py-GC-MS) was explored to address analytical gaps related to polymeric PFAS (**Paper II**). Emissions of fluorinated compounds associated with the lifecycle of lithium-ion batteries (LIBs) were investigated to evaluate their relevance as a contamination source in Europe (**Paper III**). Finally, alternative chemistries to PFAS were explored for applications in stone sealers (**Paper I**) and LIBs (**Paper IV**).

In **Paper I**, total fluorine (TF) analysis of commercial stone sealers revealed that 81% contained fluorine, with concentrations reaching up to 27 150 $\mu\text{g F/g}$, most likely originating from polymeric PFAS. Extractable organofluorine (EOF) analysis confirmed the predominantly organic nature of the fluorine, and targeted analysis identified polyfluoroalkyl phosphate esters (PAPs) as the dominant PFAS class. Six products were presumably identified as not containing intentionally added PFAS, instead relying on organosilicon-based chemistries, some of which are currently under regulatory scrutiny.

Paper II investigated the application of Py-GC-MS following initial TF screening of cookware (up to 550 000 $\mu\text{g F/g}$), textiles (up to 1 600 $\mu\text{g F/g}$), electronics (up to 2 100 $\mu\text{g F/g}$) and personal care products (up to 630 000 $\mu\text{g F/g}$). The method demonstrated the ability to detect polytetrafluoroethylene (PTFE) in unknown samples down to 0.1-0.2 wt% and to differentiate between different side-chain fluorinated polymers commonly used in textile applications.

In **Paper III**, targeted analysis of electrolyte-related anions used in LIBs, such as bis(trifluoromethanesulfonyl)imide (TFSI⁻), hexafluorophosphate (PF₆⁻) and tetrafluoroborate (BF₄⁻), revealed their widespread occurrence in water samples, with varying regional profiles. BF₄⁻ dominated the contamination profile of the Danube in Hungary, while temporal sampling of the River Erpe in Germany showed elevated TFSI⁻ and PF₆⁻ concentrations potentially connected to industrial activities, with marked decreases observed during a temporary plant shutdown. In Sweden, recycling and landfill sites were identified as major contributors to environmental TFSI⁻ and PF₆⁻, with concentrations reaching up to 300 ng/L for TFSI⁻ and 39 000 ng/L for PF₆⁻, primarily in landfill-related samples.

Paper IV examined the availability of PFAS alternatives for critical LIB components, specifically cathodes and electrolytes, through a literature review and interviews with experts from academia and industry. The results indicate that while PFAS-free LIBs appear to be technically feasible, such solutions are not yet widely established and require further evaluation with respect to performance, durability, and economic viability. Nevertheless, the outlook is rather promising, as several companies have already introduced PFAS-free solutions or are actively developing them.

Overall, the combination of fluorine mass balance approaches and Py-GC-MS proved effective for screening consumer products for PFAS and for supporting regulatory enforcement. The results further highlight substitution, guided by robust analytical data and lifecycle considerations, as the most effective long-term strategy to reduce PFAS emissions. However, alternatives require careful evaluation to avoid regrettable substitution, particularly in critical technologies such as LIBs.

Keywords: PFAS, fluorinated polymers, fluorine mass balance, stone sealers, combustion ion chromatography, pyrolysis-gas chromatography-mass spectrometry, lithium-ion batteries, emissions, PFAS alternatives.

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Table of contents

Abstract.....	ii
Sammanfattning.....	iii
Zusammenfassung	v
Επιστημονική περίληψη για το κοινό.....	vii
List of publications	viii
Abbreviations.....	x
Prologue.....	1
1. Introduction	2
1.1 The class of PFAS	2
1.2 The early days of fluorine chemistry and the first use applications	3
1.3 PFAS in today's world	5
1.4 The challenge of analysing PFAS	6
1.5 From detection to substitution: searching for alternatives	8
2. Aim and scope.....	9
3. Methods.....	10
3.1 Sampling and data acquisition	10
3.2 Sample preparation.....	10
3.3 Instrumental analysis	11
3.3.1 TF and EOF analysis by CIC in Paper I-II	11
3.3.2. Py-GC-MS analysis in Paper II	11
3.3.3. LC-MS analysis in Paper I and III	11
3.4 Identification of potential alternatives to PFAS in Paper I and IV.....	12
4. Results and Discussion	14
4.1 Analysis of fluorinated compounds across diverse product categories.....	14
4.1.1 Fluorine mass balance of sealers (Paper I)	14
4.1.2 Py-GC-MS as tool for detection of C-F bonds (Paper II)	14
4.2 Environmental occurrence of emerging PFAS from the lifecycle of LIBs (Paper III)	15
4.3 Exploring non-PFAS options for sealers and LIBs	16
4.3.1 Silicones as alternatives to PFAS in sealers (Paper I).....	16
4.3.2 Alternatives to fluorinated components in LIBs (Paper IV)	17
5. Conclusion and future outlook	18
Acknowledgements.....	21
References	22

Abstract

Per- and polyfluoroalkyl substances (PFAS) are a large group of synthetic chemicals with unique physicochemical properties that have led to their widespread use, ranging from everyday consumer products to critical technologies. Their extreme persistence, combined with other hazardous properties, raises significant concerns for human health and the environment. Limited understanding of PFAS uses across different applications, together with gaps in analytical capabilities, hampers progress towards their phase out and replacement with safer alternatives.

In this work, analytical investigations were combined with a search of potential PFAS alternatives. Fluorine-based analytical approaches were applied to assess PFAS use across different product categories (**Papers I and II**), and pyrolysis-gas chromatography-mass spectrometry (Py-GC-MS) was explored to address analytical gaps related to polymeric PFAS (**Paper II**). Emissions of fluorinated compounds associated with the lifecycle of lithium-ion batteries (LIBs) were investigated to evaluate their relevance as a contamination source in Europe (**Paper III**). Finally, alternative chemistries to PFAS were explored for applications in stone sealers (**Paper I**) and LIBs (**Paper IV**).

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In **Paper III**, targeted analysis of electrolyte-related anions used in LIBs, such as bis(trifluoromethanesulfonyl)imide (TFSI⁻), hexafluorophosphate (PF₆⁻) and tetrafluoroborate (BF₄⁻), revealed their widespread occurrence in water samples, with varying regional profiles. BF₄⁻ dominated the contamination profile of the Danube in Hungary, while temporal sampling of the River Erpe in Germany showed elevated TFSI⁻ and PF₆⁻ concentrations potentially connected to industrial activities, with marked decreases observed during a temporary plant shutdown. In Sweden, recycling and landfill sites were identified as major contributors to environmental TFSI⁻ and PF₆⁻, with concentrations reaching up to 300 ng/L for TFSI⁻ and 39 000 ng/L for PF₆⁻, primarily in landfill-related samples.

Paper IV examined the availability of PFAS alternatives for critical LIB components, specifically cathodes and electrolytes, through a literature review and interviews with experts from academia and industry. The results indicate that while PFAS-free LIBs appear to be technically feasible, such solutions are not yet widely established and require further evaluation with respect to performance, durability, and economic viability. Nevertheless, the outlook is rather promising, as several companies have already introduced PFAS-free solutions or are actively developing them.

Overall, the combination of fluorine mass balance approaches and Py-GC-MS proved effective for screening consumer products for PFAS and for supporting regulatory enforcement. The results further highlight substitution, guided by robust analytical data and lifecycle considerations, as the most effective long-term strategy to reduce PFAS emissions. However, alternatives require careful evaluation to avoid regrettable substitution, particularly in critical technologies such as LIBs.

Sammanfattning

Per- och polyfluorerade alkylsubstanser (PFAS) är en stor grupp av syntetiska kemikalier med unika fysikalisk-kemiska egenskaper vilket har lett till deras omfattande användning, från vardagliga konsumentprodukter till kritiska teknologier. Deras extrema persistens, i kombination med andra farliga egenskaper, bedöms kunna utgöra en risk för människors hälsa och miljön. En begränsad förståelse av PFAS-användning inom olika tillämpningar, tillsammans med brister i analytiska metoder, försvårar arbetet med att fasa ut PFAS och ersätta dem med säkrare alternativ.

I detta arbete kombinerades analytiska undersökningar med en kartläggning av potentiella PFAS-alternativ. Fluoroberade analytiska metoder utgjorde grunden till strategin av att bättre förstå omfattningen av PFAS-användning i olika produktkategorier (**Artikel I och II**). Pyrolys-gaskromatografimasspektrometri (Py-GC-MS) utforskades som en kompletterande teknik för att täcka analytiska kunskapsluckor relaterade till polymera PFAS (**Artikel II**). Vidare undersöktes utsläpp av fluorerade föreningar kopplade till litiumjonbatteriers livscykel för att bedöma batteriers betydelse som föroreningskälla i Europa (**Artikel III**). Slutligen studerades alternativ till PFAS för tillämpningar i impregneringar för stenmaterial (**Artikel I**) och LIB (**Artikel IV**).

I **Artikel I** visade analysen av totalt fluor i kommersiella impregneringar för stenmaterial att 81% av produkterna innehöll fluor, med koncentrationer upp till 27 150 µg F/g, sannolikt huvudsakligen från polymera PFAS. Analys av andelen extraherat organiskt fluor (extractable organofluorine) bekräftade fluorens huvudsakligen organiska karaktär och riktad analys (target analysis) identifierade polyfluorerade alkylfosfater-substanter (PAPs) som den dominerande PFAS-klassen. Sex produkter identifierades dock som troligen fria från avsiktligt tillsatta PFAS och baserades istället på organosilicium-kemi, varav vissa för närvarande är föremål för regulatorisk granskning.

I **Artikel II** undersöktes användningen av Py-GC-MS efter inledande totalt fluor-screening av köksredskap (upp till 550 000 µg F/g), textilier (1 600 µg F/g), elektronik (2 100 µg F/g) och personliga hygienprodukter (upp till 630 000 µg F/g). Metoden visade förmåga att detektera polytetrafluoreten (PTFE) i okända prover ner till 0.1–0.2 viktprocent samt särskilja mellan olika fluorerade sidokedjespolymerer (side-chain fluorinated polymers) som ofta används i textila tillämpningar.

I **Artikel III** visade riktad analys av elektrolytrelaterade anjoner i litiumjonbatterier, såsom bis(trifluormetansulfonyl)imid (TFSI⁻), hexafluorofosfat (PF₆⁻) och tetrafluorborat (BF₄⁻), deras utbredda förekomst i vattenprover med regionalt varierande belastningsprofiler. I Ungern dominerade BF₄⁻ föroreningsprofilen i Donau, medan tidsupplöst provtagning i Erpe i Tyskland visade förhöjda halter av TFSI⁻ och PF₆⁻ som potentiellt kunde kopplas till industriell verksamhet, med minskningar under en tillfällig produktionsnedstängning. I Sverige identifierades återvinnings- och deponiområden som betydande källor till TFSI⁻ och PF₆⁻, med koncentrationer upp till 300 ng/L (TFSI⁻) och 39 000 ng/L (PF₆⁻), främst i deponirelaterade prover.

I **Artikel IV** undersöktes tillgängligheten av PFAS-alternativ för kritiska litiumjonbatterier-komponenter, särskilt katoder och elektrolyter, genom litteraturstudier och intervjuer med experter från akademi och industri. Resultaten indikerar att PFAS-fria litiumjonbatterier är tekniskt genomförbara, men att sådana lösningar ännu inte är brett etablerade och kräver vidare utvärdering av prestanda, livslängd och ekonomisk bärkraft. Trots detta är framtidsutsikterna lovande, då flera företag redan har introducerat PFAS-fria lösningar eller aktivt utvecklar dem.

Sammantaget visade sig kombinationen av fluoroberade massbalansmetoder och Py-GC-MS vara effektiv för screening av konsumentprodukter med avseende på PFAS och för att stötta regulatorisk tillsyn. Resultaten understryker dock att substitution, guidad av robusta analytiska data och livscykelperspektiv,

förblir den mest effektiva långsiktiga strategin för att minska PFAS-utsläpp. Alternativ måste utvärderas noggrant för att undvika så kallade "beklagliga substitutioner" (regrettable substitution). Detta är särskilt viktigt för kritiska teknologier som litiumjonbatterier.

Zusammenfassung

Per- und polyfluorierte Alkylsubstanzen (PFAS) sind eine große Gruppe synthetischer Chemikalien mit einzigartigen physikochemischen Eigenschaften, die zu ihrer weitverbreiteten Verwendung geführt haben, von alltäglichen Produkten für Konsumenten bis hin zu kritischen Technologien. Ihre extreme Persistenz, kombiniert mit meist anderen gefährlichen Eigenschaften, gibt Anlass zu erheblichen Bedenken hinsichtlich der menschlichen Gesundheit und der Umwelt. Ein begrenztes Verständnis der Anwendungen von PFAS in verschiedenen Applikationen sowie Lücken in analytischen Methoden erschweren den Fortschritt beim gezielten Ersetzen von PFAS durch sicherere Alternativen.

In dieser Arbeit wurden analytische Untersuchungen mit der Suche nach potenziellen PFAS-Alternativen kombiniert. Fluorbasierte analytische Ansätze bildeten die Grundlage, um das Ausmaß der PFAS-Verwendung in unterschiedlichen Produktkategorien besser zu verstehen (**Publikationen I und II**). Die Pyrolyse-Gaschromatographie-Massenspektrometrie (Py-GC-MS) wurde als ergänzende Technik eingesetzt, um analytische Lücken im Zusammenhang mit polymeren PFAS zu schließen (**Publikation II**). Darüber hinaus wurden Emissionen fluorierter Verbindungen entlang des Lebenszyklus von Lithium-Ionen-Batterien untersucht, um die Relevanz von Batterien als Kontaminationsquelle in Europa abzuschätzen (**Publikation III**). Schließlich wurden PFAS-Alternativen für Anwendungen in Steinversiegelungen (**Publikation I**) und LIBs (**Publikation IV**) erforscht.

In **Publikation I** zeigte die Analyse des Gesamtfluors in kommerziellen Steinversiegelungen, dass 81% der Produkte Fluor enthielt, mit Konzentrationen von bis zu 27 150 µg F/g, die vermutlich hauptsächlich aus polymeren PFAS stammen. Die Analyse des extrahierbaren organischen Fluors bestätigte die überwiegend organische Natur des Fluors und eine zielgerichtete Analyse identifizierte polyfluorierte Alkylphosphatester (PAPs) als dominierende PFAS-Klasse. Sechs Produkte wurden jedoch vermutlich als frei von absichtlich zugesetzten PFAS eingestuft, die stattdessen auf Organosilicium-Chemien basierten, von denen einige derzeit unter regulatorischer Beobachtung stehen.

Publikation II untersuchte den Einsatz von Py-GC-MS nach initialem Gesamt-Fluor-Screening von Kochgeschirr (bis zu 550 000 µg F/g), Textilien (bis zu 1 600 µg F/g), Elektronik (bis zu 2 100 µg F/g) und Körperpflegeprodukten (bis zu 630 000 µg F/g). Die Methode zeigte die Fähigkeit, Polytetrafluorethylen (PTFE) in unbekanntenen Proben bis zu einem Gehalt von 0.1–0.2 Gewichtsprozent nachzuweisen, sowie zwischen verschiedenen seitengruppen-fluorierten Polymeren zu unterscheiden, die häufig in Textilanwendungen eingesetzt werden.

In **Publikation III** zeigte die zielgerichtete Analyse von elektrolytbezogenen Anionen in Lithium-Ionen-Batterien, darunter Bis(trifluormethansulfonyl)imid (TFSI⁻), Hexafluorophosphat (PF₆⁻) und Tetrafluoroborat (BF₄⁻), deren weite Verbreitung in wässrigen Umweltproben mit regional unterschiedlichen Belastungsprofilen. In Ungarn dominierte BF₄⁻ das Kontaminationsprofil der Donau, während zeitlich aufgelöste Probenahmen an der Erpe in Deutschland erhöhte TFSI⁻ und PF₆⁻ Konzentrationen zeigten, die potenziell mit industriellen Aktivitäten in Verbindung stehen und während einer temporären Betriebsschließung zurückgingen. In Schweden wurden Recycling- und Deponiestandorte als wesentliche Quellen für TFSI⁻ und PF₆⁻ in der Umwelt identifiziert, mit Konzentrationen von bis zu 300 ng/L (TFSI⁻) und 39 000 ng/L (PF₆⁻), überwiegend in deponiebezogenen Proben.

Publikation IV untersuchte anhand einer Literaturrecherche und Experteninterviews aus Wissenschaft und Industrie, die Verfügbarkeit von PFAS-Alternativen für zentrale LIB-Komponenten, insbesondere Kathoden und Elektrolyte. Die Ergebnisse zeigen, dass PFAS-freie Lithium-Ionen-Batterien technisch realisierbar erscheinen, solche Lösungen jedoch bislang nicht breit etabliert sind und weitere Evaluierungen hinsichtlich Leistungsfähigkeit, Lebensdauer und wirtschaftlicher Tragfähigkeit notwendig

sind. Dennoch ist der Ausblick vielversprechend, da mehrere Unternehmen bereits PFAS-freie Lösungen eingeführt haben oder aktiv an deren Entwicklung arbeiten.

Insgesamt erwies sich die Kombination aus Fluor-Massenbilanz-Ansätzen und Py-GC-MS als wirksam für das Screening von Verbraucherprodukten auf PFAS und zur Unterstützung regulatorischer Maßnahmen. Die Ergebnisse zeigen jedoch auch, dass Substitution, gestützt auf robuste analytische Daten und Lebenszyklusbetrachtungen, die wirksamste langfristige Strategie zur Reduktion von PFAS-Emissionen bleibt. Alternativen müssen sorgfältig bewertet werden, um sogenannte „bedauerliche Substitutionen“ zu vermeiden. Dies ist insbesondere für kritische Technologien wie Lithium-Ionen-Batterien wichtig, da der Übergang zu kohlenstoffarmen Lösungen nicht zu einer erhöhten chemischen Umweltbelastung führen sollte.

Επιστημονική περίληψη για το κοινό

Οι υπερ- και πολυφθοριωμένες ουσίες (PFAS) είναι ανθρωπογενείς χημικές ενώσεις που χρησιμοποιούνται σε πολλά προϊόντα καθημερινής χρήσης: από αντικολητικά μαγειρικά σκεύη και υφάσματα έως ηλεκτρονικές συσκευές και μπαταρίες. Ξεχωρίζουν επειδή απωθούν το νερό και τα λίπη και αντέχουν στη θερμότητα, όμως έχουν ένα σοβαρό μειονέκτημα: δεν διασπώνται εύκολα στο περιβάλλον. Για τον λόγο αυτό, συχνά αποκαλούνται «αιώνια χημικά» και προκαλούν αυξανόμενη ανησυχία για τις επιπτώσεις τους στην ανθρώπινη υγεία και στα περιβαλλοντικά οικοσυστήματα.

Η παρούσα μελέτη διερεύνησε σε ποια καταναλωτικά προϊόντα καθώς και σε τι βαθμό χρησιμοποιούνται οι PFAS, πώς καταλήγουν στο περιβάλλον και ποιες εναλλακτικές λύσεις υπάρχουν. Η ανάλυση των PFAS είναι δύσκολη, επειδή πρόκειται για μια μεγάλη και πολύ διαφορετική οικογένεια χημικών ουσιών, που περιλαμβάνει τόσο μικρά όσο και πολύ μεγάλα μόρια. Τα αποτελέσματα έδειξαν ότι πολλά προϊόντα, όπως στεγανωτικά βερνίκια για πέτρα, υφάσματα, μαγειρικά σκεύη και καλλυντικά, περιέχουν σημαντικές ποσότητες PFAS. Παράλληλα, εντοπίστηκαν προϊόντα χωρίς PFAS, για τα οποία χρησιμοποιούν εναλλακτικές χημικές τεχνολογίες, αποδεικνύοντας ότι η αντικατάσταση είναι εφικτή.

Ιδιαίτερη έμφαση δόθηκε στις μπαταρίες ιόντων λιθίου, που αποτελούν βασικό στοιχείο της πράσινης μετάβασης. Πιο συγκεκριμένα, διαπιστώθηκε ότι PFAS και άλλες φθοριωμένες χημικές ουσίες που χρησιμοποιούνται στις μπαταρίες μπορούν να διαρρεύσουν στο περιβάλλον κατά την παραγωγή, την χρήση, την ανακύκλωση ή την απόρριψή τους. Παρότι σήμερα υπάρχουν πιθανές λύσεις για μπαταρίες χωρίς PFAS, απαιτείται περαιτέρω ανάπτυξη ώστε να διασφαλιστεί ότι είναι αποδοτικές, ανθεκτικές και οικονομικά βιώσιμες.

Εν κατακλείδι, η μελέτη δείχνει ότι ο καλύτερος τρόπος για τη μείωση της ρύπανσης από PFAS είναι η σταδιακή αντικατάστασή τους με ασφαλέστερες εναλλακτικές, βασισμένη σε αξιόπιστα επιστημονικά δεδομένα. Αυτό είναι ιδιαίτερα σημαντικό, ώστε η μετάβαση σε «πράσινες» τεχνολογίες να μην συνοδεύεται από νέα περιβαλλοντικά προβλήματα.

List of publications

Publications summarized in this doctoral thesis:

- I Eleni K. Savvidou, Shivani Cott, Khushi Desai, Jonathan P. Benskin, Hannah L. Ray, Anna Young, Joseph G. Allen, Heather Whitehead, Graham Peaslee, Ian T. Cousins; **Fluorine Mass Balance in Commercial Stone Sealers from the US Market and Evaluation of PFAS-free Alternatives.** *Submitted manuscript.*
- II Lisa Skedung,* Eleni K. Savvidou,* Steffen Schellenberger, Anders Reimann, Ian T. Cousins, Jonathan P. Benskin; **Identification and quantification of fluorinated polymers in consumer products by combustion ion chromatography and pyrolysis-gas chromatography-mass spectrometry.** *Environmental Science: Processes & Impacts* 2024, 26 (1), 82-93.
- III Eleni K. Savvidou,* Kyra Spaan,* Malte Posselt, Amanda Rensmo, Flóra Pomázi, Christoph J. Reith, Ian T. Cousins, Jörg Lewandowski, Sándor Baranya, Karl Lilja, Jonathan P. Benskin. **Emissions of Fluorinated Substances from Lithium-Ion Battery Manufacturing and Recycling in Central and Northern Europe.** *Manuscript.*
- IV Eleni K. Savvidou, Amanda Rensmo, Jonathan P. Benskin, Steffen Schellenberger, Xianfeng Hu, Marcel Weil, Ian T. Cousins. **PFAS-Free Energy Storage: Investigating Alternatives for Lithium-Ion Batteries.** *Environmental Science & Technology* 2024, 58 (50) 21908-21917.

*Shared first authorship

Author's contributions:

- I Participated in study design; performed sample preparation, extractions, instrumental analysis and data processing; researched alternatives; took the lead in writing the manuscript.
- II Performed sample preparation, instrumental analysis (CIC) and data processing; took a lead role in writing the manuscript.
- III Participated in study design; organised part of the sampling; performed sample preparation, extractions, instrumental analysis and data processing; took the lead in writing the manuscript.
- IV Designed, organised and carried out interviews with experts; reviewed the literature; took the lead in writing the manuscript.

Other publications related to this thesis:

Amanda Rensmo, Eleni K. Savvidou, Ian T. Cousins, Xianfeng Hu, Steffen Schellenberger, Jonathan P. Benskin; **Lithium-ion battery recycling: a source of per- and polyfluoroalkyl substances (PFAS) to the environment?** *Environmental Science: Processes & Impacts* 2023, 25 (6), 1015-1030.

Robin Vestergren, Anders Appelblom, Simona A. Bălan, Sicco H. Brandsma, Thomas A. Bruton, Ian T. Cousins, Jeremy R. Gauthier, Audun Heggelund, Jenny Ivarsson, Anna Kärrman, Lisa Melymuk, Chijioke Olisah, Amanda Rosen, Eleni K. Savvidou, Steffen Schellenberger, Lisa Skedung, Petteri Talasniemi, Tonie Wickman, Jonathan Zweigle, Christian Zwiener, Jonathan P. Benskin; **A Systematic Workflow for Compliance Testing of Emerging International Classwide Restrictions on PFAS.** *Environmental Science & Technology* 2024, 58 (34), 14968-14972.

Romain Figuière, Luc T. Miaz, Eleni K. Savvidou, Ian T. Cousins; **An Overview of Potential Alternatives for the Multiple Uses of Per- and Polyfluoroalkyl Substances.** *Environmental Science & Technology* 2025, 59 (4), 2031-2042.

Abbreviations

BOB	Bis(oxalato)borate
CI	Chemical ionisation
CIC	Combustion ion chromatography
D4	Octamethylcyclotetrasiloxane
diPAP	polyfluoroalkyl phosphate diester
ECHA	European Chemicals Agency
EI	Electron ionisation
EOF	Extractable organofluorine
ESI	Electrospray ionisation
EU	European Union
EV	Electric vehicle
FEC	Fluoroethylene carbonate
GC	Gas chromatography
INAA	Instrumental neutron activation analysis
LC	Liquid chromatography
LIB	Lithium-ion battery
monoPAP	polyfluoroalkyl phosphate monoester
MS	Mass spectrometry
OECD	Organisation for Economic Co-operation and Development
PBT/vPvB	persistent, bioaccumulative, toxic/ very persistent and very bioaccumulative
PEO	Poly(ethylene oxide)
PFAA	Perfluoroalkyl acid
PFAS	Per- and polyfluoroalkyl substance
PFBS	Perfluorobutanesulfonic acid
PFCA	Perfluoroalkyl carboxylic acid
PFHxA	Perfluorohexanoic acid
PFHxS	Perfluorohexanesulfonic acid
PFOA	Perfluorooctanoic acid
PFOS	Perfluorooctanesulfonic acid
PFPEs	Perfluoropolyethers
PFPrA	Perfluoropropionic acid
PFSA	Perfluoroalkane sulfonic acid
PIGE	Particle induced gamma-ray emission spectroscopy
PLOT	Porous layer open tubular
(e)PTFE	(Expanded) Polytetrafluoroethylene
PVDF	Polyvinylidene fluoride
Py	Pyrolysis
REACH	Registration, Evaluation, Authorisation and Restriction of Chemicals
SFP	Side-chain fluorinated polymer
SPE	Solid phase extraction
TF	Total fluorine
TFA	Trifluoroacetic acid
TFMS	Trifluoromethane sulfonic acid
TFSI' /NTf₂/bis-FMeSI	Bis(trifluoromethanesulfonyl)imide
TOP	total oxidizable precursor
U.S. EPA	United States Environmental Protection Agency
WWTP	Wastewater treatment plant

Prologue

Imagine a molecule that can simultaneously repel water and oil, can resist the harshest conditions from very high temperatures to very aggressive chemicals, and that can endure for centuries without ever really breaking down. Now imagine billions of these molecules dispersed across the globe, in cookware we use to prepare food, in buildings where we live, in batteries that power our phones, but somehow also in rivers, air, soils and oceans. Everywhere around us. This is the paradox that PFAS embody: invisible chemical entities with extraordinary properties that have enabled countless technological innovations, yet have the potential to impact human and environmental health.

From the extraordinary strength of the carbon-fluorine bond at the molecular scale to the large-scale consequences observed in nature and society, PFAS exemplify the dual nature of chemistry itself. They demonstrate how chemical ingenuity can solve practical challenges, from non-stick surfaces to water repellent coatings and high-performance energy storage, while at the same time highlighting the unforeseen consequences of introducing manmade substances into complex natural systems. Over the past two decades PFAS have moved from industrial and regulatory neglect to the scientific forefront and regulatory attention, capturing concerns of scientists, policymakers and the public alike. This thesis examines their presence in modern materials, the analytical challenges that obscure their detection, and the possibilities for transitioning towards safer alternatives.

1. Introduction

1.1 The class of PFAS

According to the most recent definition by the Organisation for Economic Co-operation and Development (OECD), PFAS encompass all chemicals that contain at least one fully fluorinated $-CF_2-$ or $-CF_3$ moiety, without any hydrogen or halogen (Br, Cl, I) attached to the very same carbon atom (OECD, 2021). Based on this broad structural definition that includes low-molecular weight as well as polymeric substances (Figure 1), more than 7 million compounds are currently listed on PubChem that would be considered a PFAS (Schymanski et al., 2023).

At the core of PFAS chemistry lies the carbon-fluorine (C-F) bond, which is the strongest single bond found in organic molecules. Fluorine is the most electronegative element, resulting in a polarised C-F bond with electrostatic attributes and a large dipole moment. Additionally, the small size of the fluorine atom (1.47 Å) makes it suitable to replace hydrogen (O'Hagan, 2008). Especially highly fluorinated alkyl chains impart extreme thermal, chemical and biological stability, as well as pronounced hydrophobicity and lipophobicity (Buck et al., 2011). These characteristics have enabled their widespread use in numerous applications, ranging from industrial processes to consumer applications and advanced technologies (Glüge et al., 2020).

However, the same properties that make PFAS technologically valuable also make them environmentally problematic. Their extreme resistance to chemical and biological stressors leads to high environmental persistence, with many PFAS remaining intact or transforming to persistent degradation products (Cousins et al., 2020). In addition, there are several PFAS that are bioaccumulative (Khan et al., 2023; Taher and Lohmann, 2025; Yates et al., 2025) and capable of long-range transport (Faust, 2023; Muir et al., 2025), resulting in their widespread occurrence in the environment, wildlife and humans (De Silva et al., 2021; Evich et al., 2022). Toxicological research has so far focused only on a small subset of PFAS, particularly perfluoroalkyl acids (PFAAs), including the widely studied perfluorooctanoic acid (PFOA) and perfluorooctanesulfonic acid (PFOS). Adverse effects for these two PFAS include reproductive toxicity, hepatotoxicity, immune suppression, endocrine disruption, and cancer (Fenton et al., 2021).

Actions have therefore largely concentrated on these legacy PFAS, exemplified by the voluntary phase-out of PFOS by 3M in 2000 (U.S. EPA, 2000) and a stewardship program initiated by the United States Environmental Protection Agency (U.S. EPA) in 2006, which aimed to stop production of PFOA by 2015 (U.S. EPA, 2006). These efforts were followed by the listing of PFOS (2009), PFOA (2019), and perfluorohexanesulfonic acid (PFHxS; 2022) under the Stockholm Convention on persistent organic pollutants (POPs) (Stockholm Convention on POPs, n.d.). Nevertheless, these phase-outs have mostly resulted in the substitution of PFOA and PFOS with other PFAS, rather than a transition towards fundamentally different, non-fluorinated chemistries.

Given their extreme persistence, continuous use, and global mobility, environmental concentrations of PFAS are expected to continue increasing, raising concerns about long-term and still largely unknown consequences (Cousins et al., 2022). Reflecting this growing awareness, in 2023, five European countries (Sweden, Norway, Denmark, Germany and the Netherlands) submitted a proposal to restrict the entire PFAS class under the Registration, Evaluation, Authorisation and Restriction of Chemicals (REACH) regulation affecting all industries (ECHA, 2023), which is an unprecedented regulatory effort aiming to address the issue at the level of the chemical class rather than on a substance-by-substance approach.

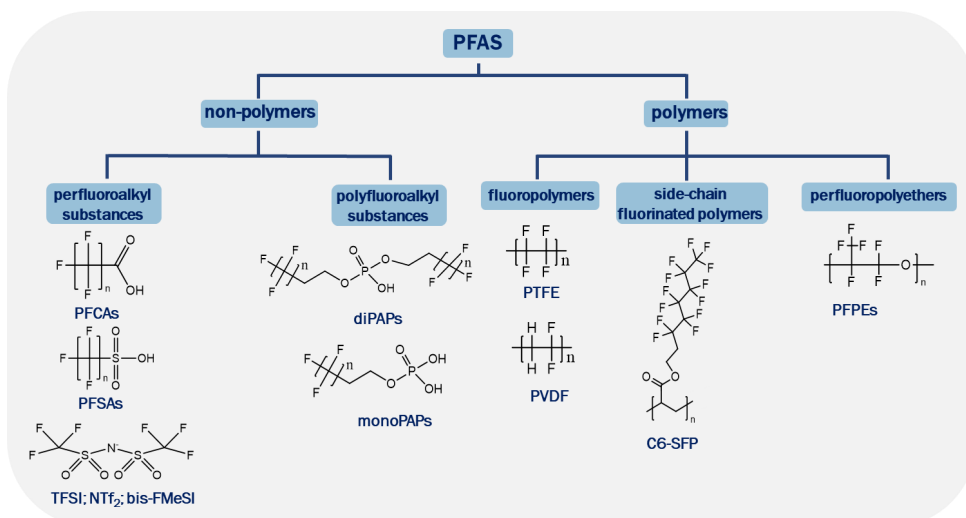


Figure1. Overview of the class of PFAS with some example compounds explored in this thesis, including perfluoroalkyl carboxylic acids (PFCAs), perfluoroalkane sulfonic acids (PFSA), bis(trifluoromethanesulfonyl)imide (TFSI; NTf₂; bis-FMeSI), polyfluoroalkyl phosphate diesters (diPAPs), polyfluoroalkyl phosphate monoesters (monoPAPs), polytetrafluoroethylene (PTFE), polyvinylidene fluoride (PVDF), C6-side-chain fluorinated polymers (C6-SFP), perfluoropolyethers (PFPEs).

1.2 The early days of fluorine chemistry and the first use applications

Organofluorine chemistry is almost exclusively synthetic, as only a few naturally occurring organic compounds incorporating fluorine in their molecular structure are known (Dolbier, 2005; Petkowski et al., 2024). Fluorine itself is a highly unusual element: it is the most electronegative element in the periodic table and highly reactive as F₂ (Dolbier, 2005). As a result, the development of organofluorine chemistry was technically challenging and progressed slowly in its early stages (Banks and Tatlow, 1994). In principle, hydrogen atoms in organic molecules can be replaced by fluorine, enabling the formation of an exceptionally large and diverse range of organofluorine compounds (Sandford, 2000).

A major milestone was the first isolation of elemental fluorine by Henri Moissan in 1886, which marked the beginning of systematic research in this field. Shortly thereafter, in 1892, Frédéric Swarts laid the foundation for organofluorine synthesis by developing reactions that converted chlorocarbons into chlorofluorocarbons using antimony trifluoride (SbF₃) (Dolbier, 2005). This so-called Swarts reaction represented a crucial advance, enabling the production of fluorinated organic compounds and paving the way for their practical application.

Building on these early synthetic developments, one of the first large-scale applications of organofluorine chemistry emerged with the introduction of chlorofluorocarbons (CFCs) as refrigerants. In the early 20th century, General Motors sought alternatives to the flammable, corrosive and toxic substances then used in refrigeration, such as sulphur dioxide (SO₂) and ammonia (NH₃) (Okazoe, 2009). Thomas Midgley Jr. identified fluorinated aliphatic compounds as promising candidates, synthesising dichlorodifluoromethane (CCl₂F₂), which led to the invention of “Freons®.” These compounds were considered initially a technological success, until their role in stratospheric ozone depletion was discovered by Mario J. Molina and F. Sherwood Rowland (1979), whose work was then recognized with the Nobel Prize in Chemistry in 1995 (Dolbier, 2005; Okazoe, 2009).

In 1938, another landmark discovery occurred when Roy J. Plunkett at DuPont accidentally synthesised polytetrafluoroethylene (PTFE) while attempting to develop novel refrigerants (Banks and Tatlow, 1994). PTFE proved to be an exceptional material, exhibiting high thermal stability, chemical resistance and low friction. Although the production of fluoropolymers was initially not economically viable, the technological demands of the Second World War accelerated their development and use in critical applications. In particular, PTFE was employed in components of the Manhattan Project for the construction of the atomic bomb, where materials capable of handling uranium hexafluoride (UF_6) were needed (Okazoe, 2009). In parallel, major advances in fluorination chemistry were achieved, including cobalt trifluoride (CoF_3)-based perfluorination in 1947 and, more importantly, Simon's electrochemical fluorination, which enabled the large-scale and commercial production of fluorinated compounds (Dolbier, 2005; Okazoe, 2009).

Building on these wartime innovations, post-war advances in organofluorine synthesis and manufacturing enabled the large-scale production of PTFE, paving the way for its widespread commercial adoption. In 1945, PTFE was registered under the tradename Teflon[®], and by 1950 DuPont opened its first commercial plant in Parkersburg, West Virginia, United States (Banks and Tatlow, 1994; "The History of Teflon[™] Fluoropolymers," n.d.). One of the most prominent early consumer applications emerged in the 1950s, when French chemist Marc Grégoire introduced PTFE-coated cookware (1954), popularising non-stick surfaces and founding the company Tefal in 1956 (McKeen, 2013; Tefal, n.d.; Thomas, 1998). Subsequent innovations expanded the range of applications, notably the development of expanded PTFE (ePTFE) membranes, commercialised as Gore-Tex[®], which revolutionised functional textiles by combining water repellence with breathability (GORE-TEX, 2022). However, rapid upscaling of PTFE production also introduced significant environmental consequences. Early manufacturing processes (from the 1950s onward) relied on PFOA as a processing aid, which was subsequently released into the environment, including from DuPont's facility in Parkersburg, West Virginia (Gaber et al., 2023; Prevedouros et al., 2006). These emissions led to widespread contamination of drinking water sources and human blood with PFOA (Richter et al., 2018; U.S. EPA, 2013), with particularly severe impacts on local communities. This contamination resulted in a large class-action lawsuit in the early 2000s, leading to substantial compensation payments and increased regulatory scrutiny (Gaber et al., 2023). The case, later brought to broader public attention through the film "Dark Waters," marked a turning point in raising public awareness of PFAS pollution and the long-term environmental, health, and economic consequences associated with fluoropolymer production.

Beyond materials science, organofluorine chemistry also had profound impact on the medical field. The incorporation of fluorine into pharmaceutical compounds can substantially alter their biological activity, stability, and bioavailability, contributing to the development of anaesthetics, antidepressants, and many other therapeutics (Dolbier, 2005; Sandford, 2000). Together, these early successes laid the foundation for the widespread adoption of fluorinated chemistries across numerous industrial sectors in the decades that followed, ultimately giving rise to an increasingly diverse range of fluorinated substances, including the class now referred to as PFAS.

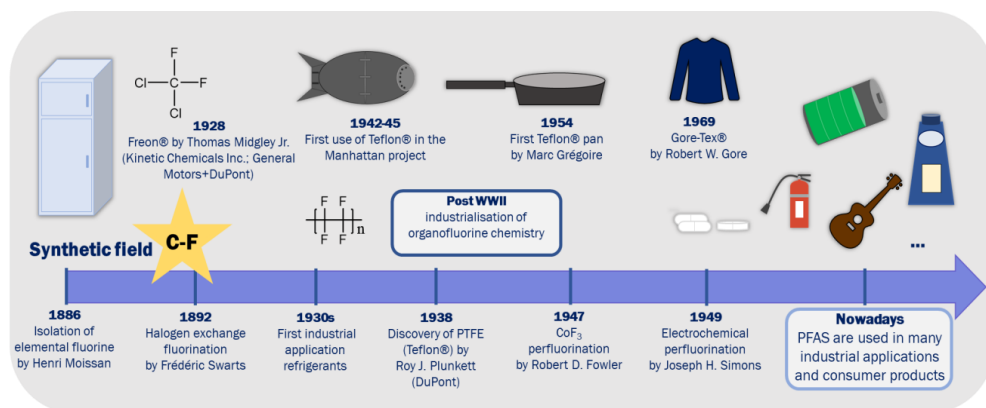


Figure 2. Historical development of synthetic methods and product applications of PFAS.

1.3 PFAS in today's world

With these historical developments, fluorinated chemistries have become deeply embedded in modern industrial processes and consumer products. Today, PFAS are used across a wide range of applications, driven by their unique physicochemical properties and functional performance. While several uses are well documented, such as in cookware, textiles, cosmetics, firefighting foams and food packaging, many applications remain poorly characterised. This knowledge gap is often due to limited transparency from manufacturers, but also reflects analytical challenges and a lack of comprehensive data.

Given their widespread presence and functional diversity, two specific use cases were examined in more detail within this thesis. Studies indicate that PFAS are widely incorporated into building and construction materials, making the building sector one of the largest users of PFAS in the Nordic countries (Glüge et al., 2020). Combined estimates suggest that coatings and buildings account for more than a quarter of total PFAS consumption in the Nordic region (Glüge et al., 2020), while the REACH restriction dossier estimates that approximately 5 000 to 13 000 metric tons of PFAS are used each year in construction-related products across the EU (ECHA, 2023). The construction sector encompasses a broad range of materials and product types, contributing to the complexity of assessing PFAS use in this domain (Green Science Policy Institute et al., 2021). One specific application examined in this work is stone sealers, which are used to protect mineral-based materials from degradation (Tressaud, 2019). To provide long-term protection, stone sealers must withstand environmental and chemical stress while interacting effectively with the stone matrix. This requires materials with low surface tension together with a suitable molecular size to allow penetration into porous stone materials (Camaiti et al., 2021). Polymeric PFAS are commonly used for this purpose to create hydrophobic surface treatment (Camaiti et al., 2021; Tressaud, 2019). These products are applied both indoors and outdoors and play an important role in the conservation of buildings, monuments, and cultural heritage. Stone sealers therefore represent a particularly relevant case for evaluating PFAS substitution strategies, especially in the context of the essential use concept (see section 1.5).

While PFAS are critical in building materials, their utility also extends to high-performance technologies, such as lithium-ion batteries (LIBs), where fluorination similarly enhances material properties, especially electrochemically (Wang et al., 2024). One application that has gained considerable attention recently, particularly in light of the proposed universal PFAS restriction, is LIBs. In LIBs, PFAS and other fluorinated compounds are used in critical components such as binders, electrolytes, but also in other parts such as separator coatings, gaskets, seals, valves and sealings ("PFAS restriction proposal - RECHARGE statement for 2nd Call for Evidence," 2021; Rensmo et al., 2023). Fluorination plays a crucial role in the design of

materials suitable for LIBs applications. The introduction of fluorine can enhance oxidative and thermal stability, improve ion conductivity, and increase tolerance to wide temperature ranges, high voltages and repeated cycling. In addition, fluorinated materials can contribute to nonflammability, supporting safer battery operation (Wang et al., 2024). The focus here is on cathode binders, which commonly use polyvinylidene fluoride (PVDF) due to its chemical inertness, stability and good adhesion. In electrolytes small molecule PFAS such as lithium bis(trifluoromethanesulfonyl)imide (LiTFSI) are mostly used as additives to improve performance (Rensmo et al., 2023; Wang et al., 2024). LiTFSI belongs to the class of so-called ionic liquids, salts composed entirely of ions that typically melt below 100 °C. Their modular pairing of cations and anions allows systematic tuning of physicochemical properties, making them attractive for electrochemical applications, including battery electrolytes. Their low vapour pressure and low flammability have further contributed to their reputation as potential “green solvents” (Beil et al., 2021; Zhou et al., 2023). The use of LiTFSI as a fluorinated ionic liquid illustrates how novel PFAS chemistries are being designed to meet increasingly demanding performance requirements, reflecting the continuous evolution of fluorinated materials in advanced materials.

Although the functional benefits of PFAS are clear during use, their environmental footprint extends across the entire lifecycle, from manufacturing to end-of-life (Barola et al., 2023; Coffin et al., 2023; Dalmijn et al., 2023; Guelfo et al., 2024). Emissions at multiple stages can lead to environmental contamination and human exposure, underscoring the need for a comprehensive understanding of PFAS release pathways. Assessing these emissions is essential for risk evaluation and for guiding mitigation strategies, including the identification of suitable substitution options. This complexity is compounded by the presence of a wide variety of PFAS in diverse product and environmental matrices. Despite regulatory actions on legacy compounds such as PFOA and PFOS, many PFAS continue to be used in industrial and consumer applications and therefore be potentially emitted, highlighting the ongoing challenges in managing this diverse class of chemicals.

1.4 The challenge of analysing PFAS

PFAS encompass an extremely large and diverse chemical space, which makes their analysis particularly challenging for analytical chemists. Due to their widespread use and occurrence in the environment, analytical methods must be applicable to a broad range of matrices with varying degrees of complexity. Traditional analytical approaches have primarily focused on targeted analysis, that is, the detection of known PFAS using authentic standards, most commonly by liquid chromatography coupled with mass spectrometry (LC-MS). Even for the characterisation of consumer products most studies have relied on such targeted methodologies (Bečanová et al., 2016; Dueñas-Mas et al., 2023; Favreau et al., 2017; Herzke et al., 2012; Janousek et al., 2019; Kotthoff et al., 2015; Vestergren et al., 2015; Ye et al., 2015).

However, targeted analysis captures only a limited fraction of the PFAS present, often missing polymeric PFAS, as these compounds are typically not readily extractable and are not well suited for analysis by LC-MS. As a result, conventional PFAS workflows are biased towards small, extractable molecules, while high-molecular-weight and non-extractable polymeric PFAS remain largely unaccounted for. In practice, no single analytical method is sufficient to obtain a comprehensive overview of PFAS in complex samples, and a combination of complementary approaches is therefore required. An effective strategy is to begin with broad screening methods and subsequently narrow the focus towards more specific analyses (**Figure 3**).

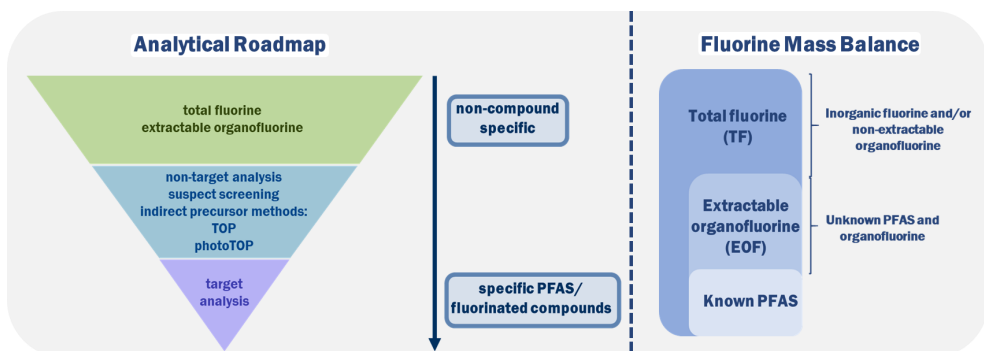


Figure 3. Analytical roadmap for PFAS analysis (left), starting with broad, non-compound methods at the top and progressively narrowing to techniques targeting individual substances, with total oxidizable precursor (TOP) and photochemically induced TOP (photoTOP) assays. The fluorine mass balance concept (right) illustrates how information gained from different measurements can be integrated and interpreted in relation to one another.

Broad analytical approaches include measurements of total fluorine (TF), which determine the fluorine content (both inorganic and organic) from the raw material sample. Following extraction and subsequent fluorine determination of the extract, the extractable organofluorine (EOF) content can be obtained. EOF measurements are designed to remove inorganic fluorine, and therefore can infer whether the detected fluorine is organic. This method is, however, limited by the extraction design and may still fail to capture all PFAS present, depending on their physicochemical properties and behaviour. Moreover, caution is warranted, as other inorganic fluorinated species not removed by the chosen extraction method may also be present within the EOF fraction. Several techniques are available for measuring TF, including combustion ion chromatography (CIC), particle induced gamma-ray emission spectroscopy (PIGE) and instrumental neutron activation analysis (INAA) (Schultes et al., 2019). CIC is the most widely applied technique, as it can be used to determine both TF and EOF, followed by PIGE as a surface sensitive-screening technique (Idowu et al., 2025). To further refine the analysis, more advanced techniques can be applied, such as non-target screening, which aims to detect previously unknown compounds, and suspect screening, which targets substances from predefined lists even when authentic standards are not available (Hollender et al., 2023). Other complementary techniques include the total oxidizable precursor (TOP) assay and the photochemically induced TOP (photoTOP), which estimate PFAS precursors, by measuring their oxidation products (Houtz and Sedlak, 2012; Zweigle et al., 2022). For the characterisation of polymeric PFAS, techniques such as pyrolysis-gas chromatography-mass spectrometry (Py-GC-MS) can provide valuable structural information by thermally degrading polymers into characteristic fragments (Akoueson et al., 2021; Muensterman et al., 2022). Py-GC-MS is particularly useful for identifying polymeric PFAS in materials where conventional extraction-based methods fail.

Together, these approaches form the basis of the fluorine mass balance strategy, which integrates broad and targeted analyses to improve PFAS characterisation. Discrepancies between TF and EOF measurements can indicate the presence of inorganic fluorine and/or non-extractable organofluorine. Further comparison of EOF with quantified, known PFAS provides insight into the proportion of unknown PFAS and other organofluorine that remains uncharacterised. The fluorine mass balance approach has increasingly been applied in recent years to both consumer products (Gonzalez de Vega et al., 2024; Lauria et al., 2022; Pütz et al., 2022; Robel et al., 2017; Schultes et al., 2018) and environmental samples (Aro et al., 2021; Cioni et al., 2023; He et al., 2025; Spaan et al., 2020). This highlights the need for complementary analytical strategies to better characterise the PFAS burden in complex matrices, particularly in materials and products where polymeric PFAS are intentionally used.

1.5 From detection to substitution: searching for alternatives

The most effective path forward for reducing PFAS emissions, exposure and associated environmental and health risks is the phase-out of PFAS and the identification of safer alternatives that can provide equivalent performance in their intended use applications. Several concepts and policy approaches have been proposed to support this transition. As part of the European Green Deal, the European Commission published the “EU Chemicals Strategy for Sustainability”, which explicitly aimed to phase out non-essential uses of PFAS (European Commission, 2020). The concept of “essential use” originates from the Montreal Protocol, which sought to phase out chlorofluorocarbons while allowing exemptions for uses deemed essential. Applied to PFAS, three essentiality categories have been proposed: essential, substitutable and non-essential. Essentiality is defined based on whether a use is necessary for health or safety and functioning of society and other critical purposes (Cousins et al., 2019). A complementary approach is “functional substitution”, which focuses on replacing the chemical based on the function it provides within a given application (Tickner et al., 2015). This concept is particularly useful when combined with the essential use framework (Roy et al., 2022). Once potential alternatives are determined, a chemical alternatives assessment should be conducted to evaluate their technical performance, environmental fate, and human health impacts, thereby minimizing the risk of regrettable substitution. Lastly, a more precautionary approach is the “safe and sustainable by design” concept, where safety and sustainability are already integer in the innovation and design of chemicals, materials and production processes (Soeteman-Hernández et al., 2024).

While these frameworks provide important guidance for phasing out PFAS, their systematic application is beyond the scope of this thesis. Instead, this work focused on identifying PFAS-free alternatives and generating data on the occurrence and properties, providing necessary knowledge that can inform future substitution strategies and safe design approaches.

2. Aim and scope

The overall hypothesis of this thesis is that the prevalence and diversity of PFAS in consumer products and technologies are substantially underestimated. This gap in understanding arises, in part, because a) only a limited suite of consumer products has been characterised for PFAS; and b) that current analytical approaches predominantly focus on known, low molecular weight PFAS, often overlooking polymeric PFAS, which are widely used yet analytically challenging to characterize. By expanding the scope of investigation of PFAS we can obtain a more comprehensive picture of PFAS use and, in turn, identify opportunities to transition towards safer, PFAS-free alternatives. Moreover, improved knowledge of PFAS use in commerce offers greater insight on the substances we might expect to find in the environment.

This thesis aimed to investigate and characterise the use of PFAS in a range of consumer and technological applications that have received limited attention to date. This specifically included sealers as a subgroup of building materials and lithium-ion batteries as a critical emerging technology. A key objective was to evaluate whether viable alternatives exist for these product types. The findings are intended to support industry and policymakers in identifying unnecessary PFAS uses and in promoting a transition towards more sustainable and less harmful solutions. In addition, the work explored complementary analytical methods for detecting and characterising polymeric PFAS and other fluorinated substances that fall outside the scope of traditional targeted workflows.

Accordingly, the objectives underlying the four papers were to:

Paper I: Characterise PFAS occurrence in under-studied product types (here stone sealers);

Paper II: Develop and evaluate analytical approaches for polymeric PFAS in consumer products;

Paper III: Investigate the environmental release of novel and emerging PFAS during the lifecycle of LIBs;

Paper I + IV: Search for potential alternatives to PFAS in stone sealers and LIBs.

These objectives guided five main research questions:

1. What is the PFAS content and how widely are they used in modern consumer products and technologies?
2. What PFAS are used or detectable in sealers, LIBs, cookware, textiles, electronics and personal care products?
3. Why are PFAS used in these product categories?
4. Do these PFAS occur in the environment?
5. Are there technologically feasible alternatives without environmental and human-health impacts?

3. Methods

3.1 Sampling and data acquisition

For **Paper I**, 35 indoor stone sealer products intended for use on countertop, table top, floor, grout and wall coatings were selected. The sampling focused on the US market and was based on online searches of major home-improvement stores as well as interviews with experts to obtain the most common and popular products. Although this study focused on sealers primarily marketed for indoor use, there are also products for outdoor application, and similar products may find use in stone conservation with monumental significance.

For **Paper II**, a total of 35 products from four product categories including cookware, textiles, electronics and personal care products were obtained from industry partners in the “POPFREE industry” project or purchased in Sweden. Samples included products known to use PFAS, but also a selection of items with unknown contents. Individual items were disassembled and sub-sampled into smaller portions depending on the product and material type, resulting in 45 product samples. Cookware samples included non-stick, PTFE and ceramic-coated items; electronics consisted of components such as charging cases, coffee makers and speakers; and personal care products contained dental floss, band aids and packaging from facial serums.

For **Paper III**, water samples were collected in three European countries where activities related to LIB manufacturing, use, and/or disposal were taking place. In Hungary, 0.5 L samples were taken in September 2024 upstream and downstream of two large LIB manufacturers along the Danube, including samples from the Vác wastewater treatment plant (WWTP) effluent and tap water from Budapest. In Germany, 0.5 L samples were collected between 2016 and 2024 from the River Erpe downstream of the Münchehofe WWTP, which receives wastewater from an electric vehicle (EV) manufacturer; additional influent and effluent samples were provided by the WWTP. In Sweden, 1 L surface water samples were collected around a recycling facility and downstream a nearby landfill in Halmstad and around a research and development facility of a (recently bankrupt) LIB producer and recycler in Västerås in early 2025, together with effluent from four WWTPs and landfill leachate from a site near Stockholm.

For **Paper IV**, the literature was reviewed to identify potential alternatives to commonly used PFAS in LIBs and to assess the state-of-the art. To capture current technological possibilities, stakeholders from both academia and industry were consulted and interviewed. These included companies that offer PFAS-free solutions as well as scientists conducting research in the battery field and have relevant expertise. Together, these perspectives were intended to provide a balanced understanding of the current technological state and potential developments.

3.2 Sample preparation

Only minimal sample preparation is needed for TF analysis, and samples can be analysed directly using small sample amounts. The same applies to analysis by Py-GC-MS. In **Paper I**, samples (in the microgram range) were weighed directly into the ceramic boats. Samples from **Paper II** were more complex; some were disassembled and cut into smaller pieces, while for others the surface material was scraped off and collected. The sample amounts were also in the microgram range for both TF and Py-GC-MS.

For the fluorine mass balance in **Paper I**, samples were extracted by adding 0.3% ammonium hydroxide in methanol to the sample (as the sealers were suspensions or emulsions), followed by ultrasonication, and removal of the supernatant (when applicable), which was then filtered. The obtained extract was divided for EOF and targeted analysis by LC-MS. Isotopically labelled internal standards were added only to aliquots intended for targeted analysis, and not to the EOF fraction.

The sample preparation in **Paper III** was more elaborated, as it was water samples with varying matrices that had to be prepared for targeted analysis. A solid phase extraction (SPE) method designed for water analysis was slightly modified to capture a wide range of PFAS including very polar ultrashort-chain PFAS and inorganic anions such as hexafluorophosphate (PF_6^-) and tetrafluoroborate (BF_4^-) but also longer-chain PFAS. In short, SPE weak ion exchange cartridges were first washed and conditioned. Samples were spiked with isotopically-labelled internal standards the day before extraction, then filtered if necessary, and the pH was set to 4. Then the samples were loaded dropwise onto the cartridges, and afterwards eluted. The extract underwent another clean up step with an activated carbon cartridge. Then the extract was evaporated under gentle nitrogen flow to almost dryness. It was reconstituted with methanol; buffer water and recovery standard were added.

3.3 Instrumental analysis

3.3.1 TF and EOF analysis by CIC in Paper I-II

TF (**Paper I-II**) and EOF (**Paper I**) were quantified using a previously established CIC method (Schultes et al., 2018). For TF analysis, 0.1-0.7 mg (**Paper I**) and 0.1-0.9 mg (**Paper II**) of sample were weighed directly onto pre-baked ceramic boats. For EOF analysis (**Paper I**), 50 μL of the extracted sample were applied to the boats. Combustion was carried out in a HF-210 Mitsubishi furnace operated at 1100 $^\circ\text{C}$, using argon (200 mL min^{-1}) and oxygen (400 mL min^{-1}) as carrier gases. The combustion gases were captured in MilliQ water using a GA-210 gas absorption unit (Mitsubishi). Subsequently, 50 μL of the absorption solution were injected onto an ion chromatograph (Dionex Integriion HPIC, Thermo Fisher Scientific). Chromatographic separation was performed on an anion exchange column set (Dionex IonPac AS19 2 \times 50 mm guard column and 2 \times 250 mm analytical column, 7.5 μm particle size) maintained at 30 $^\circ\text{C}$. Aqueous potassium hydroxide was used as mobile phase with a gradient from 8 mM to 60 mM at a flow rate of 0.25 mL min^{-1} . A conductivity detector was used for the detection of fluoride.

Combustion of organosilicate compounds in CIC is not recommended, as it can generate silicon oxide particles that compromise instrument performance. Due to these technical limitations, TF analysis in **Paper I** was additionally performed by PIGE at the University of Notre Dame. Samples were deposited onto fluorine-free Whatman 1 qualitative filter papers, which also served as procedural blanks. The prepared samples were mounted to stainless-steel frames and analysed using ex vacuo ion beam conditions. Proton irradiation was carried out using 10-50 nA and proton energies between 3.4 and 3.9 MeV, with an irradiation time of 180 seconds per sample. The proton beam was extracted through an 8 μm Kapton[®] foil and directed onto the targets in air.

3.3.2. Py-GC-MS analysis in Paper II

Small amounts of each sample (approximately 0.5 mg) were introduced to a pyrolysis chamber that was heated to 165 $^\circ\text{C}$ and purged with helium. The samples were pyrolyzed at 700 $^\circ\text{C}$ and the resulting volatiles were analysed on an Agilent 8890 GC coupled to an Agilent 5977B MSD mass spectrometer. Chromatographic separation was performed on a 30 m \times 0.25 mm i.d. Agilent HP-5MS Ultra Inert (Chrompack) column. The GC oven program was initiated at 50 $^\circ\text{C}$ for 2 minutes, then increased to a final temperature of 325 $^\circ\text{C}$ at 21 $^\circ\text{C min}^{-1}$, which was maintained for 12 minutes. Mass spectra were acquired using electron ionisation (EI) mode.

3.3.3. LC-MS analysis in Paper I and III

Target PFAS analysis was performed using a Dionex 153 Ultimate 3000 liquid chromatograph coupled to a Thermo Scientific Q Exactive HF Orbitrap operated in negative electrospray ionisation (ESI) mode. Chromatographic conditions were adapted to cover the specific PFAS relevant to each paper. Part of samples from **Paper III** were analysed on a Thermo ultrahigh-performance liquid chromatograph coupled to a Thermo TSQ Quantiva triple-quadrupole mass spectrometer (Thermo Scientific).

In **Paper I**, in addition to perfluoroalkyl carboxylic acids (PFCAs) and perfluoroalkane sulfonic acids (PFSA_s), polyfluoroalkyl phosphate mono- and diester (mono- and diPAPs) were included. Chromatographic separation was carried out on a 2.1x50 mm Acquity UPLC®BEH C18 column (1.7 μm particle size) operated at 50 °C. To enable monoPAP analysis, mobile phase A consisted of 95% water and 5% methanol with 2mM ammonium acetate and 5mM 1-methyl piperidine, while mobile phase B consisted of 75% methanol, 20% acetonitrile and 5% water with the same additives (Ullah et al., 2011). This approach was not suitable for diPAPs due to inadequate chromatographic resolution. Therefore, for diPAP analysis, water:acetonitrile (90:10, 2 mM ammonium acetate) and acetonitrile:water (99:1, 2 mM ammonium acetate) were used as mobile phases.

In **Paper III**, very polar compounds, including ultrashort-chain PFAS and inorganic anions (BF₄⁻ and PF₆⁻), were targeted and required a separate chromatographic method (He et al., 2025). Chromatographic separation was achieved on an Acclaim™ Trinity P1 HPLC column using mobile phase A (20mM ammonium acetate in water) and mobile phase B (20 mM ammonium acetate in 99 % methanol and 1% water). Short- and long-chain PFAS were analysed using a method from Waters (Waters, 2023), employing an Atlantis™ BEHTM C18 AX Mixed Mode column (2.1 × 100 mm, 1.7 μm; Waters). Mobile phase A was 2 mM ammonium acetate in water, while mobile phase B was 0.1% (v/v) ammonium hydroxide in methanol.

3.4 Identification of potential alternatives to PFAS in Paper I and IV

In **Paper I**, PFAS-free sealers were first identified through chemical analysis and examination of ingredient lists. Additional potential compounds were retrieved from a PFAS alternatives database (Figuière et al., 2025). All identified compounds were subsequently screened for their regulatory status. In **Paper IV**, potential alternatives were gathered primarily through targeted literature review and consultations with researchers and alternatives providers. This process was exploratory and not conducted as systematic alternatives assessment.

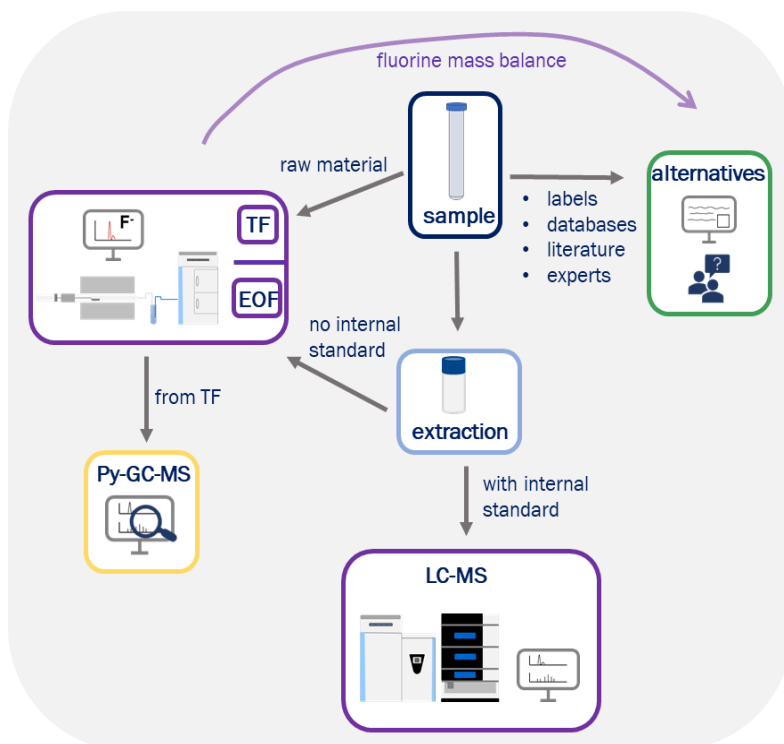


Figure 4. Schematic overview of the methodological approach in this thesis, with total fluorine (TF), extractable organofluorine (EOF), pyrolysis-gas chromatography-mass spectrometry (Py-GC-MS) and liquid chromatography-mass spectrometry (LC-MS).

4. Results and Discussion

4.1 Analysis of fluorinated compounds across diverse product categories

4.1.1 Fluorine mass balance of sealers (Paper I)

The building industry is one of the major users of PFAS, with an estimated annual use of 5 241 to 12 725 metric tons (Figuère et al., 2025). Analytical data on PFAS in building materials remain limited, and existing studies often rely on targeted analysis, thereby overlooking especially polymeric PFAS. In this work, the fluorine mass balance approach was applied to comprehensively characterise sealer products.

Among the examined 35 sealers intended for indoor use, 81% contained detectable levels of TF, with concentrations reaching up to 27 150 µg F/g. By comparison, only 52% of the products listed PFAS or fluorinated compounds on their ingredient lists. EOF analysis indicated that most of the fluorine was extractable and therefore likely of organic origin, with levels up to 24 073 µg F/g. Targeted PFAS analysis accounted for only up to 31% of the EOF, leaving a substantial fraction of unidentified fluorine, most likely originating from polymeric PFAS or other low molecular weight PFAS not included in the targeted analysis.

Screening of ingredient labels confirmed the widespread use of polymeric PFAS, often listed under vague terms such as “*fluorochemical polymer*” or “*fluorinated acrylic polymer*”. Low molecular weight PFAS were also declared, including descriptors such as “*partially fluorinated alcohol*” and “*C4 perfluorinated surfactant*” as well as more specific substances such as “*ammonium C6-C16 perfluoroalkylethyl phosphate*” (CAS 65530-70-3). The targeted analysis indicated that mono- and diPAPs were the predominant PFAS sub-class present, although PFAAs were also frequently detected. Notably, PFOA was found in seven products at high concentrations (up to 1 275 ng/g), which is of particular concern. It remains unclear whether the detected PFAAs are impurities, degradation products or intentionally added ingredients. It also remains unclear what polymeric PFAS are used, although the name “*fluorinated acrylic polymer*” and a previous study (Liu et al., 2025) could hint the presence of side-chain fluorinated polymers (SFPs).

The results highlight the presence of both polymeric and low molecular weight PFAS in stone sealers, often under vague or incomplete ingredient declarations. The substantial fraction of unidentified fluorine underscores the limitations of targeted analysis and the need for complementary approaches, such as the fluorine mass balance, to capture the full PFAS burden.

4.1.2 Py-GC-MS as tool for detection of C-F bonds (Paper II)

The universal PFAS restriction proposal provides guideline values for compliance testing of consumer products: 50 ppm (µg/g or mg/kg) fluorine for TF, including polymeric PFAS; 25 ppb for individual targeted PFAS without polymeric PFAS; and 250 ppb for the sum of targeted PFAS with optional precursor conversion (ECHA, 2023). However, substantial analytical challenges remain. While TF analysis is a valuable tool, and CIC is compatible with a broad range of matrices, compliance testing ultimately requires demonstrating that the measured TF originates from PFAS. As shown in **Paper I**, the absence of polymeric PFAS in targeted analysis can lead to large discrepancies between TF/EOF and known PFAS. To address this gap, Py-GC-MS was explored as a potential complementary tool.

Method validation was performed using standards of perfluorobutanesulfonic acid (PFBS), perfluorohexanoic acid (PFHxA), PFHxS and PFOA. Retention times increased with perfluoroalkyl chain length, and the method was able to distinguish PFAAs based on characteristic fragments. PFCAs predominantly produced ions at m/z 69 and 131, whereas PFSAs primarily yielded m/z 69. SFP reference materials (C4, C6, and C8) were also analysed and showed similarly increasing retention times with longer perfluorinated side-chains. Major fragment ions were m/z 69 for the C4-SFP, and m/z 77 for both the C6- and C8-SFPs. Their chromatograms, however, were more complex due to additional peaks originating

from the textile matrices. PTFE reference materials were also pyrolyzed and produced major ions at m/z 81 and 100.

Because electron ionisation (EI) was used, which is a hard ionization technique that causes extensive fragmentation, the method was limited in its suitability for structural elucidation. For future method development, testing a softer ionisation technique such as chemical ionization (CI) is recommended.

The method was further tested on 45 consumer product samples from the Swedish market that were either suspected or known to contain fluorinated polymers. First, TF screening was conducted, yielding ranges of 70–550 000 ppm F in cookware, <13–1 600 ppm F in textiles, <22–3 000 ppm F in electronics, and <22–630 000 ppm F in personal care products. Samples exceeding the 50 ppm threshold were subsequently analysed qualitatively by Py-GC-MS to determine whether the detected fluorine originated from PFAS. PTFE could be detected at levels as low as 0.1–0.2 wt% in samples of unknown composition, particularly within electronics, cookware and personal care products. For cookware with various coatings, ceramic and silicone-based reference materials enabled the identification of coatings in other samples. For textiles, the method successfully identified different SFP finishings. However, some electronics samples remained inconclusive, likely due to the presence of inorganic fluorine derived from mica, which is used as a pigment or filler.

4.2 Environmental occurrence of emerging PFAS from the lifecycle of LIBs (Paper III)

Novel PFAS are emerging as environmental contaminants, specifically those associated with LIBs. With increasing demand for LIBs, potential emissions of such PFAS, and their subsequent environmental impacts, are likely to rise. **Paper III** aimed to assess the lifecycle-associated emissions of PFAS from LIBs, covering manufacturing, downstream use, and recycling in Europe (Hungary, Germany and Sweden) by analysing surface water, WWTP effluent and drinking water. The focus was on selected “battery markers” TFSI⁻ and the inorganic anions PF₆⁻ and BF₄⁻, whose lithium salts are used in electrolytes (Rensmo et al., 2023). These compounds have been detected in environmental samples before (Guelfo et al., 2024; Jiao et al., 2023; Neuwald et al., 2021, 2020; Tisler et al., 2025), and are mostly associated with use in LIB applications. The “battery markers” were detected across all sites, albeit with differing profiles.

In Hungary, along the Danube, TFSI⁻ concentrations remained relatively low and stable (average 0.3 ng/L), including upstream sites suggesting the presence of additional sources beyond the investigated LIB plants. Effluent samples from the WWTP plant receiving discharges from a major LIB manufacturer showed approximately double the TFSI⁻ concentration (0.6 ng/L). Inorganic anions were dominated by BF₄⁻, with concentrations ranging from 5 263 to 9 702 ng/L in river water, and elevated levels were found in the drinking water (1 790 ng/L), suggesting limited removal during riverbank filtration and water treatment processes. Ultrashort-chain PFAS, including trifluoroacetic acid (TFA) and perfluoropropionic acid (PFPrA), were also consistently detected, with TFA averaging 1 232 ng/L in the Danube. Slightly higher levels were observed in drinking water and WWTP effluent, reflecting ongoing anthropogenic input.

In Germany, sampling in the River Erpe downstream of a WWTP receiving discharges from an EV plant revealed higher TFSI⁻ concentrations (average 0.7 ng/L) compared to upstream (0.1 ng/L). This confirms the WWTP’s contribution to local emissions. Temporal monitoring captured dynamic changes in concentrations, with a notable decrease during a one-week plant shutdown in March 2024 (TFSI⁻ decreased to 0.4 ng/L) followed by increases correlating with operational activity, reaching a maximum of 1.5 ng/L by December 2024. PF₆⁻ dominated the inorganic anion profile, peaking at 1 664 ng/L in effluent samples, while BF₄⁻ also exhibited temporal variation (up to 1286 ng/L). Ultrashort-chain PFAS, including TFA, PFPrA and trifluoromethane sulfonic acid (TFMS) were prevalent, whereas long-chain PFAS (PFOA, PFOS) remained low at 2.6 ng/L and 3.1 ng/L, respectively. Correlation analyses suggest linked sources between TFSI⁻, BF₄⁻ and PF₆⁻ and other LIB-related PFAS like TFMS and PFBS.

In Sweden, TFSI⁻ and PF₆⁻ emissions were strongly associated with recycling and landfill activities. Downstream of a landfill near Stockholm, TFSI⁻ concentrations reached up to 329 ng/L in landfill leachate from Stockholm, whereas elevated levels (106 ng/L) were detected downstream of a landfill and recycling facility in a bird protection area in Southern Sweden. This suggests transport and accumulation in receiving environments. Effluent samples from Swedish sites generally exhibited lower concentrations (≤ 1.8 ng/L for TFSI⁻), and tap water measurements were mostly below detection limits or very low, reflecting dilution and partial removal through treatment processes. Ultrashort-chain PFAS, particularly TFA and PFPrA, reached remarkably high levels in landfill leachate (TFA up to 19 782 ng/L; PFPrA up to 18 312 ng/L), highlighting the role of waste streams as secondary sources of PFAS. Other compounds, including TFMS and PFBS were elevated near recycling and landfill sites, further supporting the link between LIB lifecycle activities and local PFAS contamination.

These findings underscore the importance of considering the full lifecycle of LIBs, the varying contributions of manufacturing, recycling, and landfills, and the need for appropriate monitoring and management strategies to mitigate environmental releases of LIB-associated PFAS.

4.3 Exploring non-PFAS options for sealers and LIBs

4.3.1 Silicones as alternatives to PFAS in sealers (Paper I)

The fluorine mass balance in **Paper I** indicated that six sealer products showed no evidence of intentionally added PFAS within the applied analytical framework, with organosilicon compounds identified as their primary functional chemistry based on product labelling. The literature also reported additional PFAS-free chemistries used in sealers, including waxes, acrylics and polyurethanes. Across these systems, key performance requirements include high resistance to chemical, thermal and photo-oxidative stress; strong adhesion to the material; low surface tension and suitable molecular size for penetrating porous materials; and solubility in non-toxic solvents.

Among the most prominent PFAS-free chemistries identified in the sealers were siloxanes and silanes, which are valued for their water repellence, penetration into porous substrates, breathability, thermal stability and durability. These compounds can also be used in combination, resulting in coatings with good breathability, improved thermal stability, and enhanced protection against weathering.

Three silicone-based substances likely defining the sealing function were identified: dimethylmethoxyphenyl siloxane, triethoxyoctylsilane and octamethylcyclotetrasiloxane (D4). Regulatory assessments, however, have indicated that these and other structurally related organosilicon compounds carry potential environmental and human health hazards. These include properties that are classified as persistent, bioaccumulative, toxic/ very persistent and very bioaccumulative (PBT/vPvB), endocrine disruptive and toxic for the reproductive system. While the EU applies stricter controls, in other jurisdictions such as the United States, Canada, and Australia, different assessment outcomes and management options exist.

Additional potential PFAS alternatives were identified through the alternatives database (Figuière et al., 2025), which consisted of synthetic organic compounds and mostly silicone-based compounds, some of which have also been subject to EU regulatory scrutiny due to suspected mutagenic or reproductive hazards. Beyond synthetic materials, natural alternatives such as linseed oil have historically been used for stone treatment because of their water-repellent and consolidating properties. However, drawbacks include resinification, surface yellowing, or reduced long-term hydrophobicity (Snethlage, 2014). Comprehensive comparative assessments of performance, durability, toxicological profiles and environmental impacts for silicone-based and natural alternatives are still lacking. This highlights the importance of comprehensive testing and careful assessment to ensure that PFAS substitutes are both effective and safe, to avoid potential regrettable substitution.

4.3.2 Alternatives to fluorinated components in LIBs (Paper IV)

Attention is increasingly turning to the fluorinated chemicals that are part of the LIB design, and the question arises whether there are safer and sustainable alternatives available that can also realistically replace PFAS in such a critical technology.

Particularly cathodes rely on the use of fluoropolymers such as PVDF and PTFE as binders because of their chemical inertness, high thermal stability and strong adhesive properties. However, fluoropolymers are of environmental concerns, because their production and disposal can cause PFAS emissions. Additionally, the cathode processing involves N-methyl-2-pyrrolidone (NMP), a toxic solvent needed to dissolve PVDF. While PFAS-free binders are already established for anodes, finding suitable alternatives for cathodes remains significantly more challenging. Despite this, active research efforts and industrial developments are advancing several potential PFAS-free solutions, which include water-soluble polymer binders, novel carbon-based structures, inorganic binders and biobased materials. A few manufacturers have already commercialized PFAS-free binder technologies, or are on the way to do so. These developments indicate that PFAS-free cathode materials are beginning to emerge, but their performance and suitability for large-scale industrial production is still subject to further research.

The electrolyte commonly uses lithium hexafluorophosphate (LiPF_6) as the main salt, which is highly moisture-sensitive and can decompose to form hydrofluoric acid (HF), leading to corrosion of the cell. To mitigate these issues, PFAS-based salts like LiTFSI and fluorinated solvents like fluoroethylene carbonate (FEC) are added, improving cycling performance and stability, while also contributing to non-flammability, thereby enhancing overall battery safety. Fluorine-free electrolyte salts exist, and include perchlorate (ClO_4^-), bis(oxalato)borate (BOB), tris(oxalato)phosphate, tetracyanoborate, and dicyanotriazolate. Their main drawback compared to LiPF_6 is a reduced ability to passivate the aluminium current collector, which is crucial for preventing corrosion from the electrolyte chemicals and, consequently, for extending the lifespan of the LIB (Du et al., 2024). A problem as in **Paper I**, is that most electrolyte formulations are trade secrets, which makes it more difficult to get an overview of what fluorinated compounds are used and if there are viable non-fluorinated alternatives. However, the outlook is that it seems like they are technically capable to adapt towards PFAS-free formulations if the industry demand increases.

Solid-state batteries are a promising next-generation technology and offer opportunities for innovation towards fluorine-free materials. Although they are approaching commercialisation, technical hurdles remain. Many solid and gel electrolytes under development are based on fluorinated polymers like PVDF, although non-fluorinated options such as poly(ethylene oxide) (PEO) also exist and are explored for application. It remains uncertain whether high-performance PFAS-free solid-state batteries will emerge in the near future. Sodium-ion batteries are an advancing alternative technology to LIBs, but research suggests that they may also rely on fluorinated additives. As for LIBs, it will also be essential for sodium-ion batteries to map their PFAS use for better understanding and management of potential environmental consequences.

Fluorinated substances pose a challenge to the recycling processes of LIBs. Formation of HF and other potentially harmful byproducts pose a risk to occupational health, equipment and the environment. High-temperature pyrometallurgy (up to 1600 °C) may destroy most of the PFAS, but industry is shifting more towards hydrometallurgy process that uses lower temperatures and achieves higher yields of the metals. With insufficiently high temperatures there is a risk that PFAS are not fully mineralised, and may therefore be introduced into waste streams. Reducing PFAS or fluorine content could help improve recycling efficiency and safety.

A transition away from PFAS in LIBs is possible but not straightforward, especially with remaining uncertainties regarding costs. Promising alternatives already exist, and with effort from academia and industry but also regulators, it should be possible to realise PFAS-free LIBs.

5. Conclusion and future outlook

The findings of this thesis support the overarching hypothesis that current knowledge substantially underestimates both the use and occurrence of PFAS in consumer products and technologies, largely due to limited publicly available information and constraints of traditional analytical methods. Across the investigated product categories, PFAS use was shown to be more widespread and chemically diverse than suggested by ingredient disclosures or targeted analyses alone. In addition, emerging PFAS, particularly those associated with newer technologies, were found to be widely present in the environment.

Paper I, and in part **Paper II**, confirm previous findings that integrated fluorine analysis, particularly fluorine mass balance, is an effective strategy for assessing a broad range of products with diverse matrices and for capturing the majority of PFAS present. **Paper II** further demonstrates that analytical gaps associated with the widespread use of polymeric PFAS can be partially addressed by Py-GC-MS. However, this approach still requires further methodological development, including the application of softer ionisation techniques such as CI to preserve more structural information from the investigated molecules. In addition, expanding the number of reference materials, covering low-molecular-weight to polymeric fluorinated substances, and establishing spectral databases would improve compound identification. Nevertheless, some products may contain modified or proprietary polymers for which reference materials do not exist, further underscoring the value of softer ionisation methods, combined with a GC column such as a porous layer open tubular (PLOT) column (U.S. EPA, 2025) capable of separating and retaining highly volatile fluorinated substances.

Based on the EU PFAS restriction proposal, **Paper II** contributed to the development of a stepwise approach to support compliance enforcement for consumer products (Vestergren et al., 2024). The workflow consists of three main steps. Initial screening by TF allows rapid identification of products with elevated fluorine content. Where TF exceeds 50 ppm, additional investigation is required to determine the origin of the fluorine. In cases where supply-chain information is unavailable or insufficient, Py-GC-MS (**Paper II**) can be applied to assess whether the fluorine is of organic origin. Products confirmed to contain organofluorine above the threshold would be considered non-compliant with the proposed regulation. For products with low TF or no indication of organofluorine, targeted analysis of selected PFAS (C1-C16 PFAAs) can provide further refined and risk-relevant information. If targeted analysis indicates concentrations above 25 ppb for any individual PFAS or 250 ppb for the sum of PFAS, then the product is non-compliant. In practice, this tiered workflow can guide both regulators and industry in determining whether consumer products meet the forthcoming PFAS restriction. The increasing role of e-commerce and the import of products from outside the EU complicate regulatory oversight, raising questions about how effectively compliance with EU chemical regulations can be ensured in practice.

If PFAS are more prevalent in different products than previously assumed, the question arises as to what happens to them beyond the use phase of the product itself. **Paper III** highlights that PFAS emissions can occur at multiple stages of a product's lifecycle, as illustrated here for LIBs. While legacy PFAS such as PFOA and PFOS have been subject to regulatory and voluntary phase-outs and show declining environmental trends, substitution and innovation has often resulted in the introduction of more complex and novel PFAS chemistries rather than a transition to PFAS-free materials. Consequently, environmental monitoring must extend beyond legacy PFAS and include novel fluorinated compounds, such as TFSI. Additionally, monitoring strategies need to increasingly incorporate advanced analytical approaches, including suspect and non-target screening workflows, to effectively capture the evolving PFAS landscape over time. This is particularly crucial for drinking water surveillance, as these substances do not appear to be sufficiently removed by conventional wastewater treatment processes. Beyond PFAS, the detection of other fluorinated compounds such as PF_6^- and BF_4^- is also of concern, especially as these substances are often claimed by industry to readily hydrolyse in the environment. Their occurrence in drinking water highlights the need for degradation testing under environmentally relevant conditions, as well as

comprehensive (eco)toxicological evaluation. Moreover, these compounds represent only a small fraction of fluorinated substances used in LIBs. Numerous additional compounds and degradation products are likely present but currently remain undetected. Addressing these emissions is therefore complex not only due to the analytical limitations, but also because of limited transparency regarding PFAS use by industry and constraints within existing regulatory frameworks.

A key finding across **Papers I-IV** is the need for improved product labelling and greater transparency. Clearer information is essential for both consumers and professionals to make informed decisions regarding sustainable materials and products. In parallel, regulators face the challenge of defining what constitutes a “PFAS-free” product in a scientifically robust and enforceable manner. At present, manufacturers increasingly label products as “PFAS-free”, often without providing detailed supporting information. Initiatives within the EU, such as the digital product passport (The European Parliament and The Council of the European Union, 2024) and the battery passport (The European Parliament and The Council of the European Union, 2023), aim to improve transparency and access to information on chemical content in products. This is especially important in the context of a circular economy, where recycling and reuse of materials are central, and the reintroduction of hazardous substances should be avoided. However, under the current framework of the battery passport, substance identities may remain vague due to confidentiality and competition concerns. There is therefore a need for improved systems that allow substance identities to be shared, at least to some extent, with the scientific community, or for stronger incentives for companies to conduct and disclose independent studies. Reliance solely on industry-related data, however, carries an inherent risk of bias.

Increased transparency from PFAS users would enable the refinement of targeted and suspect screening lists, ensuring that analytically relevant compounds are monitored and thereby facilitating more effective environmental surveillance. Greater openness would also support analytical chemists in developing suitable methods for emerging substances. Furthermore, the case of LiPF₆ illustrates the need for greater rigor in the preparation and evaluation of substance registration dossiers, as information provided by manufacturers and importers is not always transparent, consistent, or sufficiently substantiated. Ultimately, while preventing PFAS emissions at the source should remain the primary goal, industry engagement in funding monitoring and remediation efforts is also essential. These challenges raise a central question: where PFAS use is problematic, can they be replaced?

Looking ahead, the proposed universal EU PFAS restriction provides an important foundation for reducing PFAS emissions and exposure. However, the results of this thesis indicate that meaningful progress will require a shift towards genuinely PFAS-free or fluorine-free chemistries (**Papers I and IV**), supported by sustained investment in alternative materials and technologies. Developing such alternatives remains challenging, as they must meet demanding technical performance criteria while also ensuring environmental and human health safety. Inevitably, trade-offs will need to be carefully evaluated, underscoring the importance of integrating analytical innovation, lifecycle thinking, and substitution frameworks to guide future decision-making. The identification and implementation of suitable alternatives also depend on transparency from alternative material providers, particularly regarding chemical composition and functional performance. Concepts such as the essential use framework provide a valuable basis for evaluating the necessity of chemicals in specific applications and, when combined with an understanding of chemical function, can support the identification of appropriate substitutes. At the national level, countries such as Denmark and Sweden have established research (“PFAS Center Denmark,” n.d.) and substitution (“The Swedish Centre for Chemical Substitution | RISE,” n.d.) centres dedicated to this purpose. Establishing a similar, centrally coordinated European platform (WSP, 2025) could facilitate knowledge transfer and accelerate substitution efforts across Europe, including in countries with more limited research capacity.

Ensuring inclusivity is critical, as countries facing financial, infrastructural, or political constraints should not be excluded from the transition towards safer chemistries and a cleaner environment. Inclusivity must also extend to industrial sectors: critical industries, including those central to the “green energy transition,” must remain accountable for the materials and chemicals they employ rather than being exempt from responsibility. While society bears long-term environmental and economic consequences of PFAS contamination, decisions regarding their design, use and disclosure are largely made upstream by industry. Addressing the PFAS challenge therefore requires governance frameworks that move beyond downstream monitoring and remediation, and instead place greater responsibility on chemical producers and product manufacturers to prevent emissions at the source. Achieving this will require close collaboration among materials chemists, environmental scientists, regulators, and other domain experts for specific use applications, underscoring the importance of interdisciplinary approaches as a pathway towards addressing the PFAS challenge collectively.

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