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Investigation of the occurrence of PFAS (per- and polyfluorinated alkyl compounds) in waste streams

Final Report

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Investigation of the occurrence of PFAS (per- and polyfluorinated alkyl compounds) in waste streams

Final Report

by

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Kurzbeschreibung: Untersuchung des Vorkommens von PFAS (Per- und polyfluorierte Alkylverbindungen) in Abfallströmen

Zur Abschätzung des Risikos von per- und polyfluorierten Verbindungen (PFAS) für Mensch und Umwelt besteht Forschungsbedarf hinsichtlich ihres Verbleibs in der Umwelt und hier im Besonderen im Hinblick auf ihre Persistenz in Abfallströmen.

Um diese Fragestellung zu adressieren, hat das Umweltbundesamt das Forschungsvorhaben „Untersuchung des Vorkommens von PFAS (Per- und polyfluorierte Alkylverbindungen) in Abfallströmen“ initiiert. Dieses Vorhaben hat das Ziel einer ersten Identifizierung, Quantifizierung und Bewertung des Vorkommens von PFAS in bestimmten Abfallströmen. Die Notwendigkeit dieser Erfassung und des daraus abgeleiteten Forschungsbedarfes ergibt sich direkt aus dem Stockholmer Übereinkommen über persistente organische Schadstoffe (POP), in dessen Anhang A bereits Vertreter aus der Gruppe der PFAS (PFOS, ihre Salze und PFOSF, PFOA ihre Salze und PFOA-verwandte Verbindungen sowie PFHxS, ihre Salze und PFHxS-verwandte Verbindungen) aufgenommen wurden.

Um eine repräsentative Probennahme durchführen zu können, wurde hierzu in einem ersten Schritt die aktuelle Situation anhand der verfügbaren Literatur dargestellt und hieraus relevante Abfallströme identifiziert.

Auf Grundlage dieser Ergebnisse wurde ein Probenplan für die als relevant identifizierten Abfallströme entwickelt und eine gezielte Probennahme vorbereitet und durchgeführt. Anhand der erhaltenen Ergebnisse aus der Literaturrecherche und der Analytik wurden die Risiken für Mensch und Umwelt sowie die Auswirkungen auf die Abfallwirtschaft abgeschätzt, sowie Vorschläge zur Grenzwertsetzung und zu möglichen Entsorgungswegen abgeleitet.

Abstract: Investigation of the occurrence of PFAS (per- and polyfluorinated alkyl substances) in waste streams.

In order to assess the risk of PFAS to humans and the environment, there is a need for research on their fate in the environment and in particular, with regard to their persistence, in waste streams.

To address this issue, the Federal Environment Agency has initiated the research project "Investigation of the occurrence of PFAS (per- and polyfluorinated alkyl substances) in waste streams". This project aims at the initial identification, quantification and evaluation of the occurrence of PFAS in specific waste streams. The necessity of this survey and the research needs derived from it arise directly from the Stockholm Convention on Persistent Organic Pollutants (POPs), in whose Annex A representatives from the group of PFAS (PFOS, their salts and PFOSF, PFOA their salts and PFOA-related compounds as well as PFHxS, their salts and PFHxS-related compounds) have already been included.

In order to be able to carry out a representative sampling, the first step is to describe the current situation based on the available literature and to identify relevant waste streams.

Based on these results, a sampling plan was developed for the waste streams identified as relevant and a targeted sampling was prepared and carried out. Based on the results obtained, which include organic fluorine in the form of the sum parameter (EOF), the risks to humans and the environment as well as the effects on waste management were estimated, and suggestions for setting limit values and possible disposal routes were made.

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Abbreviation	Meaning
CLP-VO	(Regulation (EC) No. 1272/2008 of the European Parliament and of the Council of 16 December 2008 on classification, labeling and packaging of substances and mixtures
10:2 diPAP	10:2 fluorotelomer phosphate diester
11Cl-PF3OUdS	11-Chloroeicosafluoro-3-oxaundecane-1-sulfonic acid
4:2 FTOH	2-(Perfluorobutyl)ethyl alcohol
10:2 FTOH	2-(Perfluorodecyl)ethyl alcohol
6:2 FTOH	2-(Perfluorohexyl)ethyl alcohol
8:2 FTOH	2-(Perfluorooctyl)ethyl alcohol
HFPO-DA	2,3,3-Tetrafluoro-2-(heptafluoropropoxy)propionic acid, its salts and its acyl halides
7:3 FTCA	2H,2H,3H,3H-Perfluorodecanoic acid
H4PFUnDA	2H,2H,3H,3H-Perfluorundecanoic acid
H2PFDA	2H,2H-Perfluorodecanoic acid
5:3 FTCA	4,4,5,6,6,7,7,8,8-undecafluorooctanoic acid
4:2 FTS	4:2 fluorotelomer sulfonic acid
6:2 diPAP	6:2 Fluorotelomer phosphate diester
6:2 / 8:2 diPAP	6:2/8:2 fluorotelomer phosphate diester
6:2 FTS; H4PFOS	6:2-Fluorotelomer sulfonic acid
Cl-PFHxPA	6-Chloroperfluorohexylphosphoric acid (-)
HPFHpA	7H-dodecafluoroheptanoic acid
7H-PFHpA	7H-perfluoroheptanoic acid
8:2 diPAP	8:2 Fluorotelomer phosphate diester
8:2 FTS	8:2 fluorotelomer sulfonic acid
8:2 / 10:2 di PAP	8:2/10:2 fluorotelomer phosphate diester

Abbreviation	Meaning
9Cl-PF3ONS	9-Chlorohexadecafluoro-3-oxanonane-1-sulfonic acid
AOF	Adsorbable organic fluorine
ADONA / H-DONA	Ammonium 2,2,3-trifluoro-3-(1,1,2,3,3-hexafluoro-3-(trifluoromethoxy)propoxy)propanoate
APFO	Ammonium pentadecafluorooctanoate
BVT	Best available techniques (Orig. German: beste verfügbare Techniken)
CAS	Chemical Abstracts Service
ChemG	Chemicals Act (Orig. German: Chemikaliengesetz)
ChemVerbotsV	Chemicals Prohibition Ordinance (Orig. German: Chemikalien-Verbotsverordnung)
KrWG	Circular Economy Act (Orig. German: Kreislaufwirtschaftsgesetz)
CIC	Combustion Ion Chromatography
SCIP	Consumer database on hazardous substances in products (Substances of Concern In articles as such or in complex objects (Products))
DNEL	Derived-no-effect-level (Derived exposure limit below which a substance does not cause adverse effects on human health)
dTOP	Direct TOP assay
WFD	Directive 2000/60/EC establishing a framework for Community action in the field of water policy - Water Framework Directive
diSAmPAP	Di-substituted polyfluoroalkyl phosphate esters of N-ethyl perfluorooctane sulfonamidoethanol.
TM	Dry matter (same meaning as dry substance)
ECHA	European Chemicals Agency
ER	European Council (Orig. German: Europäischer Rat)

Abbreviation	Meaning
EP	European Parliament
EPO	European Patent Office
EU	European Union
EOF	Extractable organic fluorine
UBA	Federal Environmental Agency (Orig. German: Umweltbundesamt)
BImSchG	Federal Immission Control Act (Orig. German: Bundes-Immissionsschutzgesetz)
BfR	Federal Institute for Risk Assessment (Orig. German: Bundesinstitut für Risikobewertung)
BMUV	Federal Ministry for the Environment, Nature Conservation, Nuclear Safety and Consumer Protection (Orig. German: Bundesministerium für Umwelt, Naturschutz, nukleare Sicherheit und Verbraucherschutz)
BMBF	Federal Ministry of Education and Research (Orig. German: Bundesministerium für Bildung und Forschung)
StBA	Federal Statistical Office (Orig. German: Statistisches Bundesamt)
AFFF	Fire extinguishing foams (from aqueous film forming foam)
LFGB	Food and Feed Code (Orig. German: Lebensmittel- und Futtermittelgesetzbuch)
AbfRRL	Framework Directive 2008/98/EC on Waste (Orig. German: Rahmenrichtlinie 2008/98/EG über Abfälle)
GrwV	Groundwater Ordinance (Orig. German: Grundwasserverordnung)
LW	Health guide (Orig. German: Gesundheitlicher Leitwert)
GOW	Health orientation values (Orig. German: Gesundheitliche Orientierungswerte)
N-MeFOSA	Heptadecafluoro-N-methyloctanesulfonamide
HR-CS-GFMAS	High resolution continuum source-graphite furnace-molecular absorption spectroscopy
HPLC	High-performance liquid chromatography
IED	Industrial Emissions Directive 2010/75/EU
DepV	Landfill Ordinance (Orig. German: Deponieverordnung)
LPCL	Low POP concentration limit
MS	Mass spectrometry
6:2 PAP	Mono[2-(perfluorohexyl)ethyl] phosphate

Abbreviation	Meaning
EtFOSAA	N-Ethylperfluorooctanesulfonamidoacetic acid
N-EtFOSAA	N-Ethylperfluorooctanesulfonamidoacetic acid
N-MeFOSAA	N-methylperfluoro-1-octanesulfonamidoacetic acid
NGO	Non-governmental organization (NGO)
GefStoffV	Ordinance on Hazardous Substances (Orig. German: Gefahrstoffverordnung)
DümV	Ordinance on the Marketing of Fertilizers, Soil Additives, Cultivation Substrates and Plant Auxiliaries (Fertilizer Ordinance) (Orig. German: Verordnung über das Inverkehrbringen von Düngemitteln, Bodenhilfsstoffen, Kultursubstraten und Pflanzenhilfsmitteln (Düngemittelverordnung)
POP Waste Monitoring Ordinance	Ordinance on the Separate Collection and Monitoring of Non-hazardous Waste Containing Persistent Organic Pollutants
OECD	Organisation for economic co-operation and development
PFAS	Per- and polyfluorinated alkyl substances
PFAA	Perfluoroalkyl acids, perfluoro acids (perfluorocarboxylic and -sulfonic acids)
PFBS	Perfluorobutanesulfonic acid
PFBA	Perfluorobutanoic acid
PFCA	Perfluorocarboxylic acids
PFDS	Perfluorodecane sulfonic acid
PFDA	Perfluorodecanoic acid
8:2 PAP	Perfluorodecyl phosphate
PFDoDS	Perfluorododecane sulfonic acid
PFDoDA	Perfluorododecanoic acid
PFHpS	Perfluoroheptane sulfonic acid
PFHpA	Perfluoroheptanoic acid
PFHxDA	Perfluorohexane decanoic acid
PFHxS	Perfluorohexane-1-sulfonic acid and its salts
PFHxA	Perfluorohexanoic acid
PFNS	Perfluoronon sulfonic acid
PFNA	Perfluorononanoic acid
PFOSA	Perfluorooctane sulfonamide
SAmPAP	Perfluorooctane sulfonamidoethanol-based phosphate esters

Abbreviation	Meaning
PFOS	Perfluorooctane sulfonic acid
PFODA	Perfluorooctanedecanoic acid
FOSAA	Perfluorooctanesulfonamidoacetic acid
PFOA	Perfluorooctanoic acid
PFPS	Perfluoropentanesulfonic acid
PFPeA	Perfluoropentanoic acid
PFECHS	Perfluoro-p-ethylcyclohexylsulfonic acid
PFPA	Perfluorophosphoric acids
PFPrA	Perfluoropropanoic acid
PFSA	Perfluorosulfonic acids
PFSA	Perfluorosulfonic acids
PFTeDA	Perfluorotetradecanoic acid
PFTrDA	Perfluorotridecanoic acid
PFTrDS	Perfluorotridecansulfonic acid
PFUnDS	Perfluorundecane sulfonic acid
PFUnDA	Perfluorundecanoic acid
POP	Persistent organic pollutants (POPs)
PTFE	Polytetrafluoroethylene
PNEC	predicted no effect concentration
RDF	Refuse Derived Fuel, Substitute fuel (Orig. German: Ersatzbrennstoff, EBS)
REACH-VO	Regulation (EC) No. 1907/2006 concerning the Registration, Evaluation, Authorization and Restriction of Chemicals
POP-VO	Regulation (EU) 2019/1021 on persistent organic pollutants
AbfKlärV	Sewage Sludge Ordinance (Orig. German: Klärschlammverordnung)
SPE	Solid phase extraction
SC	Stockholm Convention
SVHC	Substance of very high concern
SPIN	Substances in Products in the Nordic Countries

Abbreviation	Meaning
N-EtFOSA	Sulfluramide
OGewV	Surface Water Ordinance (Orig. German: Oberflächengewässerverordnung)
TWI	Tolerable weekly intake
TFAA	Trifluoroacetic acid
EPA	U.S. Environmental Protection Agency
UTC	Unintentional trace contamination
USPTO	United States Patent and Trademark Office
UPCL	Upper POP concentration limit
AVV	Waste Catalogue Ordinance (Orig. German: Abfallverzeichnisverordnung)
WHG	Water Resources Act (Orig. German: Wasserhaushaltsgesetz)
WHO	World Health Organization

Zusammenfassung

Hintergrund und Ziele

In den vergangenen Jahren hat die Bedeutung von per- und polyfluorierten Alkylverbindungen (PFAS) stetig zugenommen. Die Produktionsmenge an PFAS folgte in den vergangenen Jahrzehnten einem stark steigenden Trend. PFAS sind Wasser, Schmutz und Fett abweisend. Sie werden weltweit verwendet und man findet sie in zahllosen Produkten, von der Outdoorjacke über die Teflonpfanne bis hin zu Feuerlöschschäumen (UBA, 2020a). Da PFAS so vielfältig verwendet werden, gibt es auch viele Wege, wie sie in die Umwelt gelangen können: Bei der Herstellung der Chemikalien selbst, ihrer Weiterverarbeitung zu Erzeugnissen, beim Gebrauch der Produkte und schließlich bei und nach der Entsorgung.

Mit der steigenden Bedeutung dieser Stoffgruppe, die hauptsächlich auf ihren funktionalen Eigenschaften für viele technische Prozesse beruht, steigt auch die Freisetzung in die Umwelt. PFAS können sich über die Luft, Flüsse und Meere bis in entlegene Gebiete wie die Arktis verteilen. Sie sind kaum abbaubar und bleiben daher für einen sehr langen Zeitraum in der Umwelt. Einige PFAS reichern sich in Tieren, Pflanzen und im Menschen an und wirken zudem gesundheitsschädigend. Erhöhte Konzentrationen von PFOA und PFOS im menschlichen Blut können Wirkungen von Impfungen vermindern, die Neigung zu Infekten erhöhen, zu erhöhten Cholesterinwerten führen und bei Nachkommen ein verringertes Geburtsgewicht zur Folge haben (UBA, 2020a).

Aufgrund ihrer Eigenschaften ist die Abschätzung des Risikos von PFAS für Mensch und Umwelt notwendig und es besteht Forschungsbedarf hinsichtlich ihres Verbleibs in der Umwelt und hier im Besonderen im Hinblick auf ihre Persistenz sowie Eintragsmengen aus Abfallströmen. Um diese Fragestellung zu adressieren hat das Umweltbundesamt das Forschungsvorhaben „Untersuchung des Vorkommens von PFAS (Per- und polyfluorierte Alkylverbindungen) in Abfallströmen“ initiert.

Dieses Vorhaben hat das Ziel einer ersten Identifizierung, Quantifizierung und Bewertung des Vorkommens von PFAS in Abfallströmen. Weiterhin soll eine Grundlage für weitere Forschungsschwerpunkte entstehen und Handlungsbedarf abgeleitet werden. Die Notwendigkeit der Erfassung von PFAS in Abfällen ergibt sich direkt aus dem Stockholmer Übereinkommen über persistente organische Schadstoffe (POP), in dessen Anhang A (Eliminierung) bereits Vertreter aus der Gruppe der PFAS (PFOS, ihre Salze und PFOSF, PFOA ihre Salze und PFOA-verwandte Verbindungen sowie PFHxS, ihre Salze und PFHxS-verwandte Verbindungen) aufgenommen wurden.

Herangehensweise/Methodik

Um die Projektziele zu erreichen wurden zunächst, basierend auf einer Hintergrundrecherche (siehe Kapitel 2.1und 3.1), relevante Abfallströme identifiziert, ein Probenplan entwickelt und eine gezielte Probennahme und anschließende chemisch-physikalische Analytik durchgeführt, bei der das organische Fluor in Form des Summenparameters (EOF) erfasst wird (siehe Kapitel 2.2, 2.3, 2.4 und 3.2).

Auf Grundlage der Messergebnisse und den daraus abzuleitenden PFAS-Konzentrationen (siehe Kapitel 3.2) sollten PFAS-Massenströme in den jeweiligen Abfallströmen errechnet, und darauf aufbauend, Betrachtungen zu möglichen Entsorgungswegen sowie zu möglichen PFAS-Grenzwerten im Bereich des Abfallrechts angestellt werden. Allerdings konnten aus den PFAS-Messungen der Proben keine Konzentrationen für Einzel-PFAS-Substanzen erhalten werden, sondern nur Summenparameter (siehe Kapitel 3.2). Damit fehlte die Grundlage, um PFAS-Massenströme zu errechnen und für darauf aufbauende Fragestellungen.

Um die Massenströme ausrechnen und die Abfallgrenzwerte herleiten zu können, wurde deshalb in Abstimmung mit dem Umweltbundesamt entschieden, geeignete Literaturwerte zu recherchieren und auszuwerten. Hierfür wurde eine zusätzliche Literaturrecherche für die ausgewählten Abfallströme (Papier, Klärschlamm, Böden und Textilien) durchgeführt (siehe Kapitel 2.5 3.3). Die gefundenen Werte wurden extrahiert, gemittelt und verwendet, um die Massenströme (siehe Kapitel 2.7 und 3.4) zu ermitteln.

Zur Abschätzung der Umweltrelevanz wurden die ausgewählten Abfallströme in einen Umweltkontext gebracht (siehe Kapitel 2.8 und 3.5) und Betrachtungen zu möglichen Grenzwerten (siehe Kapitel 2.9 und 3.6) angestellt.

Auf Grundlage der Ergebnisse wurden Empfehlungen zur Etablierung möglicher abfallwirtschaftlicher Grenzwerte abgeleitet (siehe Kapitel 3.6) sowie Handlungsanweisungen für die Öffentlichkeit und den abfallwirtschaftlichen Vollzug formuliert (siehe Kapitel 3.7 und 3.8).

Analysen und Analysenergebnisse

Aus der durchgeführten Literaturrecherche wurden in Absprache mit dem UBA die folgenden Abfallströme als relevant identifiziert:

- Textilien
- Klärschlämme
- Papier
- Böden
- Baumarktprodukte, die für den Außenbereich vorgesehen sind (z.B. Farben)

Entsprechend dieser vorgenommenen Einschätzung wurden Proben aus diesen Bereichen für die EOF-Untersuchungen von der BAM genommen. Im Falle der Textilien wurden Proben untersucht, die der Altkleidersammlung zugeführt wurden. Klärschlämme wurden von Kläranlagenbetreibern angefragt mit dem besonderen Augenmerk auf industrielle Kläranlagen aus dem Papiersektor, sowie kommunale Kläranlagen, bei denen ein bekannter Einleiter aus dem Bereich der PFAS einsetzenden Betriebe bekannt ist (z.B. Galvanikbetriebe). Das untersuchte Papier stammt aus einer Altpapiersortieranlage. Die Böden wurden von verschiedenen Betreibern von Deponien, Landesuntersuchungsämtern oder vor Ort selbst genommen. Die untersuchten Farben und andere Produkte für die Anwendung im Außenbereich wurden als einzige in einem nicht gealterten Zustand, sondern neuen Zustand, untersucht. Die Untersuchung aller Proben erfolgte mittel HR-CS-MAS hinsichtlich des Summenparameters EOF nach geeigneter Probenvorbereitung, Extraktion und Aufreinigung.

Textilen

Die Untersuchung der Textilien erfolgte nach dem Zerkleinern und Homogenisieren der Proben. Der Probenumfang hierbei beträgt zehn unterschiedliche Jacken, zwei Taschen und einen Teppich die an verschiedenen Tagen aus der Altkleidersammlung entnommen wurden. Der Gesamtprobenumfang entspricht somit 13 Proben. Die Untersuchung der Jacken zeigte dabei einen EOF-Gehalt im Bereich von 37 µg/kg bis zu einem EOF-Gehalt von 1163 µg/kg. In einer Tasche konnte kein EOF nachgewiesen werden und für die zweite Tasche wurde ein Gehalt von 195 µg/kg ermittelt. Auch bei dem Teppich konnte ein EOF-Gehalt von 83 µg/kg nachgewiesen werden. Diese Spanne, die zwei Größenordnungen abdeckt, zeigt, dass es sich bei Alttextilien um einen stark heterogenen Abfallstrom handelt.

Klärschlämme

Die in dieser Studie untersuchten Klärschlämme wurden alle durch den Betreiber der jeweiligen Kläranlagen entnommen und vor Ort gefriergetrocknet. Es wurden insgesamt 17 Klärschlammproben in dieser Studie betrachtet. Beprobt wurden zum einen Kläranlagen industrieller Art, die mit PFAS-haltigen Produkten assoziiert sind, und zum anderen kommunale Kläranlagen, deren Klärschlämme bereits in der Vergangenheit erhöhte PFAS-Werte aufwiesen.

Bei den industriellen Kläranlagen ließ sich im Klärschlamm ein maximaler EOF-Gehalt von ca. 5000 µg/kg ermitteln. Bei den Klärschlämmen aus kommunalen Kläranlagen ließ sich ein EOF-Gehalt von bis zu ca. 450 µg/kg feststellen. Dieser Wert liegt über dem von typischen Target-PFAS-Untersuchungen und zeigt, dass eine Unterbewertung mittels Target-Analysemethoden auftreten kann. Auch zeigt sich, dass ein Eintrag von organischem Fluor in die Kläranlagen sowie eine Anreicherung in der Schadstoffsenke Klärschlamm stattfindet.

Papier

Die untersuchten Proben wurden aus einer Altpapiersortieranalage entnommen und hinsichtlich ihres EOF-Gehaltes untersucht. Dabei wurden zwei Fraktionen entnommen. Zum einen grafisches Papier und zum anderen Verpackungskartonage. Die Proben wurden zerkleinert und homogenisiert. Die anschließende Untersuchung ergab für die Papierprobe einen EOF-Wert von ca. 120 µg/kg und für die Verpackungskartonage einen Wert von ca. 240 µg/kg. Dieses zeigt deutlich, dass der recycelte Rohstoff Papier und Kartonage einen Grundgehalt an PFAS hat, der im Rahmen der Wiederverwendung in neue Produkte verschleppt werden kann und aus diesen oder während des Prozessierens in die Umwelt gelangen kann.

Böden

In dieser Studie wurden neun Böden unterschiedlicher Herkunft nach Gefriertrocknung untersucht. Dabei wurden Böden ohne bekannte Belastung sowie Böden aus bekannten kontaminierten Regionen untersucht. Die vorbelasteten Böden zeigen eine EOF-Belastung von bis zu 3 mg/kg. Die restlichen Böden zeigen alle eine EOF-Belastung (< 0,25 mg/kg), die jedoch unabhängig von ihrem Probenahmeort sehr ähnlich sind. Dies deutet darauf hin, dass eine messbare Hintergrundbelastung mit fluorierten organischen Verbindungen besteht.

Baumarktprodukte

Produkte, die im Außenbereich eingesetzt werden, sind häufig mit PFAS versetzt, um Ihnen wasser- und schmutzabweisende Eigenschaften zu verleihen. Aus diesem Grund wurden in dieser Studie verschiedene Baumarktprodukte untersucht, die für den Außenbereich vorgesehen sind. Dabei wurden fünf Farben und drei Dichtungsmaterialien untersucht. Die Dichtungsmaterialien weisen keinen nachweisbaren EOF-Gehalt auf. Bei den untersuchten Farben konnte hingegen in allen Proben PFAS nachgewiesen werden. Die Spanne des EOF-Gehaltes lag zwischen 40 µg/kg und 180 µg/kg. Die so erhaltenen Daten legen nahe, dass Farben für den Außenbereich PFAS enthalten, die möglicherweise nach ihrer Applikation ausgewaschen werden und in die Umwelt gelangen können. Des Weiteren kann es durch die Entsorgung von Resten sowie der entsprechend behandelten Flächen zu einer Belastung des entsprechenden Abfallstroms kommen.

Trotz der Identifizierung zahlreicher „Hotspot“-Proben konnte in keiner der Proben mittels Target-Analytik ein belastbarer Wert für eine Einzelsubstanz gemessen werden. Dies zeigt deutlich, wie wichtig das Screening von Proben ist, um eine Einschätzung der Belastung zu möglichen. Auch legt dies nahe, dass weitere Methoden entwickelt werden müssen, um den Bedarf an der Messung von Zielanalyten decken und somit eine Abschätzung der potenziell ausgehenden Gefahr von Abfällen und Produkten treffen zu können.

PFAS-Konzentrationen in den ausgewählten Abfallströmen

Da aus den in diesem Projekt gemessenen Proben keine Messwerte für einzelne PFAS erhalten werden konnten, wurde eine weitere Literaturrecherche zu PFAS-Messdaten in den Abfallströmen Textilien, Klärschlämme Papier und Böden durchgeführt. Für diese wurden PFAS-Literaturdaten mit Fokus auf deutschen Proben recherchiert, extrahiert und aus den erhaltenen Daten Mittel- und Medianwerte gebildet. Diese Werte sind wegen verschiedener Einschränkungen mit einer gewissen Unsicherheit belastet. Die Einschränkungen ergeben sich insbesondere aus (1) der unterschiedlichen Anzahl an analysierten PFAS in den identifizierten Studien, (2) einer Verzerrung der Ergebnisse, weil häufig gezielt Verdachtsfälle analysiert werden (außer bei Böden wurden keine oder nur sehr wenige Hintergrundmessungen vorgenommen bzw. identifiziert) und (3) Abweichungen, die sich aus der Berechnung der Summenkonzentrationen ergeben. Die Daten sollten unter Berücksichtigung der Einschränkungen interpretiert werden.

Textilien

PFAS werden in Textilien eingesetzt, um diesen wasser- und fettabweisende Eigenschaften zu verleihen. Besonders hohe PFAS-Konzentrationen wurden in Markisen und in Outdoorjacken gefunden, welche von der Verwendung von Fluortelomeralkoholen stammen. Table 8 fasst die errechneten Mittel- und Medianwerte zusammen.

Tabelle 1: PFAS-Mittel- und Medianwerte in den gefundenen Textilproben. Alle Daten in µg/kg.

Substanz	Mittelwert	Median
∑PFCA	87,56	8,36
∑PFSA	20,65	0,00
∑PFPA	0,00	0,00
∑Präkursoren	280,70	3,46
∑PFAS	**388,91**	**74,05**

Die gemittelten PFAS-Werte für die Textilien stimmen gut mit den in diesem Vorhaben beprobten Ergebnissen für Textilien überein.

In einem aktuellen Bericht von Wood, (2020) werden Konzentrationen von 0,025-0,05% für Leder (250-500 mg/kg), 0,03% für synthetische Teppiche (300 mg/kg) und 2-3% für Textilien und Möbel (20.000-30.000 mg/kg) angegeben. Die Konzentrationen liegen dabei oberhalb der hier ausgerechneten Konzentrationen in Table 8, was daran liegen kann, dass die Konzentrationen von Wood (2020) auf den durchschnittlichen PFOS-Konzentrationen in Textilien aus UNEP (2017) basieren.

Ähnliche Konzentrationen werden ebenfalls im gesamt-PFAS-Beschränkungsverfahren angegeben. Diese schwanken zwischen <0,1% und 7% je nach Anwendung. Manche Textilien bestehen zu 100% aus PFAS wie z.B. PTFE Membranen (Annex XV, 2023a). Generell liegen auch hier die Konzentrationen höher als die ausgerechneten Werte in Table 8, was daran liegen kann, dass das Beschränkungsverfahren alle PFAS umfasst (auch Polymere) und nicht nur die kurzkettigen Säuren und einige Vorläufer wie in diesem Vorhaben.

Um die tatsächliche Belastung von Textilien und deren Abfällen mit PFAS möglichst repräsentativ darstellen zu können, sollten gezielte Messungen von verschiedenen Textilien durchgeführt werden, welche dann extrapoliert werden können.

Klärschlamm

Klärschlamm fällt in Kläranlagen an, in denen kommunale und/oder industrielle Abwässer behandelt werden. Zu den wichtigsten PFAS-Punktquellen gehören Abwässer aus Industrien, die PFAS herstellen oder in ihren Prozessen verwenden, wie etwa die Papier-, Textil-, Metallbeschichtungs- und Halbleiterindustrie. Da PFAS jedoch auch in einer Vielzahl von Konsumgütern und Haushaltsanwendungen verwendet werden, sind sie auch im rein kommunalen Abwasser zu finden, z. B. durch das Auswaschen aus Textilien.

Die PFAS reichern sich in den Kläranlagen zum Teil im Klärschlamm an oder sie werden zum anderen Teil über den Ablauf in Oberflächengewässer eingeleitet.

Um einen Überblick über die PFAS-Konzentrationen in deutschem Klärschlamm zu erhalten, wurde eine Vielzahl von Klärschlammproben aus unterschiedlichen Quellen analysiert. Es wurden nur wenige Daten zu industriellen Kläranlagen identifiziert. Solche Anlagen weisen höhere PFAS-Konzentrationen auf als kommunale Anlagen. Die wenigen Daten zu industriellen Kläranlagen wurden bei der Berechnung der PFAS-Konzentrationswerte nicht betrachtet. Auf der Grundlage der ermittelten Daten für kommunale Anlagen wurden die in Table 9 dargestellten Mittel- und Medianwerte berechnet.

Tabelle 2: PFAS-Mittel- und Medianwerte in den gefundenen Klärschlammproben. Alle Daten in µg/kg.

Substanz	Mittelwert	Median
∑PFCA	18,03	16,60
∑PFSA	48,80	31,30
∑PFPA	0,00	0,00
∑Präkursoren	0,36	0,00
∑PFAS	**67,18**	**47,40**

Eine Langzeitstudie (von 2008-2013) zeigte, dass die PFAS-Konzentrationen im Klärschlamm sowohl für regulierte als auch für nicht regulierte Stoffe insgesamt abnahmen. Aktuelle Trends könnten durch neue Messungen ermittelt werden.

Die in Table 9 errechneten PFAS-Mittel- und Medianwerte können mit den in diesem Vorhaben gemessenen EOF-Werten verglichen werden. Während in industriellen Kläranlagen EOF-Werte bis zu 5.000 µg/kg nachgewiesen werden konnten, liegen die PFAS-Konzentrationen aus den kommunalen Kläranlagen zumeist in einem Bereich unter 500 µg/kg (Mittelwert 338 µg/kg). Die errechneten Werte aus Table 9 liegen leicht unterhalb dieser Werte.

Die errechneten Werte liegen in einem ähnlichen Bereich wie Daten, die im gesamt PFAS-Beschränkungsdossier im Februar 2023 veröffentlich wurden (Annex XV, 2023b).

Papier

PFAS werden insbesondere in Lebensmittelkontaktpapieren eingesetzt, um dem Papier wasser- und fettabweisende Eigenschaften zu verleihen. Da Papiere in Deutschland zum Großteil recycelt werden, können die PFAS jedoch auch in anderen Papieren vorkommen. Dies konnte auch durch die Altpapier- und Altkartonanalysen (grafische Papiere und Kartonverpackungen) in diesem Vorhaben bestätigt werden (siehe Kapitel 2.2.4 und 3.2.2).

Generell ist der Begriff der Lebensmittelkontaktpapiere sehr weit gefasst. Hierzu gehören z.B. Burger-Papierverpackungen, Papiertüten beim Bäcker, Backpapier, Pappteller, Papierstrohhalme und Pizzaboxen jedoch auch Papier- und Pappeverpackungen von z.B. Tiefkühlware. Je nach Anwendung werden mehr oder weniger PFAS verwendet.

Auf der Grundlage der ermittelten Daten für überwiegend Lebensmittelkontaktpapiere wurden die in Table 10 dargestellten Mittel- und Medianwerte berechnet.

Tabelle 3: PFAS-Mittel- und Medianwerte in den gefundenen Papierproben. Alle Daten in µg/kg.

Substanz	Mittelwert	Median
∑PFCA	155,87	0,60
∑PFSA	1,48	0,70
∑PFPA	0,00	0,00
∑Präkursoren	116,47	0,00
∑PFAS	**273,81**	**10,40**

Im Rahmen des gesamt-PFAS-Beschränkungsverfahrens werden Fluorkonzentration in Lebensmittelkontaktpapiern von 537 mg/kg (Mittelwert) und 1.200 mg/kg (Maximum) angegeben. Diese Werte beziehen sich nur auf das Fluor, jedoch bestehen PFAS-Moleküle auch noch aus anderen Atomen wie z.B. Kohlenstoff und Sauerstoff. Die Ersteller des Dossiers haben deshalb angenommen, dass das Fluor ca. 50% des Gesamtgewichtes der PFAS-Moleküle ausmacht (vgl. PFHxA ~66% Fluoranteil und Seitenkettenfluorierte PFAS ~12,8% Fluoranteil) (Annex XV, 2023a). Somit ergeben sich PFAS-Konzentrationen von 1.074 mg/kg (Mittelwert) und 2.400 mg/kg (Maximum) in Lebensmittelkontaktpapieren. Diese Werte sind bedeutend höher als die in Table 10 errechneten Werte, was daran liegen kann, dass im Beschränkungsdossier alle PFAS mitbetrachtet werden (Säuren, Polymere, seitenkettenfluorierte Polymere etc.), während hier nur die kurzkettigen Säuren sowie einige Vorläufer betrachtet werden.

Um die tatsächliche Belastung von Textilien und deren Abfällen mit PFAS möglichst repräsentativ darstellen zu können, sollten gezielte Messungen von verschiedenen Papieren durchgeführt werden.

Böden

In Böden werden PFAS nicht absichtlich eingesetzt, jedoch finden sie auch Anwendung in PFAS-haltigen Produkten wie z.B. Pflanzenschutzmitteln. In den meisten Fällen treten PFAS als unbeabsichtigte Verunreinigung in Böden auf, beispielsweise durch das frühere Aufbringen von PFAS-kontaminierten Klärschlämmen oder Papierabfällen, die Komposten beigemischt wurden. Eine weitere Quelle für Verunreinigungen in Böden ist die Verwendung von PFAS-haltigen Feuerlöschschäumen sowie die industrielle Herstellung von PFAS und den damit verbundenen Emissionen u.a. in die Luft und anschließender Deposition auf den Böden.

Der Datensatz zu PFAS in Böden umfasst bei Weitem die meisten literaturbasierten Proben in diesem Vorhaben mit etwa 8.000 individuellen Messungen. Der überwiegende Teil der Proben wurden bei Verdachtsfällen genommen, jedoch wurden auch einige Hintergrundmessungen vorgenommen. In vielen der Verdachtsfälle konnten jedoch keine PFAS nachgewiesen werden, da die Messwerte unterhalb des Detektionslimits liegen (Detektionslimit: 1µg/kg).

Auf Grundlage aller identifizierten Werte für Böden wurden die in Table 11 dargestellten Mittel- und Medianwerte berechnet.

Tabelle 4: PFAS-Mittel- und Medianwerte in den identifizierten Bodenproben. Alle Proben aus Rastatt wurden miteinbezogen. Alle Daten in µg/kg.

Substanz	Mittelwert	Median
∑PFCA	11,90	0,14
∑PFSA	3,50	0,00
∑PFPA	0,00[1]	0,00
∑Präkursoren	2,14	0,00
∑PFAS	**17,53**	**0,21**

1 Die Konzentration für PFPA ist hier nicht null, jedoch statistisch gesehen irrelevant

Da der größte Teil der Daten aus dem Landkreis Rastatt stammt, sind diese Daten nicht zwangsläufig repräsentativ für ganz Deutschland.

Zusätzlich konnten Vergleichswerte aus Nordrhein-Westphalen erhalten werden. In unbelasteten Ackerböden konnten ∑ PFAS-Werte von bis zu ~7,6 µg/kg nachgewiesen werden. Die meisten Werte lagen jedoch unterhalb von 1 µg/kg. Diese Konzentrationen setzen sich größtenteils aus PFOS, PFOA und PFBA zusammen. Der errechnete Mittelwert im Bereich von etwa 18 µg/kg in Table 11 stimmt gut mit diesen Werten überein, wenn bedacht wird, dass dieser Wert sowohl belastete als auch unbelastete Boden beinhaltet.

Zuletzt können die errechneten PFAS-Konzentrationen aus Table 11 mit den in diesem Vorhaben gemessenen EOF-Werten verglichen werden. In den gemessenen belasteten Böden konnten EOF-Werte von bis zu 3.000 µg/kg nachgewiesen werden (Mittelwert 1.249 µg/kg). In den unbelasteten Böden schwanken die EOF-Werte zwischen 73 und 209 µg/kg und liegen somit höher als die errechneten Werte.

Abfalldaten und PFAS Flüsse in den ausgewählten Abfallströmen

Auf Grundlage der recherchierten Daten zu den ausgewählten Abfallströmen und den ermittelten PFAS-Konzentrationen als Mittel- und Medianwerte, werden die PFAS-Frachten in den Abfallströmen eingeschätzt. Dargestellt sind die Frachten für die Perfluorcarbon-, -sulfon und -phosphorsäuren, die Präkusoren sowie ein Summenwert für alle PFAS.

Der größte Teil der Textilabfälle in Deutschland wird recycelt (69%) und thermisch behandelt (ca 28 %). Knapp 3% werden auf sonstige Weise entsorgt.

Der größte Teil des Klärschlamms aus kommunalen Kläranlagen in Deutschland wird durch thermische Behandlung (77%) und bodenbezogene Verwertung (22%) verwertet. Etwa 1% wird auf andere Weise entsorgt (z.B. Kompostierung/Vergärung).

Es konnten keine schlüssigen Daten bezüglich des Altpapierabfallstroms gefunden werden. Für dieses Vorhaben wird eine Quelle der Papierindustrie für das Jahr 2020 verwendet und eine stoffliche Verwertungsquote von 80% angenommen. Für die restlichen 20% wird überwiegend energetische Verwertung (ca. 19,7%) sowie ein kleiner Anteil Deponierung (ca. 0,3%) angenommen.

Von 128,9 Millionen Tonnen Abfallböden die in 2018 in Deutschland anfielen, wurden 14,95 % beseitigt, während 85,05 % verwertet wurden. Der größte Teil des beseitigten Abfalls wurde deponiert (14,89 %), d. h. er wurde in/auf einer Deponie/einer speziell angelegten Deponie abgelagert oder dauerhaft umschlossen gelagert, z. B. in einem Bergwerk.. Nur ein kleiner Teil wurde verbrannt (0,02 %) oder, wenn die Kontamination in Form von flüssigen oder schlammigen Abfällen vorliegt, biologisch abgebaut (0,04 %). Was die Verwertung betrifft, so wurde der größte Teil des Bodens (68,87 %) durch Verfüllung verwertet. 16,17 % wurden durch Recycling verwertet, d. h. durch die Wiederaufbereitung von organischem Material, mit dem Ziel es wie ursprünglich vorgesehen zu verwenden. Nur ein kleiner Teil des kontaminierten Bodens (0,01 %) wurde energetisch verwertet.

In Table 12 und Table 13 sind die jährlichen PFAS-Massenströme in den betrachteten Abfallströmen zusammengefasst. Dabei wird zwischen der Art der Behandlung unterschieden, sodass die jährlichen Mengen an recycelten, deponierten und sonstig verwerteten (z.B. verfüllten) PFAS dargestellt ist.

Tabelle 5: Zusammenfassung der errechneten PFAS-Massenströme aus dem Mittelwert der ausgewählten Abfallströme. Alle Angaben in kg.

Abfallstrom	Gesamt PFAS-Menge	Menge PFAS recycelt/stofflich verwertet	Menge PFAS energetisch verwertet	Menge PFAS deponiert/sonstiges
Textilien	758,71	523,74	215,20	19,78
Klärschlamm	116,94	26,13	89,69	1,12
Papier	3.961,46	3.169,17	780,13	12,16
Böden	2260,37	1922,22	0,55	337,60
Gesamt	**7.097,48**	**5.641,26**	**1.085,57**	**370,66**

Wird der Mittelwert in Table 12 betrachtet, so sind in den analysierten Abfallströmen insgesamt rund 7.100 kg PFAS enthalten. Mehr als die Hälfte davon (~56 %) stammt aus Papierabfällen, während nur ~1,6% aus Klärschlamm stammen. Die hohe PFAS-Fracht bei den Papierproben wird allerdings als nicht realistisch angesehen, da nur PFAS-Messwerte aus Lebensmittelkontaktpapier gefunden werden konnten. Viele Papiere im Papierabfallstrom sind jedoch nicht oder nur teilweise mit PFAS belastet (z.B. Zeitungen, Bücher etc.), wodurch die PFAS-Fracht vermutlich überschätzt wird. Die hohen PFAS-Mengen der Bodenproben können auf die vergleichsweise sehr großen Mengen an erfassten Abfallböden zurückgeführt werden (~130.000.000 t im Vergleich zu ~17.000.000 t Altpapier).

Tabelle 6: Zusammenfassung der errechneten PFAS-Massenströme aus dem Medianwert der ausgewählten Abfallströme. Alle Angaben in kg.

Abfallstrom	Gesamt PFAS-Menge	Menge PFAS recycelt/stofflich verwertet	Menge PFAS energetisch verwertet	Menge PFAS deponiert/sonstiges
Textilien	144,47	99,73	40,98	3,77
Klärschlamm	82,50	18,43	63,28	0,79
Altpapier	150,47	120,37	29,63	0,46
Böden	26,54	22,57	0,01	3,96
Gesamt	**403,98**	**261,1**	**133,9**	**8,98**

Werden die Medianwerte in Table 13 betrachtet, so sind in den untersuchten Abfallströmen insgesamt~400 kg PFAS enthalten.

Für die weiteren Fragestellungen des Vorhabens werden die Daten mit Bezug zu den Mittelwerten betrachtet. Sie sind also diesbezüglich als Worstcase-Szenario zu interpretieren.

Betrachtungen und Empfehlungen zu Abfallgrenzwerten für PFAS

Zur Ableitung von abfallwirtschaftlichen Grenzwerten wurde eine bestehende Methode zur Ableitung von POP-Abfallgrenzwerten herangezogen. Die Methodik begrenzt dabei den Konzentrationsbereich eines möglichen Grenzwertes einer Substanz nach unten und oben anhand verschiedener Kriterien. Bei der Anwendung der Methode auf die Substanzgruppe der PFAS ergaben sich jedoch einige Schwierigkeiten und Herausforderungen.

Auswahl der zu untersuchenden PFAS

Eine wesentliche Herausforderung ist dabei die Frage, welche PFAS bei der Grenzwertableitung betrachtet werden sollen. Es wird eine Methode benötigt, durch welche die Vorläufersubstanzen mit in die Grenzwertbetrachtung einbezogen werden können, ohne sie als Einzelsubstanzen zu messen.

Aufgrund der großen Diversität der PFAS kann nicht jede Substanz einzeln analytisch nachgewiesen werden, wodurch die gesamte PFAS-Fracht oft unbekannt ist. Besonders bei den Vorläuferverbindungen ist die Diversität sehr hoch. Um diese Verbindungen mitbetrachten zu können wird deshalb vorgeschlagen das TOP-Assay anzuwenden. Dabei werden die Vorläuferverbindungen größtenteils zu den analogen Perfluorcarbonsäuren oxidiert. Da für dieses Verfahren derzeit noch kein Standard besteht und damit die Ergebnisse der benötigen Analysen vergleichbar sind, wird empfohlen einen Standard für das TOP-Assay zu etablieren.

Für einen Abfallgrenzwert könnten einfach detektierbare Substanzen als Grundlage verwendet werden. Beispielsweise werden in der DIN 38407-42 und der DIN 38414 10 bestimmte PFAS analysiert. Es wird vorgeschlagen, die Auswahl an Perfluorsulfonsäuren auszuweiten und den Carbonsäuren anzupassen, damit alle C4-C10 Säuren beider Substanzgruppen betrachtet werden. Dadurch ergibt sich eine Anzahl an 14 zu messenden PFAS (C4-C10 Perfluorcarbonsäuren und C4-C10 Perfluorsulfonsäuren). Diese 14 Substanzen sollten vor und nach einem TOP-Assay gemessen werden, um die abfallgrenzwertrelevanten Konzentrationen der betrachteten Abfälle sowie die gesamte PFAS-Fracht der Abfälle zu bestimmen.

Herleitung von Abfallgrenzwerten für PFAS

Eine weitere Herausforderung ist die Herleitung des Grenzwertes. Da vorgeschlagen wird 14 individuelle PFAS zu messen (siehe Kapitel 3.6), müssten auch 14 individuelle Grenzwerte entwickelt werden, welche für einen Summengrenzwert addiert werden. Es konnten in diesem Vorhaben jedoch nicht ausreichend Daten für jede Substanz identifiziert werden. Insbesondere bezüglich der Umweltrisiken fehlt es an aussagekräftigen Daten, um ein oberes Begrenzungskriterium für mögliche Abfallgrenzwerte ermitteln zu können.

In diesem Vorhaben wurden Betrachtungen zu einem möglichen Abfallrenzwert deshalb für die Einzelsubstanzen PFOS und PFOA angestellt. Aufgrund der Komplexität der Herleitung der Abfallgrenzwerte und der teilweise mangelhaften Datenlage wurden die Betrachtungen für den beispielhaft anhand der Bodendaten angestellt, da für diesen Abfallstrom die meisten Messdaten vorliegen. Es werden jedoch auch Daten für Textilien, Klärschlamm und Papier dargestellt, soweit identifiziert.

Für den Abfallstrom der Abfallböden wurden jeweils für PFOS und PFOA die unteren und oberen Begrenzungskriterien ermittelt. Des Weiteren wurden die wirtschaftlichen Folgen und Hintergrundkontaminationen berücksichtigt. Es zeigte sich, unter den Rahmenbedingungen der zugrunde gelegten Methode zur Abschätzung des Abfallgrenzwertes, dass die oberen Konzentrationsgrenzwerte für PFOS- und PFOA die oberen Begrenzungskriterien für Umweltrisiken überschreiten würden. Würden solche Böden somit wiederverwendet werden, bestünde ein Risiko für negative Umweltauswirkungen. Es ist daher nicht gelungen, einen sinnvollen Abfallrenzwert für diese Stoffe unmittelbar auf der Grundlage der angewendeten Methode herzuleiten.

Dies geht darauf zurück, dass die recherchierten PNEC-Werte für PFOS und PFOA in einem sehr niedrigen Konzentrationsbereich liegen. Diese Werte werden bei der zugrunde gelegten Methode mit einem Sicherheitsfaktor (10.000) multipliziert (Potrykus et al., 2015), da angenommen werden kann, dass bei geeigneter Abfallbehandlung nur ein Bruchteil der gefährlichen Stoffe in die Umwelt gelangen. Dies ist bei den Böden jedoch nicht der Fall, da diese direkt wieder in die Umwelt gelangen, wodurch der Faktor hier keine Anwendung findet.

Eine ähnliche Situation herrscht bei den Klärschlämmen, da diese in der Landwirtschaft verwendet werden können, wodurch die PFAS direkt in die Umwelt gelangen können. Hier findet der Sicherheitsfaktor ebenfalls keine Anwendung. Eine Herleitung für einen Abfallrenzwert wurde im Rahmen dieses Vorhabens jedoch nicht vorgenommen und sollte in Zukunft noch durchgeführt werden.

Die Papiere und Textilien sind von dieser Einschränkung jedoch nicht betroffen, da bei sachgerechter Behandlung dieser Abfälle, das Risiko eines Eintrages von PFAS in die Umwelt, als gering angesehen wird. Generell könnte die in diesem Vorhaben beschriebene Methode auf diese beiden Abfallströme angewendet werden und es wird davon ausgegangen, dass ein sinniger Grenzwert abgeleitet werden kann. Dies wurde in diesem Vorhaben jedoch nicht durchgeführt und sollte daher in Zukunft erfolgen. Weiterhin sollten weitere analytische Messdaten für das Vorkommen von PFAS in diesen und weiteren Abfallströmen erhoben werden, um eine solide Datengrundlage für die Herleitung von Abfallgrenzwerten bereitzustellen.

Darstellung des Umweltkontextes für die ausgewählten Abfallströme und Empfehlungen für Entsorgungswege

Um Empfehlungen für Entsorgungswege abzuleiten, werden die errechneten PFAS-Massenströme der vier ausgewählten Abfallströme in einen Umweltkontext gebracht. Hierfür wird eine Risikobewertung durchgeführt, bei welcher die folgenden Fragen zugrunde liegen:

- Sind die angewendeten Beseitigungs- und Verwertungsverfahren geeignet, um die enthaltenen PFAS zu zerstören oder unumkehrbar umzuwandeln?
- Können bei diesen Prozessen neue PFAS entstehen bzw. werden die bestehenden PFAS umgewandelt?
- Verursachen Beseitigung- oder Verwertungsverfahren möglicherweise eine Umwelt- oder Gesundheitsgefährdung (d.h. eine relevante Exposition von Mensch oder Umwelt)? Hierbei werden auch andere relevante Grenzwerte betrachtet. Eine Emission von signifikanten PFAS Mengen in die Umwelt oder deren Verschleppung im Wertstoffkreislauf wird generell als Risiko angesehen.

Bei der Betrachtung werden mögliche Expositionspfade berücksichtigt und es wird diskutiert, welche Verfahren besonders zu einer Gefährdung von Mensch und Umwelt führen können beziehungsweise, welche Maßnahmen getroffen werden sollten, um dies zu vermeiden. Aus den Betrachtungen werden schlussendlich Empfehlungen für geeignete Entsorgungswege abgeleitet.

In Table 14 sind die Risiken und die Entsorgungswege der Abfallströme sowie damit verbundene Risiken und Empfehlungen dargestellt.

Tabelle 7: Ausgewählte Abfallströme, Entsorgungswege, Risiken und Empfehlungen

Abfallstrom bzw. Entsorgungsweg	Risiken und Empfehlungen
TEXTILIEN **Insgesamt fielen 2018 ~2 Millionen Tonnen an Textilabfällen in Deutschland an, was einer gesamten PFAS-Fracht von ~760 kg entspricht (durchschnittliche PFAS-Belastung: ~389 µg/kg). Davon wurden ca. 70% dem Recycling zugeführt, ca. 28% thermisch behandelt und ca. 2% sonstig entsorgt.**	
Stoffliche Verwertung ~70%	PFAS werden während der stofflichen Verwertung von Textilien nicht zerstört. Es kann zu einer Verschleppung der PFAS kommen z.B. durch das Recycling und die Wiederverwendung der Textilien. Werden die Textilfasern recycelt, so können die PFAS ausgewaschen werden und über den Abwasserpfad in die Kläranlagen gelangen. Insbesondere flüchtige PFAS können in die Luft gelangen und möglicherweise zu einer Belastung von exponiertem Personal führen. Da in einer Recyclinganlage für entsprechende Textilabfälle erhöhte Konzentrationen von PFAS in der Luft auftreten könnten, sollte in Erwägung gezogen werden, persönliche Schutzmaßnahmen zu ergreifen, um das Risiko einer PFAS-Exposition durch Einatmen und Hautkontakt zu vermeiden. Dies könnte z.B. das Tragen von Atemschutzmasken und Handschuhen umfassen. Um die Notwendigkeit für solche Schutzmaßnahmen festzustellen, können geeignete Luftmessungen durchgeführt werden. Es wird empfohlen Produkte, die mit hoher Wahrscheinlichkeit erheblich mit PFAS belastet sind, wie z.B. Outdoorjacken, Arbeitskleidung sowie Outdoortextilien wie Markisen frühzeitig zu identifizierten, auszusortieren und der thermischen Verwertung zuzuführen, um eine Verschleppung von PFAS zu vermeiden.
Thermische Behandlung ~28%	In Europa müssen Müllverbrennungsanlagen mit einer Mindesttemperatur von 850°C und eine Verweilzeit von mindestens 2 Sekunden betrieben werden. Da es sich hierbei um ein Minimumkriterium handelt, liegen die tatsächlichen Temperaturen in der Regel etwas höher. Diverse wissenschaftliche Studien konnten zeigen, dass diese Bedingungen ausreichen, um PFAS weitgehend zu zerstören. Insignifikante Mengen an PFAS konnten in der Asche nachgewiesen werden. Relevante Konzentrationen von kurzkettigen Fluorgasen in der Abluft konnten bisher nicht identifiziert werden. Es wird daher empfohlen solche Gase im Abgas von Müllverbrennungsanlagen in Europa zu analysieren, um die entsprechende Zerstörungseffizienz der Anlagen bestimmen zu können. Derzeit ist die thermische Behandlung die empfohlene Entsorgungsmethode für PFAS-belastete Abfälle, da davon ausgegangen wird, dass PFAS bei dieser Behandlungsmethode weitgehend zerstört werden. Deshalb wird derzeit empfohlen, PFAS belastete Fraktionen von Textilabfällen möglichst einer thermischen Abfallbehandlung zuzuführen. Die Ergebnisse weiterer Untersuchungen hinsichtlich möglicher luftgetragener PFAS-Emissionen sind dabei zu berücksichtigen.
Ablagerung auf Deponien/sonstige Behandlung ~2%	In Deutschland dürfen Textilien nicht deponiert werden. Empfehlungen zur Deponierung sind daher nicht relevant.
KLÄRSCHLAMM	

Abfallstrom bzw. Entsorgungsweg	Risiken und Empfehlungen
Insgesamt fielen 2020 ~1,74 Millionen Tonnen an Klärschlamm in Deutschland an, was einer gesamten errechneten PFAS-Fracht von ~117 kg entspricht (durchschnittliche PFAS-Belastung: ~67 µg/kg). Davon wurden ca. 22% der stofflichen Verwertung zugeführt, ca. 77% thermisch entsorgt und ca. 1% auf andere Weise entsorgt.	
Stoffliche Verwertung ~22%	Bei der stofflichen Verwertung der Klärschlämme werden diese bodenbezogen als Düngemittel in der Landwirtschaft, oder bei landschaftsbaulichen Maßnahmen verwendet. Zerstört werden PFAS während der stofflichen Verwertung nicht, sie gelangen daher i.d.R. direkt in die Umwelt. In den Kläranlagen konnten außerdem flüchtige PFAS in der Luft nachgewiesen werden, wodurch ein mögliches Risiko für Personal besteht. Gemessene PFAS-Konzentrationen in der Luft von Kläranlagen liegen jedoch im ng/m³ Bereich und liegen somit weit unterhalb typischer PFAS-Arbeitsplatzgrenzwerte im mg/m³ Bereich. Grenzwerte für PFAS im Klärschlamm existieren bereits in der Düngemittelverordnung (100 µg/kg für die Summe aus PFOS und PFOA), wodurch der Eintrag dieser Substanzen in die Umwelt verringert wird. Die Anzahl der für einen geeigneten Grenzwert zu messenden Substanzen und die Höhe des Grenzwertes sollten diskutiert werden. Generell wird empfohlen die PFAS-Belastung von Klärschlämmen zu bestimmen, um dann die belasteten Schlämme einer thermischen Behandlung zuzuführen, um die PFAS möglichst weitgehend zu zerstören.
Thermische Behandlung ~77%	Derzeit ist die thermische Behandlung die empfohlene Entsorgungsmethode für PFAS-belastete Abfälle, da die PFAS bei dieser Behandlungsmethode weitgehend zerstört werden (siehe auch thermische Behandlung zu Textilien). Deshalb wird empfohlen, PFAS belastete Klärschlämme einer thermischen Abfallbehandlung zuzuführen.
Ablagerung auf Deponien/sonstige Behandlung ~1%	Unter die sonstige direkte Entsorgung der Klärschlamme fällen z.B. die Abgabe an Trocknungsanlagen und andere nicht spezifizierte Entsorgungswege. Da die genauen Behandlungswege unbekannt sind, können keine konkreten Risiken und Empfehlungen abgeleitet werden. Es wird davon ausgegangen, dass PFAS in Trocknungsanlagen nicht zerstört werden, wobei die Risiken und Empfehlungen der stofflichen Verwertung gelten.
PAPIER **Insgesamt fielen 2020 ~14,5 Millionen Tonnen an Altpapieren in Deutschland an, was einer gesamten PFAS-Fracht von ~4.000 kg entspricht (durchschnittliche PFAS-Belastung: ~274 µg/kg). Davon wurden ca. 80% der stofflichen Verwertung zugeführt, ca. 19,7% thermisch entsorgt und ca. 0,3% deponiert.**	
Stoffliche Verwertung ~80%	Bei der stofflichen Verwertung der Altpapiere werden die vorhandenen PFAS nicht zerstört. Die PFAS werden entweder in neue Produkte verschleppt oder sie gelangen über die Wasserphase in lokale Kläranlagen und schließlich teilweise in die Umwelt. Grenzwerte für Abwasser aus der Herstellung von Papier, Karton oder Pappe existieren bereits für diverse Schadstoffe, jedoch sind in Anhang 28 der Abwasserverordnung keine Grenzwerte für PFAS gelistet. Diese könnten in Zukunft etabliert werden. Während des Recyclings können flüchtige PFAS in die Luft übertreten, wodurch ein mögliches Risiko für das Personal besteht. Es ist möglich, dass PFAS-Konzentrationen in Papierrecyclinganlagen und Anlagen, welche Recyclingpapier verwenden erhöht sein könnten, jedoch sollte dies durch Messungen geprüft werden.

Abfallstrom bzw. Entsorgungsweg	Risiken und Empfehlungen
	Die Kompostierung von Rückständen aus der Papierindustrie und deren bodenbezogene Nutzung sollte generell nicht durchgeführt werden. Weiterhin wird empfohlen, stark kontaminierte Papierabfälle bzw. Papierabfälle mit hohem Risiko PFAS zu enthalten von den übrigen Papierabfällen zu trennen und der thermischen Behandlung zuzuführen. Dabei werden die PFAS weitgehend zerstört. Analog zu den Textilien könnten auch hier Maßnahmen zur Bewusstseinsbildung in der Öffentlichkeit hilfreich sein.. PFAS haltige Verpackungen für Lebensmittel dürfen laut Verpackungsgesetz nicht im Restmüll entsorgt werden, sondern müssen zusammen mit den Leichtverpackungen in der gelben Tonne recycelt werden.
Thermische Behandlung ~19,7%	Derzeit ist die thermische Behandlung die empfohlene Entsorgungsmethode für PFAS-belastete Abfälle, da die PFAS bei dieser Behandlungsmethode weitgehend zerstört werden (siehe auch thermische Behandlung zu Textilien). Deshalb wird empfohlen, auch PFAS belastete Fraktionen von Papierabfällen möglichst einer thermischen Abfallbehandlung zuzuführen.
Ablagerung auf Deponien/sonstige Behandlung ~0,3%	Ein geringer Anteil der Papierrückstände wird systembedingt noch deponiert und dadurch kann PFAS ins Sickerwasser gelangen (Die Papierindustrie, 2020). Das Sickerwasser kann in den Boden der Deponie eintreten, wird jedoch meistens in die lokale Kläranlage geleitet, wo es behandelt wird. PFAS werden bei der Deponierung nicht zerstört, sondern können ins Sickerwasser der Deponie gelangen. Durch eine fachgerechte Sickerwasserbehandlung (z.B. durch Aktivkohlefilteranlagen) kann die weitere Verbreitung der PFAS vermindert werden.
BÖDEN **Insgesamt fielen 2018 ~129 Millionen Tonnen an Abfallböden in Deutschland an, was einer gesamten PFAS-Fracht von ~2.300 kg entspricht (durchschnittliche PFAS-Belastung: ~18 µg/kg). Davon wurden ca. 85% der stofflichen Verwertung zugeführt, ca. 0,2% thermisch behandelt und ca. 15% deponiert.**	
Stoffliche Verwertung ~85%	Bei der stofflichen Verwertung der Abfallböden werden die vorhandenen PFAS nicht zerstört. Abfallböden werden meistens verfüllt. PFAS-Grenzwerte für die Wiederverwertung von Böden existieren bereits. Generell können die PFAS aus den kontaminierten Böden ausgewaschen werden und ins Grundwasser gelangen. Es gibt jedoch Unterschiede in der Bindungskraft der Böden, da manche PFAS stärker binden, jedoch fehlt es hierzu noch an konkreten Studien. Es wird daher empfohlen hierzu Daten zu erheben. Während des Aushebens können insbesondere flüchtige PFAS in die Luft übergehen und eingeatmet werden. Es könnte deshalb in Erwägung gezogen werden geeignete Schutzausrüstung (z.B. Schutzmaske und Handschuhe) bei solchen Maßnahmen zu tragen. Um die Notwendigkeit für solche Schutzmaßnahmen festzustellen könnten geeignete Luftmessungen durchgeführt werden. Es wird generell empfohlen PFAS-belastete Böden vor der Wiederverwendung zu sanieren, um die enthaltenen PFAS zu entfernen bzw. zu zerstören. Diverse chemische und thermische Verfahren sind in der Lage PFAS effektiv zu zerstören, jedoch sind diese oftmals noch nicht großtechnisch etabliert, weshalb hier noch Forschungsbedarf besteht. Alternativ können PFAS auch immobilisiert, ausgewaschen bzw. mittels Pump-and-Treat-Verfahren aus dem Boden entfernt werden.

Abfallstrom bzw. Entsorgungsweg	Risiken und Empfehlungen
Thermische Behandlung ~0,2%	Derzeit ist die thermische Behandlung die empfohlene Entsorgungsmethode für PFAS-belastete Abfälle, da die PFAS bei dieser Behandlungsmethode weitgehend zerstört werden (siehe auch thermische Behandlung zu Textilien). Deshalb wird empfohlen, auch stark PFAS belastete Fraktionen von Bodenabfällen möglichst einer thermischen Abfallbehandlung zuzuführen.
Ablagerung auf Deponien/sonstige Behandlung ~15%	Ca. 15% des als Abfall anfallenden Bodenmaterials wird auf Deponien abgelagert. Es gelten dieselben Risiken und Empfehlungen wie bei der Deponierung von anderen PFAS haltigen Abfällen (siehe Deponierung von Papier). PFAS werden bei der Deponierung nicht zerstört, sondern können ins Sickerwasser der Deponie gelangen. Durch eine fachgerechte Sickerwasserbehandlung (z.B. durch Aktivkohlefilteranlagen) kann die weitere Verbreitung der PFAS vermindert werden. Die thermische Behandlung oder die Ablagerung auf geeignete Deponien mit entsprechenden Sicherungsmaßnahmen (Basisabdichtung, Oberflächenabdichtung) ist die derzeit empfohlene Form der Entsorgung von kontaminierten Böden. Ebenfalls kann die Untertageverbringung z.B. in ehemalige Bergwerke in Betracht gezogen werden.

Summary

Background and goals

In recent years, the importance of perfluorinated and polyfluorinated alkyl compounds (PFAS) has steadily increased. The production volume of PFAS has followed a rapidly increasing trend over the past decades. PFAS are water, dirt and grease repellent. They are used worldwide and can be found in countless products, from outdoor jackets to Teflon pans to firefighting foams (UBA, 2020a). Because PFAS are used in so many ways, there are also many ways they can enter the environment: During the manufacture of the chemicals themselves, their further processing into products, during the use of the products, and finally during and after disposal.

With the increasing importance of this group of substances, based mainly on their functional properties for many technical processes, the release into the environment is also increasing. PFAS can spread through the air, rivers and oceans to remote areas such as the Arctic. They are hardly degradable and therefore remain in the environment for a very long period of time. Some PFAS accumulate in animals, plants and humans and also have adverse health effects. Elevated concentrations of PFOA and PFOS in human blood can reduce effects of vaccinations, increase the tendency to infections, lead to elevated cholesterol levels, and result in reduced birth weight in offspring (UBA, 2020a).

Due to their properties, it is necessary to assess the risk of PFAS for humans and the environment and there is a need for research on their fate in the environment, in particular with regard to their persistence and input quantities from waste streams. To address this issue, the Federal Environment Agency has initiated the research project "Investigation of the occurrence of PFAS (per- and polyfluorinated alkyl compounds) in waste streams".

This project aims at a first identification, quantification and evaluation of the occurrence of PFAS in waste streams. Furthermore, a basis for further research priorities shall be created and the need for action shall be derived. The need to record PFAS in waste arises directly from the Stockholm Convention on Persistent Organic Pollutants (POPs), in whose Annex A (elimination) representatives from the group of PFAS (PFOS, their salts and PFOSF, PFOA their salts and PFOA-

related compounds as well as PFHxS, their salts and PFHxS-related compounds) have already been included.

Approach/methodology

In order to achieve the project goals, first of all, based on a background research (see chapter 2.1and 3.1), relevant waste streams were identified, a sampling plan was developed, and targeted sampling and subsequent physico-chemical analysis were performed, in which the organic fluorine is recorded in the form of the sum parameter (EOF) (see chapter 2.2, 2.3, 2.4 and 3.2).

Based on the measurement results and the PFAS concentrations to be derived from them (see chapter 3.2), PFAS mass flows in the respective waste streams were to be calculated and, based on this, considerations of possible disposal routes and possible PFAS limit values in the area of waste legislation were to be made. However, no concentrations for individual PFAS substances could be obtained from the PFAS measurements of the samples, but only sum parameters (see chapter 3.2). Thus, the basis for calculating PFAS mass flows and for questions based on this was missing.

In order to be able to calculate the mass flows and derive the waste limit values, it was therefore decided in consultation with the Federal Environment Agency to research and evaluate suitable literature values. For this purpose, an additional literature search was carried out for the selected waste streams (paper, sewage sludge, soils and textiles) (see chapter 2.5 3.3). The values found were extracted from the literature, averaged and used to calculate the mass flows (see chapter 2.7 and 3.4) were determined.

To estimate the environmental relevance, the selected waste streams were placed in an environmental context (see chapter 2.8 and 3.5) and considerations of possible limit values (see chapter 2.9 and 3.6) have been made.

Based on the results, recommendations for the establishment of possible waste management limits were derived (see chapter 3.6) and instructions for the public and for waste management enforcement were formulated (see chapter 3.7 and 3.8).

Analyses and analysis results

From the literature review conducted, the following waste streams were identified as relevant in consultation with UBA:

- Textiles
- Sewage sludge
- Paper
- Soils
- Hardware store products intended for outdoor use (*e.g.* paints).

In accordance with this assessment, samples from these areas were collected by BAM for the EOF examinations. In the case of textiles, samples were examined that were sent to the used clothing collection. Sewage sludge was requested from wastewater treatment plant operators with a special focus on industrial wastewater treatment plants from the paper sector, as well as municipal wastewater treatment plants with a known discharger from the PFAS-using sector (*e.g.*, electroplating plants). The paper studied was from a wastepaper sorting facility. Soils were taken from various operators of landfills, state investigation offices, or on site itself. The examined paints and other products for outdoor use were the only ones examined in a non-aged condition, but new condition. The examination of all samples was carried out by means of HR-CS-MAS with regard to the sum parameter EOF after suitable sample preparation, extraction and purification.

Textiles

The textiles were examined after the samples had been milled and homogenized. The samples include ten different jackets, two bags and one carpet, which were taken from the used clothing collection on different days. The total sample size thus corresponds to 13 samples. The examination of the jackets showed an EOF content in the range of 37 µg/kg up to an EOF content of 1163 µg/kg. In one bag no EOF could be detected and for the second bag a content of 195 µg/kg was determined. An EOF content of 83 µg/kg was also detected in the carpet. This range, which covers two orders of magnitude, shows that used textiles are a highly heterogeneous waste stream.

Sewage sludge

The sewage sludges examined in this study were all taken by the operator of the respective wastewater treatment plants and freeze-dried on site. A total of 17 sewage sludge samples were considered in this study. Samples were taken from industrial wastewater treatment plants associated with PFAS-containing products, on the one hand, and municipal wastewater treatment plants whose sewage sludges had already exhibited elevated PFAS levels in the past, on the other.

In the case of industrial sewage treatment plants, a maximum EOF content of approx. 5000 µg/kg could be determined in the sewage sludge. In the sewage sludge from municipal wastewater treatment plants, an EOF content of up to approx. 450 µg/kg could be determined. This value is above that of typical target PFAS investigations and shows that underestimation can occur using target analysis methods. It also shows that there is an input of organic fluorine into the wastewater treatment plants as well as an accumulation in the pollutant sink sewage sludge.

Paper

The samples were taken from a wastepaper sorting plant and analyzed for their EOF content. Two fractions were taken. One was graphic paper and the other was packaging board. The samples were milled and homogenized. The subsequent analysis showed an EOF value of approx. 120 µg/kg for the paper sample and a value of approx. 240 µg/kg for the packaging board. This clearly shows that the recycled raw material paper and cardboard has a basic content of PFAS, which can be carried over into new products during reuse and can be released into the environment from these or during processing.

Soils

In this study, nine soils of different origins were examined after freeze-drying. Soils without known contamination as well as soils from known contaminated regions were investigated. The pre-contaminated soils show an EOF load of up to 3 mg/kg. The remaining soils all show EOF contamination (< 0.25 mg/kg), but they are very similar regardless of their sampling location. This indicates that there is a measurable background level of fluorinated organic compound contamination.

Hardware store products

Products used outdoors often contain PFAS to give them water- and dirt-repellent properties. For this reason, this study examined various DIY products intended for outdoor use. Five paints and three sealants were examined. The sealing materials showed no detectable EOF content. In contrast, PFAS was detected in all samples of the paints examined. The range of EOF content was between 40 µg/kg and 180 µg/kg. The data thus obtained suggest that outdoor paints contain PFAS, which may be washed out after their application and released into the environment. Furthermore, the disposal of residues as well as of the correspondingly treated surfaces may lead to a contamination of the corresponding waste stream.

Despite the identification of numerous "hotspot" samples, no reliable value for a single substance could be measured in any of the samples by means of target analysis. This clearly demonstrates the importance of screening samples in order to provide a possible assessment of contamination. It also suggests that further methods need to be developed to meet the need for measurement of target analytes to provide an estimate of the potential hazard of wastes and products.

PFAS concentrations in the selected waste streams.

Since no measured values for individual PFAS could be obtained from the samples measured in this project, a further literature search was conducted for PFAS measurement data in the waste streams textiles, sewage sludge paper and soils. For these, PFAS literature data focusing on German samples were researched, extracted, and mean and median values were formed from the data obtained. These values are subject to some uncertainty due to several limitations. In particular, the limitations result from (1) the different number of PFAS analyzed in the identified studies, (2) a bias in the results because suspect cases are often analyzed specifically (except for soils, no or very few background measurements were made or identified), and (3) deviations resulting from the calculation of the sum concentrations. The data should be interpreted in light of the limitations.

Textiles

PFAS are used in textiles to give them water- and grease-repellent properties. Particularly high PFAS concentrations were found in awnings and in outdoor jackets, which originate from the use of fluorotelomer alcohols. Table 8 summarizes the calculated mean and median values.

Table 8: PFAS mean and median values in the textile samples found. All data in µg/kg.

Substance	Mean value	Median
∑PFCA	87.56	8.36
∑PFSA	20.65	0.00
∑PFPA	0.00	0.00
∑Precursors	280.70	3.46
∑PFAS	**388.91**	**74.05**

The averaged PFAS values for the textiles agree well with the results sampled for textiles in this project.

In a recent report by Wood (2020), concentrations of 0.025-0.05% are reported for leather (250-500 mg/kg), 0.03% for synthetic carpets (300 mg/kg), and 2-3% for textiles and furniture (20,000-30,000 mg/kg). These concentrations are above the concentrations calculated here in Table 8 which may be due to the fact that the concentrations of Wood (2020) are based on the average PFOS concentrations in textiles from UNEP (2017).

Similar concentrations are also given in the total PFAS restriction procedure. These vary between <0.1% and 7% depending on the application. Some textiles consist of 100% PFAS such as PTFE membranes. (Annex XV, 2023a). In general, the concentrations here are also higher than the calculated values in Table 8 which may be due to the fact that the restriction procedure covers all PFAS (including polymers) and not only the short-chain acids and some precursors as in this project.

In order to be able to represent the actual contamination of textiles and their waste with PFAS as representatively as possible, targeted measurements of different textiles should be carried out, which can then be extrapolated.

Sewage sludge

Sewage sludge is generated in wastewater treatment plants treating municipal and/or industrial wastewater. Major PFAS point sources include wastewater from industries that manufacture or use PFAS in their processes, such as the paper, textile, metal coating, and semiconductor industries. However, since PFAS are also used in a variety of consumer products and household applications, they can also be found in purely municipal wastewater, *e.g.*, by washing them out of textiles.

The PFAS accumulate in the sewage sludge in the wastewater treatment plants or are discharged into surface waters via the effluent.

To obtain an overview of PFAS concentrations in German sewage sludge, a large number of sewage sludge samples from different sources were analyzed. Only a few data on industrial wastewater treatment plants were identified. Such plants have higher PFAS concentrations than municipal plants. The few data on industrial WWTPs were not considered in the calculation of PFAS concentration values. Based on the identified data for municipal plants, the PFAS concentrations shown in Table 9 were calculated on the basis of the data obtained for municipal plants.

Table 9: PFAS mean and median values in the sewage sludge samples found. All data in µg/kg.

Substance	Mean value	Median
∑PFCA	18.03	16.60
∑PFSA	48.80	31.30
∑PFPA	0.00	0.00
∑Precursors	0.36	0.00
∑PFAS	**67.18**	**47.40**

A long-term study (from 2008-2013) showed that PFAS concentrations in sewage sludge decreased overall for both regulated and non-regulated substances. Current trends could be determined by new measurements.

The Table 9 calculated PFAS mean and median values can be compared with the EOF values measured in this project. While EOF values up to 5,000 µg/kg could be detected in industrial wastewater treatment plants, the PFAS concentrations from the municipal wastewater treatment plants are mostly in a range below 500 µg/kg (mean value 338 µg/kg). The calculated values from Table 9 are slightly below these values.

The calculated values are in a similar range as data published in the overall PFAS restriction dossier in February 2023 (Annex XV, 2023b).

Paper

PFAS are used in particular in food contact papers to give the paper water- and grease-repellent properties. However, since papers are largely recycled in Germany, PFAS may also be present in other papers. This could also be confirmed by the wastepaper and waste cardboard analyses (graphic papers and cardboard packaging) in this project (see chapter 2.2.4 and 3.2.2).

In general, the term food contact paper is very broad. This includes, for example, burger paper packaging, paper bags at the bakery, baking paper, paper plates, paper straws and pizza boxes, but also paper and cardboard packaging of, for example, frozen goods. Depending on the application, more or less PFAS are used.

Based on the data obtained for predominantly food contact papers, the data presented in Table 10 were calculated on the basis of the data obtained for predominantly food contact papers.

Table 10: PFAS mean and median values in the paper samples found. All data in µg/kg.

Substance	Mean value	Median
∑PFCA	155.87	0.60
∑PFSA	1.48	0.70
∑PFPA	0.00	0.00
∑Precursors	116.47	0.00
∑PFAS	**273.81**	**10.40**

Under the total PFAS restriction procedure, fluorine concentrations in food contact papers of 537 mg/kg (mean) and 1,200 mg/kg (maximum) are reported. These values refer only to fluorine, but PFAS molecules also consist of other atoms such as carbon and oxygen. The preparers of the dossier have therefore assumed that fluorine accounts for about 50% of the total weight of PFAS molecules (compered to: PFHxA ~66% fluorine content and side-chain fluorinated PFAS ~12.8% fluorine content) (Annex XV, 2023a). This results in PFAS concentrations of 1,074 mg/kg (mean) and 2,400 mg/kg (maximum) in food contact papers. These values are significantly higher than those given in Table 10 which may be due to the fact that all PFAS are considered in the restriction dossier (acids, polymers, side-chain fluorinated polymers, etc.), whereas only the short-chain acids and some precursors are considered here.

In order to be able to represent the actual contamination of textiles and their waste with PFAS as representatively as possible, targeted measurements of different papers should be carried out.

Soils

PFAS are not intentionally applied to soils, but they are also used in products containing PFAS, such as pesticides. In most cases, PFAS occur as unintentional contaminants in soils, for example, from the past application of PFAS-contaminated sewage sludge or paper waste added to composts. Another source of contamination in soils is the use of firefighting foams containing PFAS, as well as the industrial production of PFAS and the associated emissions to air and subsequent deposition to soils, among others.

The data set on PFAS in soils includes by far the most literature-based samples in this project, with about 8,000 individual measurements. The majority of the samples were taken in suspected cases, but some background measurements were also taken. In many of the suspected cases, however, PFAS could not be detected because the measured values were below the detection limit (detection limit: 1µg/kg).

Based on all identified values for soils, the values shown in Table 11 were calculated on the basis of all identified values for soils.

Table 11: PFAS mean and median values in the identified soil samples. All samples from Rastatt were included. All data in µg/kg.

Substance	Mean value	Median
∑PFCA	11.90	0.14
∑PFSA	3.50	0.00
∑PFPA	0.00[1]	0.00
∑Precursors	2.14	0.00
∑PFAS	**17.53**	**0.21**

1 The concentration for PFPA is not zero here, but statistically irrelevant.

Since most of the data come from the district of Rastatt, this data is not necessarily representative for all of Germany.

In addition, comparative values could be obtained from North Rhine-Westphalia. In uncontaminated arable soils, ∑ PFAS values of up to ~7.6 µg/kg could be detected. However, most values were below 1 µg/kg. These concentrations are largely composed of PFOS, PFOA, and PFBA. The calculated mean value in the range of about 18 µg/kg in Table 11 agrees well with these values when it is considered that this value includes both contaminated and uncontaminated soil.

Last, the calculated PFAS concentrations obtained from Table 11 can be compared with the EOF values measured in this project. In the measured contaminated soils, EOF values of up to 3,000 µg/kg were detected (mean value 1,249 µg/kg). In the uncontaminated soils, the EOF values vary between 73 and 209 µg/kg and are thus higher than the calculated values.

Waste data and PFAS fluxes in the selected waste streams.

Based on the researched data for the selected waste streams and the determined PFAS concentrations as mean and median values, the PFAS loads in the waste streams are estimated. The loads for the perfluorocarboxylic, -sulfone and -phosphoric acids, the precursors as well as a sum value for all PFAS are shown.

The majority of textile waste in Germany is recycled (69%) and thermally treated (approx. 28%). Less than 3% are disposed of in other ways.

Most of the sewage sludge from municipal wastewater treatment plants in Germany is recovered by thermal treatment (77%) and soil-related recycling (22%). About 1% is disposed of in other ways (*e.g.*, composting/fermentation).

No conclusive data could be found regarding the wastepaper stream. For this project, a paper industry source for the year 2020 is used and a material recovery rate of 80% is assumed. For the remaining 20%, mainly energy recovery (approx. 19.7%) and a small share of landfilling (approx. 0.3%) are assumed.

Of 128.9 million tons of waste soil generated in Germany in 2018, 14.95% were disposed of, while 85.05% were recycled. Most of the disposed waste was landfilled (14.89%), i.e. it was deposited in/on a landfill/special landfill or permanently stored in a container, e.g. in a mine. Only a small part was incinerated (0.02%) or, if the contamination is in the form of liquid or sludge waste, biodegraded (0.04%). In terms of recovery, most of the soil (68.87%) was recovered by backfilling. 16.17% were recovered by recycling, that is, by reprocessing organic material with the aim of using it as originally intended. Only a small part of the contaminated soil (0.01%) was recovered by energy recovery.

Table 12 and Table 13 summarize the annual PFAS mass flows in the waste streams considered. A distinction is made between the type of treatment, so that the annual quantities of PFAS recycled, landfilled and otherwise recovered (*e.g.*, backfilled) are shown.

Table 12: Summary of calculated PFAS mass flows from the average of the selected waste streams. All figures in kg.

Waste stream	Total PFAS quantity	Amount of PFAS recycled/materially recovered	Amount of PFAS energetically recovered	Quantity of PFAS deposited/other
Textiles	758.71	523.74	215.20	19.78
Sewage sludge	116.94	26.13	89.69	1.12
Paper	3.961.46	3.169.17	780.13	12.16
Soils	2260.37	1922.22	0.55	337.60
Total	**7,097.48**	**5.641.26**	**1,085.57**	**370.66**

If the mean value in Table 12 is considered, the waste streams analyzed contain a total of about 7,100 kg of PFAS. More than half of this (~56%) comes from paper waste, while only ~1.6% comes from sewage sludge. However, the high PFAS load in the paper samples is not considered realistic, as only PFAS readings from food contact paper could be found. However, many papers in the paper waste stream are not or only partially contaminated with PFAS (*e.g.*, newspapers, books, *etc.*), which probably overestimates the PFAS load. The high PFAS amounts in the soil samples can be attributed to the comparatively very large amounts of waste soil collected (~130,000,000 t compared to ~17,000,000 t of wastepaper).

Table 13: Summary of calculated PFAS mass flows from median of selected waste streams. All figures in kg.

Waste stream	Total PFAS quantity	Amount of PFAS recycled/materially recovered	Amount of PFASenergetically recovered	Quantity PFAS deposited/other
Textiles	144.47	99.73	40.98	3.77
Sewage sludge	82.50	18.43	63.28	0.79
Wastepaper	150.47	120.37	29.63	0.46
Soils	26.54	22.57	0.01	3.96
Total	**403.98**	**261.1**	**133.9**	**8.98**

If the median values in Table 13 are considered, the waste streams investigated contain a total of ~400 kg PFAS.

For the further questions of the project, the data are considered with reference to the mean values. In this respect, they are to be interpreted as a worst-case scenario.

Considerations and recommendations on waste limits for PFAS

An existing methodology for deriving POP waste limit values was used to derive waste management limit values. The methodology thereby limits the concentration range of a possible limit value of a substance downwards and upwards on the basis of various criteria. However, some difficulties and challenges arose when applying the method to the substance group of PFAS.

Selection of the PFAS to be investigated

A major challenge is the question of which PFAS should be considered in the limit derivation. A method is needed by which the precursor substances can be included in the limit value consideration without measuring them as individual substances.

Due to the great diversity of PFAS, not every substance can be detected analytically and individually, which means that the total PFAS load is often unknown. Especially the precursor compounds are very diverse. Therefore, it is suggested to use the TOP assay in order to include these compounds in the analysis. Here, the precursor compounds are largely oxidized to the analogous perfluorocarboxylic acids. Since there is currently no standard for this method and to achieve comparability of the results of the required analyses, it is recommended that a standard is established for the TOP assay.

For a waste limit value, easily detectable substances could be used as a basis. For example, DIN 38407-42 and DIN 38414 analyze 10 specific PFAS. It is proposed to extend the selection of perfluorosulfonic acids and to adapt it to the carboxylic acids so that all C4-C10 acids of both substance groups are considered. This results in a number of 14 PFAS to be measured (C4-C10 perfluorocarboxylic acids and C4-C10 perfluorosulfonic acids). These 14 substances should be measured before and after a TOP assay to determine the waste limit relevant concentrations of the considered wastes as well as the total PFAS load of the wastes.

Derivation of the waste limit values for PFAS

Another challenge is the derivation of the waste limit values. Since it is proposed to measure 14 individual PFAS (see chapter 3.6), 14 individual limit values would also need to be developed, which would be summed for a cumulative limit value. However, in this project the data identified for each substance is not sufficient. In particular, with regard to environmental risks, there is a lack of meaningful data to determine an upper limiting criterion for the waste limit values.

In this project, considerations of a possible limit waste value were therefore made for the individual substances PFOS and PFOA. Due to the complexity of the derivation of the waste limit values and the partially inadequate data situation, the considerations were made on the basis of soil data as an example, since most measurement data are available for this waste stream. However, data for textiles, sewage sludge and paper are also presented where identified.

For waste soils, the lower and upper limiting criteria were determined for PFOS and PFOA, respectively. Furthermore, the economic consequences and background contaminations were considered. It was shown, under the framework conditions of the underlying method for estimating the limit value, that the upper concentration limits for PFOS and PFOA would exceed the upper limiting criteria for environmental risks. Thus, if such soils were reused, there would be a risk of adverse environmental impact. It has therefore not been possible to derive a meaningful limit value for these substances directly on the basis of the method used.

This is due to the fact that the researched PNEC values for PFOS and PFOA are in a very low concentration range. These values are multiplied by a safety factor (10,000) in the underlying method (Potrykus et al., 2015), since it can be assumed that with appropriate waste treatment only a fraction of the hazardous substances will be released into the environment. However, this is not the case for soils, as they are directly reintroduced into the environment, which means that the factor does not apply here.

A similar situation prevails with sewage sludges, as these can be used in agriculture, which means that the PFAS can be released directly into the environment. Here, the safety factor is also not applied. However, a derivation for a limit value was not carried out within the scope of this project and should still be carried out in the future.

However, papers and textiles are not affected by this restriction, as the risk of PFAS entering the environment is considered low if these wastes are handled properly. In general, the method described in this project could be applied to these two waste streams and it is assumed that a reasonable limit can be derived. However, this was not done in this project and should be done in the future. Further analytical measurement data for the occurrence of PFAS in these and other waste streams should be collected to provide a solid data basis for limit considerations.

Environmental context for the selected waste streams and recommendations for disposal pathways

In order to derive recommendations for disposal routes, the calculated PFAS mass flows of the four selected waste streams are put into an environmental context. For this purpose, a risk assessment is carried out based on the following questions:

- Are the disposal and recovery processes applied suitable to destroy or irreversibly transform the PFAS contained?
- Can new PFAS be generated during these processes or are the existing PFAS transformed?
- Do disposal or recovery processes possibly cause an environmental or health hazard (*i.e.*, relevant exposure of humans or the environment)? Other relevant limit values are also considered here. An emission of significant amounts of PFAS into the environment or their carry-over in the recyclable material cycle is generally considered a risk.

Possible exposure pathways are taken into account, and it is discussed which processes in particular can lead to a risk to humans and the environment or which measures should be taken to avoid this. Finally, recommendations for suitable disposal routes are derived from the considerations.

Table 14 shows the risks and the disposal routes of the waste streams as well as associated risks and recommendations.

Table 14: Selected waste streams, disposal routes, risks and recommendations

Waste stream or disposal route	Risks and recommendations
TEXTILES **A total of ~2 million tons of textile waste was generated in Germany in 2018, corresponding to a total PFAS load of ~760 kg (average PFAS load: ~389 µg/kg). Of this, ~70% was recycled, ~28% thermally treated, and ~2% otherwise disposed.**	
Material recovery ~70%	PFAS are not destroyed during the recycling of textiles. A carry-over of PFAS can occur, *e.g.*, through the recycling and reuse of textiles. If the textile fibers are recycled, the PFAS can be washed out and enter the wastewater treatment plants via the wastewater pathway. In particular, volatile PFAS may become airborne and potentially result in exposure of exposed personnel. Since elevated concentrations of PFAS in air

Waste stream or disposal route	Risks and recommendations
	could occur in a recycling plant for appropriate textile wastes, consideration should be given to taking personal protective measures to avoid the risk of PFAS exposure through inhalation and skin contact. This could include, for example, wearing respirators and gloves. Appropriate air measurements may be taken to determine the need for such protective measures. It is recommended that products that are highly likely to be significantly contaminated with PFAS, such as outdoor jackets, work clothing and outdoor textiles such as awnings, be identified at an early stage, sorted out and sent for thermal recycling in order to prevent the carry-over of PFAS.
Thermal treatment ~28%	In Europe, waste incineration plants must operate at a minimum temperature of 850 °C and a residence time of at least 2 seconds. Since this is a minimum criterion, the actual temperatures are usually somewhat higher. Various scientific studies have shown that these conditions are sufficient to destroy PFAS to a large extent. Insignificant amounts of PFAS could be detected in the ash. Relevant concentrations of short-chain fluorine gases in the exhaust air could not be identified so far. It is therefore recommended to analyze such gases in the exhaust gas of waste incineration plants in Europe in order to be able to determine the corresponding destruction efficiency of the plants. Currently, thermal treatment is the recommended disposal method for PFAS-contaminated waste, as it is assumed that PFAS are largely destroyed by this treatment method. Therefore, it is currently recommended that PFAS-contaminated fractions of textile waste are sent for thermal waste treatment if possible. The results of further investigations regarding possible airborne PFAS emissions are to be taken into account.
Landfilling/other treatment ~2%	In Germany, textiles are not allowed for landfill. Recommendations for landfilling are therefore not relevant.
SEWAGE SLUDGE **A total of ~1.74 million tons of sewage sludge was generated in Germany in 2020, corresponding to a total calculated PFAS load of ~117 kg (average PFAS load: ~67 µg/kg). Of this, ~22% was sent for material recycling, ~77% was disposed of thermally, and ~1% was disposed of in other ways.**	
Material recovery ~22%	When sewage sludge is recycled, it is used as a soil-related fertilizer in agriculture or in landscaping measures. PFAS are not destroyed during recycling, so they are generally released directly into the environment. Volatile PFAS were also detected in the air at the wastewater treatment plants, posing a potential risk to personnel. However, measured PFAS concentrations in the air of wastewater treatment plants are in the ng/m³ range and are thus far below typical PFAS occupational exposure limits in the mg/m³ range. Limit values for PFAS in sewage sludge already exist in the Fertilizer Ordinance (100 µg/kg for the sum of PFOS and PFOA), which reduces the input of these substances into the environment. The number of substances to be measured for a suitable limit value and the level of the limit value should be discussed. In general, it is recommended to determine the PFAS contamination of sewage sludges and then to subject the contaminated sludges to thermal treatment in order to destroy the PFAS as far as possible.
Thermal treatment ~77%	Currently, thermal treatment is the recommended disposal method for PFAS-contaminated waste, as the PFAS are largely destroyed by this treatment

Waste stream or disposal route	Risks and recommendations
	method (see also thermal treatment on textiles). Therefore, it is recommended that PFAS-contaminated sewage sludge be submitted to thermal waste treatment.
Landfilling/other treatment ~1%	Other direct disposal of sewage sludge includes, for example, delivery to drying plants and other unspecified disposal routes. Since the exact treatment routes are unknown, no concrete risks and recommendations can be derived. It is assumed that PFAS are not destroyed in drying plants, whereby the risks and recommendations of material recycling apply.
PAPER **A total of ~14.5 million tons of wastepaper was generated in Germany in 2020, corresponding to a total PFAS load of ~4,000 kg (average PFAS load: ~274 µg/kg). Of this, approx. 80% was sent for material recycling, approx. 19.7% was disposed of thermally and approx. 0.3% was landfilled.**	
Material recovery ~80%	During the recycling of wastepaper, the PFAS present are not destroyed. The PFAS are either carried over into new products or they enter local wastewater treatment plants via the water phase and finally partially enter the environment. Limit values for wastewater from the production of paper, board or cardboard already exist for various pollutants, but no limit values for PFAS are listed in Annex 28 of the Wastewater Ordinance. These could be established in the future. During recycling, volatile PFAS may be released into the air, posing a potential risk to personnel. It is possible that PFAS concentrations may be elevated in paper recycling facilities and facilities using recycled paper, but this should be verified by measurements. Composting of residues from the paper industry and their soil-related use should generally not be carried out. Furthermore, it is recommended to separate highly contaminated paper waste or paper waste with a high risk of containing PFAS from the other paper waste and to send it for thermal treatment. In this process, the PFAS are largely destroyed. Analogous to textiles, public awareness measures could be helpful here as well... According to the Packaging Act, PFAS-containing food packaging may not be disposed of in residual waste, but must be recycled together with lightweight packaging in the yellow garbage can.
Thermal treatment ~19,7%	Currently, thermal treatment is the recommended disposal method for PFAS-contaminated waste, as the PFAS are largely destroyed in this treatment method (see also thermal treatment to textiles). It is therefore recommended that PFAS-contaminated fractions of paper waste should also be sent for thermal waste treatment wherever possible.
Landfilling/other treatment ~0,3%	A small proportion of paper residues are still landfilled due to the system and thus PFAS can enter the leachate (The Paper Industry, 2020). The leachate can enter the soil of the landfill but is mostly discharged to the local wastewater treatment plant where it is treated. PFAS are not destroyed during landfilling but can enter the leachate of the landfill. Proper leachate treatment (*e.g.*, by activated carbon filter systems) can reduce the further spread of PFAS.
SOILS **A total of ~129 million tons of waste soils were generated in Germany in 2018, corresponding to a total PFAS load of ~2,300 kg (average PFAS load: ~18 µg/kg). Of this, ~85% was sent for material recycling, ~0.2% was thermally treated, and ~15% was landfilled.**	

Waste stream or disposal route	Risks and recommendations
Material recovery ~85%	The recycling of waste soils does not destroy the PFAS present. Waste soils are mostly backfilled. PFAS limits for the recycling of soils already exist. In general, PFAS can be leached from the contaminated soils and enter the groundwater. However, there are differences in the binding strength of soils, as some bind PFAS more strongly, but there is still a lack of concrete studies on this. It is therefore recommended to collect data on this. During excavation, volatile PFAS in particular may become airborne and be inhaled. Consideration could therefore be given to wearing suitable protective equipment (e.g. protective mask and gloves) during such operations. To determine the need for such protective measures, appropriate air measurements could be taken. It is generally recommended to remediate PFAS-contaminated soils before reuse in order to remove or destroy the PFAS contained. Various chemical and thermal processes are capable of effectively destroying PFAS, but these are often not yet established on a large scale, which is why there is still a need for research in this area. Alternatively, PFAS can also be immobilized, washed out or removed from the soil by pump-and-treat processes.
Thermal treatment ~0,2%	Currently, thermal treatment is the recommended disposal method for PFAS-contaminated waste, as the PFAS are largely destroyed in this treatment method (see also thermal treatment on textiles). Therefore, it is recommended that even heavily PFAS-contaminated fractions of soil waste should be submitted to thermal waste treatment if possible.
Landfilling/other treatment ~15%	Approximately 15% of the soil material generated as waste is landfilled. The same risks and recommendations apply as for the landfilling of other PFAS-containing wastes (see Landfilling of paper). PFAS are not destroyed during landfilling, but may leach into the landfill leachate. Proper leachate treatment (e.g., activated carbon filtration systems) can reduce the further spread of PFAS. Thermal treatment or disposal in suitable landfills with appropriate safeguards (base sealing, surface sealing) is the currently recommended form of disposal for contaminated soils. Likewise, underground disposal, e.g. in former mines, can be considered.

1 Background and objective

1.1 Background on the PFAS substance class

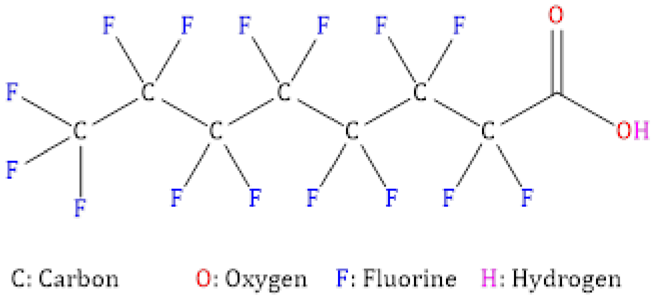
The compound class of per- and polyfluorinated alkyl compounds (PFAS) is characterized by the fact that they are compounds containing one or more carbon atoms on which all hydrogen atoms have been replaced by fluorine atoms. In this regard, per- and polyfluorinated alkyl compounds differ in the degree of fluorination (Buck et al., 2011).

Perfluorinated alkyl compounds are those compounds in which all hydrogen atoms bonded to carbon atoms have been replaced by fluorine atoms.

Polyfluorinated alkyl compounds are compounds in which all hydrogen atoms on at least one, but not all, carbon atoms of a compound have been substituted by fluorine atoms.

The basic structure of PFAS is shown in Figure 1 using the molecule perfluorooctanoic acid (PFOA). In this molecule, all hydrogen atoms (-H) on the carbon atoms (-C-) have been replaced by fluorine atoms (-F) and as a functional group this substance has an acid function (-COOH).

Figure 1: Structural formula of PFOA



Source: Own representation

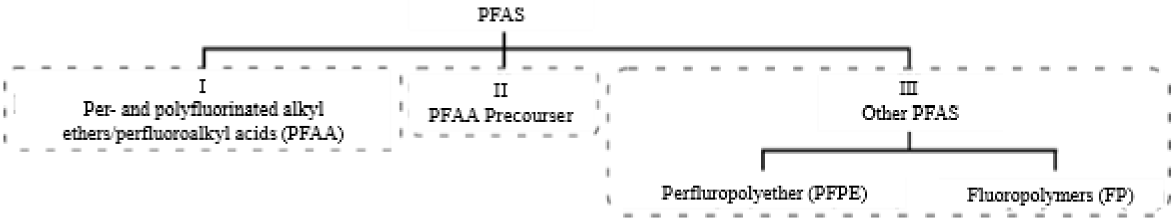
In addition to the term PFAS, the synonymous term per- and polyfluorinated chemicals (PFCs) is also used to refer to fluorinated compounds.

Because of the wide range of modification possibilities at the functional groups, which in turn have a direct influence on the functionality and properties of the compounds, there is a great variety of substances. For example, in pharmaceuticals, the introduction of fluorine atoms into drug molecules can increase fat solubility, improve the interaction of the substance with catalytic centers and delay metabolism. When used in technical applications, a positive effect on the materials used can also be observed in many respects. Here, mainly due to their high thermal and chemical stability as well as their both hydrophobic and lipophobic nature, these compounds have been used in a variety of products and processes since the 1950s. Examples include grease-resistant paper, surfactants, processing aids, specialty firefighting foams (C. Zhang et al., 2019).

This rough definition of the PFAS substance class can be further refined, e.g., based on the functional group, such as acid (-COOH) or alcohol (-OH) groups, which provides a simpler overview given the abundance of more than 4730 substances currently registered (OECD, 2018).

Such a classification was made by the OECD and the higher-level groups of this classification are shown in Figure 2.

Figure 2: Breakdown of per- and polyfluorinated compounds according to OECD.



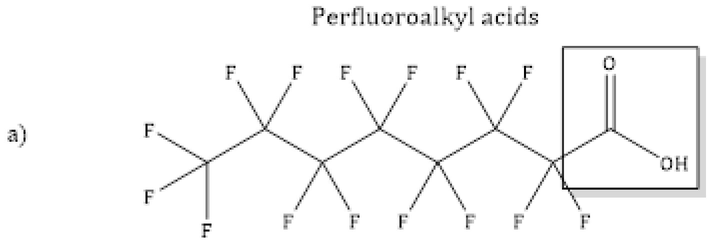
Source: Illustration adapted from OECD (2018)

The data shown in Figure 2 can be further refined and include the following compounds:

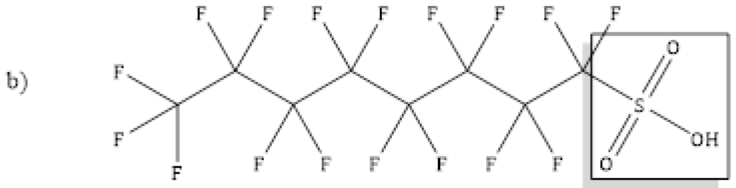
Per- and polyfluorinated alkyl ethers/perfluoroalkyl acids (PFAA)

The group of these compounds includes per- and polyfluorinated compounds that have as a functional group either an ether (Figure 4, C-O-C) or acid group (Figure 3, -COOH), the group of acids comprising various acids. For example, it can be a carboxylic acid (Figure 3 a, (-COOH)), a sulfonic acid (Figure 3 b, $(-SO_3H)$), a phosphonic acid (Figure 3 c, $(-PO_3H_2)$ or a phosphinic acid (Figure 3 d, $(-PO_2H_2)$.

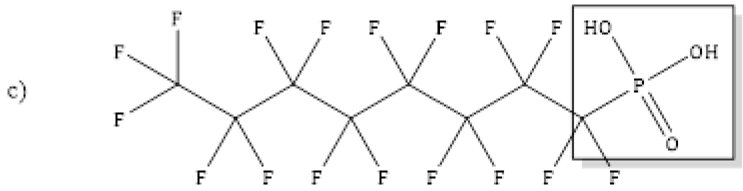
Figure 3: Subgroups of perfluoroalkyl acids according to OECD classification.



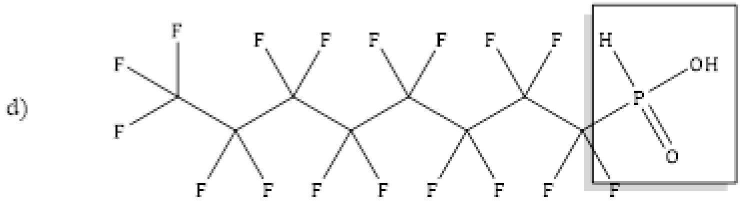
Perfluorocarboxylic acid (PFCA), e.g. perfluorooctanoic acid (PFOA)



Perfluorosulfonic acid (PFSA) e.g. perfluorooctanesulfonic acid (PFOS)



Perfluoroposphonic acid (PFPA), e.g. perfluorooctanephosphonic acid



Perfluorphosphinsäure (PFPiA), e.g. perfluorophosphinic acid

Source: Own representation

This subgroup includes perfluorooctanoic acid (PFOA) and perfluorooctanesulfonic acid (PFOS), the two best-known substances from the PFAS family. The high presence of these two substances in the literature (Waterfield et al., 2020), which deals with the environmental fate and toxicity of these two substances, follows from their high use levels and subsequent release. Due to the properties of PFOS to repel water and grease, it has been used as an impregnating agent, *e.g.*, for carpets or clothing, in chrome plating, or in firefighting foams. Due to its high environmental persistence, bioaccumulation as well as toxicity (Daly et al., 2018; Lofstedt Gilljam et al., 2016; Nian et al., 2020; Pelch et al., 2019; Waterfield et al., 2020) the production as well as the use of this substance were regulated as early as 2009 by inclusion in Annex B of the Stockholm Convention on Persistent Organic Pollutants (SC). In 2010, PFOS was included in Annex I of the POPs Regulation[1] (see chapter 1.2.3). Thus, PFOS is suspected of being carcinogenic (Waterfield et al., 2020). The release amount of this substance into the environment is estimated by Paul *et al.* (2009) already estimated at 45,250 t for the period 1970-2002. Due to the high residence time, this release already represents a major environmental risk and the substance can be detected in many environmental compartments (Paul et al., 2009).

The same applies to the substance PFOA. This substance has been used primarily as an emulsifier (Post et al., 2012). Due to similar properties as PFOS in terms of persistence, bioaccumulation and toxicity, this substance was also included in Annex A of the Stockholm Convention in 2019 and in Annex I of the POPs Regulation in 2020 [2](European Parliament, 2019). In this context, the total production of PFOA and its salts in the period 1951-2004 ranged from 3 600 - 5 700 t (Prevedouros et al., 2006). The high total production amount shows the relevance of this substance and the high potential to be released into the environment.

The second part of the first group of the OECD classification includes the per- and polyfluorinated ether carboxylic acids (at least one ether functional group (-O-) and one acid group (- COOH), Figure 4 (a)) and the per- and polyfluorinated ether sulfonic acids (at least one ether functional group (-O-) as well as one sulfonic acid group (-SO_3H), Figure 4 b)).

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[1] Amendment to Annex I POP Regulation: Commission Regulation (EU) No. 757/2010 of August 24, 2010, amending Regulation (EC) No. 850/2004 of the European Parliament and of the Council on persistent organic pollutants as regards Annexes I and III, available at http://data.europa.eu/eli/reg/2010/757/oj.

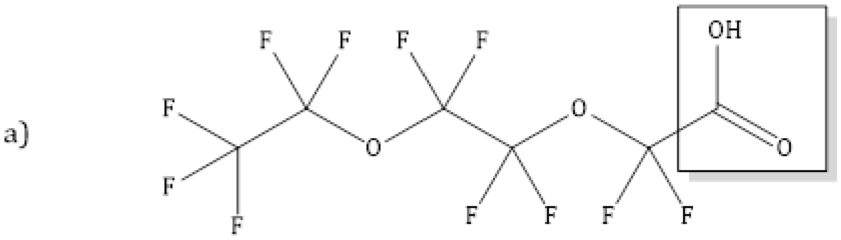
Amendment of Annexes IV and V of the POP Regulation: Commission Regulation (EU) No 1342/2014 of 17 December 2014 amending Regulation (EC) No 850/2004 of the European Parliament and of the Council on persistent organic pollutants as regards Annexes IV and V Text with EEA relevance, available at http://data.europa.eu/eli/reg/2014/1342/oj

[2] Amendment of Annex I POP Regulation: Commission Delegated Regulation (EU) 2020/784 of 8 April 2020 amending Annex I to Regulation (EU) 2019/1021 of the European Parliament and of the Council as regards the inclusion of perfluorooctanoic acid (PFOA), its salts and PFOA-related compounds, available at http://data.europa.eu/eli/reg_del/2020/784/2020-06-15

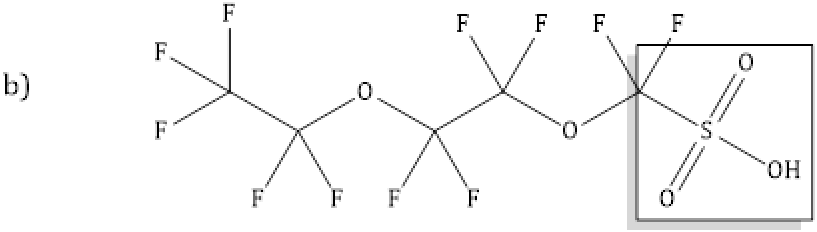
Amendment of Annexes IV and V of the POP Regulation: Commission Delegated Regulation (EU) 2021/115 of 27 November 2020 amending Annex I to Regulation (EU) 2019/1021 of the European Parliament and of the Council as regards perfluorooctanoic acid (PFOA), its salts and PFOA-related compounds, available at https://eur-lex.europa.eu/legal-content/EN/TXT/?uri=celex%3A32021R0115

Figure 4: **Subgroups of perfluorinated and polyfluorinated alkyl ethers according to the OECD classification.**

Per- and polyfluorinated alkyl ethers



Perfluoroether carboxylic acid (PFECA)



Perfluoroethersulfonic acid (PFESA)

Source: Own representation

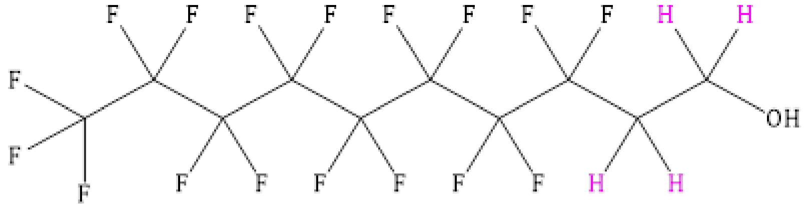
The substances in this subgroup have found significant use in the metal coating industry in China since the early 1970s (Lin et al., 2017; Ruan et al., 2015). With the regulation of both PFOS and PFOA, this class of substances has found increased application, *e.g.*, as emulsifiers (ADONA, (ammonium salt of perfluoro-4,8-dioxa-3H-nonanoic acid) or perfluoro(2-propoxypropanoic acid)) in fluoropolymer production, partially replacing PFOA there (Munoz et al., 2019; C. Zhang et al., 2019). Although these two substances are already found in the environment, including in German river systems (Heydebreck et al., 2015), research points to a need for definitive clarification of the toxicity of these substances (Gaballah et al., 2020).

PFAA Precursor

The second major group of the OECD classification comprises the fluorinated precursor substances. These are mainly substances that are used to manufacture the target substances. For example, perfluorooctane sulfonyl fluoride (POSF) is one of the precursor compounds of PFOS. Since these substances do not always convert 100 % during further processing, they can also be found in the end product and thus represent a burden for the environment. In a study conducted in 2009 by Paul *et al.* the authors report that the production of POSF had a worldwide production of 96,000 t in the period 1970-2002. With the inclusion of PFOS in Annex B of the Stockholm Convention, production at previous sites has largely ceased - however, due to exemptions allowed by Annex B, increased production in China is observable. Production there more than quadrupled from under 50 t per year to over 200 t per year between 2004 and 2006. Of this amount, 100 t per year was imported into the European Union, Brazil, and Japan (Knepper et al., 2014; Paul et al., 2009).

A large representative of this group are the fluorotelomers, which, like PFOA and PFOS, have a very high water- and grease-repellent effect and are therefore used, for example, in the textile industry (Gremmel *et al.*, 2016). The fluorotelomers are also widely used in the production of per- and polyfluorinated target compounds, such as polymers. An example of a common fluorotelomer is illustrated using 8:2 fluorotelomer alcohol (FTOH; the first number (8) indicates the number of fluorinated carbons, while the second number (2) indicates the number of non-fluorinated carbons), which is shown in Figure 5. Here, the general molecular formula for n:2 FTOH is $CF_3\ (CF\ )_{2n-1}\ CH_2\ CH_2\ OH$.

Figure 5: Structural formula of 8:2 FTOH



Source: Own representation

Because fluorotelomers can be metabolized and transformed, and thus the structural formula can change in the environment, it is difficult to assess and evaluate the environmental toxicological potential of fluorotelomers, but this is essential due to their occurrence (Butt et al., 2014; Field & Seow, 2017).

Other PFAS

The third group of the classification of PFAS according to the OECD includes the fluoropolymers and the perfluoropolyethers. Known representatives from this group are, for example, polytetrafluoroethylene (PTFE) or perfluoroalkoxy polymers (PFA), which are used, for example, as non-stick coatings.

1.2 Legal background

1.2.1 General overview and developments

The current EU regulatory landscape for PFAS is fragmented. Certain PFAS, such as PFOS, PFOA and PFHxS, are regulated in various EU legal instruments. Many of these legal instruments are EU regulations, which means that the regulations are directly applicable and also trigger direct obligations for industry and authorities. In addition to the presentation of the EU level, this chapter highlights those cases where German law regulates PFAS beyond the EU regulatory framework.

It should be noted that a number of Member States are currently preparing a restriction dossier for the whole group of PFAS under Regulation (EC) No 1907/2006 on the Registration, Evaluation, Authorisation and Restriction of Chemicals (REACH Regulation) (as of February 2023) (Annex XV, 2023b). Restricting the entire group of PFAS under REACH would have an impact on existing regulatory provisions on PFAS under other EU chemical and product legislation. However, the extent of this impact is not yet clear and will depend on the final form of the proposed restriction, if adopted.

1.2.2 Stockholm Convention

The Stockholm Convention (SC) aims to protect human health and the environment from POPs in accordance with the precautionary principle. To this end, different levels of regulation (elimination or restriction) are specified for the various substances. According to their level of regulation, the substances are assigned to three annexes of the Stockholm Convention.

Attachment A

Substances listed in this annex are to be completely eliminated from the markets of the respective contracting parties. In detail, certain time-limited exemptions for use as intermediates or other specifications are regulated for the listed substances.

Attachment B

The production and use of substances in Annex B are restricted in accordance with the specifications in the Annex. This is not a general prohibition, but a specific regulation for each use. In addition, there may be exceptions for the use of these substances.

There are currently three PFAS listed in the SC's appendices:

- Perfluorooctane sulfonic acid and its derivatives (PFOS) (Appendix B, Restriction) and
- Perfluorooctanoic acid (PFOA), its salts and PFOA-related compounds (Annex A with various exceptions, elimination).
- Perfluorohexane sulfonic acid (PFHxS), its salts and PFHxS-related compounds (Annex A without exceptions, elimination).

1.2.3 POPs Regulation

1.2.3.1 Production, placing on the market and use of POPs

As a party to the Stockholm Convention, the EU has committed to implementing the Stockholm Convention. This commitment is addressed under Regulation (EU) 2019/1021 (hereinafter referred to as POP Regulation) of the European Parliament and of the Council of 20 June 2019 on persistent organic pollutants.

Currently, three PFAS are listed in Annex I of the POPs Regulation:

- perfluorooctane sulfonic acid and its derivatives (PFOS) and
- Perfluorooctanoic acid (PFOA), its salts and PFOA-related compounds.
- Perfluorohexanoic acid (PFHxS), its salt and PFHxS-related compounds

According to Article 3 (1), the manufacture, placing on the market and use of these substances on their own, in mixtures or in articles are prohibited. It should be noted that this prohibition includes an exemption for substances present as unintentional trace contamination (UTC) in mixtures or articles.

For PFOS, Annex I of the POPs Regulation lists a UTC concentration of not more than 10 mg/kg (0.001% by weight) when present in substances. For semi-finished products or articles or components thereof, Annex I lists a UTC concentration for PFOS of less than 0.1% by weight, calculated in relation to the mass of structurally or microstructurally diverse components containing PFOS or, for textiles or other coated materials, if the PFOS content is less than 1 µg/m^2 of the coated material. The prohibition on the production, marketing and use of PFOS goes beyond the restriction on this substance set out in Annex B (Restriction) to the Stockholm Convention on Persistent Organic Pollutants, which lists various permitted purposes for production and use and specific exemptions for use that Parties to the Convention may apply for.

The ban on the production, marketing and use of PFOA is consistent with the listing of the substance in Annex A (Elimination) of the Stockholm Convention, which prohibits the production and use of PFOA subject to a limited number of specific exemptions. For PFOA or any of its salts, Annex I of the POPs Regulation lists a UTC concentration of no more than 0.025 mg/kg (0.0000025% by weight) when present in substances, mixtures or articles. For each individual PFOA-related compound or combination of PFOA-related compounds, Annex I lists a UTC concentration of no more than 1 mg/kg (0.0001% by weight) when present in substances, mixtures or articles. Annex I of the POPs Regulation lists a UTC concentration of no more than 1 mg/kg (0.0001% by weight) when PFOA and its salts are present in polytetrafluoroethylene (PTFE) micropowders produced by ionizing radiation or thermal degradation and in mixtures and articles for industrial and professional use containing PTFE micropowders[3] . If PFOA-related compounds are present in a substance that is intended to be used as a transported isolated intermediate as defined in Article 3(15)(c) of REACH in the manufacture of fluorochemicals with a perfluorocarbon chain of no more than six atoms and that meets strictly controlled conditions as defined in Article 18(4)(a) to (f) of that Regulation, a UTC concentration of no more than 20 mg/kg (0.002% by weight) applies.

The same limits that apply to PFOA also apply to PFHxS: 0.0025 mg/kg for PFHxS and its salts and 1 mg/kg for PFHxS-related compounds. Furthermore, a separate UTC of 0.1 mg/kg applies to the use of PFHxS and PFHxS-related compounds in firefighting foams, but this will be reviewed by the Commission in the future.

In addition to the UTC exemptions, Annex I of the POPs Regulation lists a number of more specific exemptions under the entries for both PFAS.

National law in the area of POPs is largely determined by EU law. Within the framework of national laws and sub-legislative regulations, apart from concretizing regulations and questions of enforcement, there are only isolated provisions that go beyond the European regulations in terms of content. The central legislative element of German substance law is the Chemicals Act (ChemG). This is where the provisions of European regulations are implemented/concretized. Under the ChemG, there are, in particular, the following ordinances with relevance for the regulation of POPs:

- Ordinance on Hazardous Substances (GefStoffV): regulates the classification, packaging and labeling of hazardous substances, as well as their handling and use,
- Chemicals Prohibition Ordinance (ChemVerbotsV): regulates the placing on the market or the restriction of the placing on the market of hazardous substances.

1.2.3.2 POP waste management

PFOS is also listed in Annex IV of the POPs Regulation, which is relevant for waste management, with a concentration limit of 50 mg/kg ("lower POP concentration limit" (LPCL) according to Article 7(4)a of the POPs Regulation). Similarly, PFOA and PFHxS are listed in Annex IV with an upper concentration limit of 1 mg/kg for the substance itself and its salts, and of 40 mg/kg for the PFOA- and PFHxS-related compounds. This means that, according to Article 7(2) of the POPs Regulation, all waste containing these PFAS above this concentration limit must be treated in a manner that ensures that the PFAS is destroyed or irreversibly transformed so that the remaining waste and releases do not exhibit the characteristics of the PFAS. In addition, producers and

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[3] With regard to this specific UTC, Annex I of the POPs Regulation specifies that all emissions of PFOA from the production and use of PTFE micropowders shall be avoided and, if this is not possible, reduced as far as possible. Furthermore, Annex I states that this exemption will be reviewed and evaluated by the Commission by 5/7/2022 at the latest.

holders of waste must make all reasonable efforts to avoid contamination of that waste with PFAS in accordance with Article 7(1) of the POPs Regulation. In addition to the LPCL, there is also an "upper POP concentration limit limit" (UPCL) according to Article 7(4)b of the POPs Regulation. This is defined in Annex V, Part 2 of the POPs Regulation as 50 mg/kg for PFOS. For PFOA and PFHxS, the limit is also 50 mg/kg for the substance itself and its salts, and a limit of 2,000 mg/kg for the PFOA- or PFHxS-related compounds.

Article 7(4) provides for two exceptions to the above obligations:

- First, waste containing or contaminated with these PFAS may be disposed of or recovered by other means in accordance with the relevant Union legislation, provided that the PFOS content in the waste is below the concentration limit of 50 mg/kg and 2,000 mg/kg, respectively.
- Secondly, in exceptional cases, a Member State or its designated competent authority may allow waste from thermal processes containing or contaminated with these PFAS up to 50 mg/kg or 2,000 mg/kg, respectively, to be permanently stored provided that a number of criteria set out in Article 7(4) and Part 2 of Annex V of the POP Regulation are met.

In Germany - in addition to the directly applicable regulations of the EU POP Regulation - according to the Federal Landfill Ordinance (DepV), waste containing POPs may not be disposed of above ground, *i.e.* only in underground landfills (all of which are located in salt rock) or by backfilling in salt rock. The upper POP limits are therefore not relevant for the disposal practice of POP-containing waste in Germany.

In other respects, the management of waste is governed by the Closed Substance Cycle Waste Management Act (KrWG). For the disposal of waste containing POPs with a content below the limit value in Annex IV of the EU-POP-V, it is of particular importance that, within the framework of the application of the waste hierarchy (§§ 6-8 KrWG), "priority shall be given to the measure which best ensures the protection of man and the environment in the generation and management of waste, taking into account the precautionary principle and the principle of sustainability. The entire life cycle of the waste is to be taken as a basis for the consideration of the effects on humans and the environment according to sentence 1" (§ 6 KrWG).

While certain ("old") POP wastes[4] are considered hazardous according to the German Waste Catalogue Ordinance (AVV), if the limits of the POP Ordinance are exceeded, and are thus subject to the corresponding requirements of the KrWG for hazardous waste, the POP Waste Monitoring Ordinance (POP-Abfall-ÜberwachV) applies to newer POPs. This was designed for the purpose of ensuring that all waste containing POPs is collected separately, not mixed, and monitored to a comparable extent, regardless of its classification as hazardous or non-hazardous waste. The types of waste listed in § 2 POP-Abfall-ÜberwachV are all assigned to non-hazardous "mirror entries" in the waste list.

The classification of PFAS according to the requirements of the CLP Regulation (EC) No. 1272/2008 (see chapter 1.2.5), in particular a harmonized classification, has in the area of the so-called "mirror entries"[5] effects on the classification of waste as hazardous or non-hazardous according to the AVV.

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[4] Annex to § 2 para. 1) 2.2.3: polychlorinated dibenzo-p-dioxins (PCDD) and polychlorinated dibenzofurans (PCDF), 1,1,1-trichloro-2,2-bis(4-chlorophenyl)ethane (DDT), chlordane, hexachlorocyclohexanes (including lindane), dieldrin, endrin, heptachlor, hexachlorobenzene, chlordecone, aldrin, pentachlorobenzene, mirex, toxaphene, hexabromobiphenyl or PCBs.

[5] Mirror entries are waste types in the Waste Catalogue Ordinance (AVV) which - depending on the specific individual case - are sometimes to be regarded as hazardous and sometimes as non-hazardous.

The classification of waste as hazardous in accordance with the AVV, in turn, triggers still additional specifications on the management of hazardous waste, for example with regard to permissible treatment options.

1.2.4 REACH Regulation

The following PFAS have been included in the candidate list[6] according to Article 59 of the REACH Regulation and are therefore substances of very high concern (SVHC):

- Perfluorobutane sulfonic acid (PFBS) and its salts;
- Perfluorohexane-1-sulfonic acid and its salts (PFHxS);
- perfluorodecanoic acid (PFDA) and its sodium and ammonium salts; and
- 2,3,3-Tetrafluoro-2-(heptafluoropropoxy)propionic acid, its salts and its acyl halides (HFPO-DA).

The addition of the above PFAS to the candidate list has a number of regulatory implications.

First, substances included in the candidate list are subject to evaluation for inclusion in Annex XIV REACH Regulation. According to Article 56 (1) REACH Regulation, a manufacturer, importer or downstream user may not place on the market or use for its own account a substance that has been included in Annex XIV for a use unless an authorization has been granted by the European Commission in accordance with the relevant provisions of the Regulation.

- Article 7(2) of REACH states that any producer or importer of articles must notify the European Chemicals Agency (ECHA) that a substance has been identified as a substance of very high concern if both of the following conditions are met:
    - a) The substance is present in these articles in quantities totaling more than one ton per year per producer or importer;
    - b) the substance is present in these articles in a concentration greater than 0.1% by mass (w/w).
- Article 31(1)(c) of REACH stipulates that the supplier of a substance or mixture must provide the recipient with a safety data sheet in accordance with Annex II of REACH if
    - a) the substance or mixture meets the criteria for classification as hazardous under CLP Regulation (EC) No 1272/2008, or
    - b) if the substance is persistent, bioaccumulative and toxic or very persistent and very bioaccumulative according to the criteria of Annex XIII REACH Regulation or
    - c) if the substance is included in the candidate list for reasons other than those listed in (a) and (b).
- Article 33(1) REACH Regulation states that any supplier of an article containing a substance on the SVHC list in a concentration higher than 0.1% by mass (w/w) must provide the recipient of the article with sufficient information available to the supplier to enable safe use of the article, including at least the name of this substance. Based on Article 9(2) of the Framework

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[6] https://echa.europa.eu/de/candidate-list-table

Directive 2008/98/EC on Waste (Waste Framework Directive), ECHA has established a database (SCIP) where suppliers must provide the data required by Article 33 (1) of REACH in accordance with national law.

- In Germany, implementation was discussed as part of the amendment to the KrWG. The version finally adopted (new § 16f ChemG) provides (1:1 in the wording of the EU Waste Framework Directive) for an obligation to "make available" the relevant information to the ECHA "as of January 5, 2021", but there are no specifications on the format and also no deadline by which the information must be made available. For the role of national chemicals legislation in relation to EU regulations outside this SCIP complex, see, mutatis mutandis, the comments in the area of POPs (see chapter 1.2.3).

Furthermore, long-chain perfluorinated carboxylic acids with a chain length of C9-C14 are restricted in Annex XVII, which means that these PFAS may no longer be manufactured or placed on the market. However, some exemptions exist such as for use in oil and water repellent textiles to protect workers from hazardous liquids, in the manufacture of PTFE and polyvinylidene fluoride for certain applications, and in photographic coatings for films. However, these exemptions all have a time limit, which means that C9-C14 perfluorocarboxylic acids in these applications must also be phased out.

As mentioned above, a restriction dossier is currently being prepared by a group of member states for all uses of all substances falling under the group of PFAS. Such a comprehensive restriction is likely to have consequences for the existing regulatory provisions on PFAS under the REACH Regulation, as described above. However, the extent of these consequences is not yet clear and will depend on the final form of the proposed restriction if adopted (status January 2023).

It should be noted that various other restriction dossiers for specific PFAS are being prepared by different member states (Rijksinstituut voor Volksgezondheid en Milieu RIVM, n.d.). Thus, there is a restriction procedure for PFHxA and their salts (ECHA, 2022b) and one for PFAS in firefighting foams (ECHA, 2022a). It is not clear how these efforts will relate to the comprehensive PFAS dossier mentioned above.

1.2.5 CLP Regulation

A number of PFAS have been included in the list of harmonized classification and labeling of hazardous substances (Table 3) of Annex VI of Regulation (EC) No 1272/2008 on classification, labeling and packaging of substances and mixtures (CLP Regulation):

- Perfluorooctanoic acid (PFOA),
- Ammonium Pentadecafluorooctanoate (APFO),
- Perfluorononane-1 acid (PFNA) and its sodium and ammonium salts,
- Perfluorodecanoic acid (PFDA) and its sodium and ammonium salts.

Inclusion in the list of harmonized classification and labeling of hazardous substances has a number of regulatory implications. CLP Article 17 (1) states that a substance or mixture classified as hazardous and contained in packaging must be accompanied by a label containing the elements specified in the same Article. The label must also comply with the labeling requirements laid down in other Articles of Title III of CLP Regulation. CLP Article 35 states that packaging containing hazardous substances or mixtures must comply with a number of requirements set out in the same Article.

On the role of national chemicals legislation in relation to EU regulations, see analogously the comments in the area of POPs (see chapter 1.2.3).

1.2.6 Water

1.2.6.1 Water Framework Directive

Directive 2000/60/EC establishing a framework for Community action in the field of water policy (Water Framework Directive, WFD) establishes a framework for the protection of inland surface waters, transitional waters, coastal waters and groundwater in the EU. Article 16 (1) of the WFD states that the European Parliament and the Council shall adopt specific measures against pollution of waters by individual pollutants or groups of pollutants presenting a significant risk to or via the aquatic environment, including such risks to waters used for the abstraction of drinking water. According to Article 16 (2), the European Commission shall submit a proposal containing a list of priority substances selected from those posing a significant risk to or via the aquatic environment. The substances are prioritized for action on the basis of the risk to or via the aquatic environment identified using an established risk assessment framework.

Article 16 (8) of the Water Framework Directive states that the European Commission shall, within two years of the inclusion of the substance concerned in the list of priority substances, submit proposals at least for emission controls from point sources and for environmental quality standards.

Annex X of the Water Framework Directive contains the above-mentioned list of priority substances. Entry 35 of the list lists PFOS and indicates that this substance is identified as a priority hazardous substance. Directive 2008/105/EC on environmental quality standards in the field of water policy (EQS Directive) sets environmental quality standards (EQS) for priority substances identified under the Water Framework Directive and lists PFOS in its Annex II with quality standards (limit values) for different types of surface waters and in biota.

In the context of a revision, the European Commission proposes to include 24 PFAS in the Water Framework Directive (European Commission, 2022b).

The WFD is implemented in Germany via the Water Resources Act (WHG) and ordinances based on it, such as the Groundwater Ordinance (GrwV) of 2010 and the Surface Water Ordinance of 2016 (OGewV). In Germany, the obligation to achieve good chemical water status arises from the aforementioned legal acts. The GrwV and the OGewV provide for the corresponding threshold values or quality standards. The OGewV implements the threshold values and quality standards for PFOS.

1.2.6.2 Drinking water guideline

Directive (EU) 2020/2184 on the quality of water intended for human consumption (Drinking Water Directive) is the first legal instrument in the EU to adopt a group approach regulating the totality of PFAS. However, this parameter value will only apply once technical guidelines for monitoring this parameter have been developed in accordance with Article 13(7). According to the Drinking Water Directive, these will be established by the Commission by January 12, 2024 (Article 13 (7)).

Article 4 (1) of the Drinking Water Directive states that Member States shall take the necessary measures to ensure that water intended for human consumption is fit for human consumption and pure. For the purposes of the minimum requirements of the Directive, water intended for human consumption is considered fit for human consumption and pure if a number of specified requirements are met. One of these requirements is that the water meets the minimum requirements set forth in Parts A, B, and D of Annex I.

Annex I, Part B (chemical parameters) lists a parameter value of 0.5 µg/L for the total of PFAS. The same Part B lists a parametric value of 0.1 µg/L for the sum of PFAS listed in Annex III, Part B, Number 3, which are considered to be of concern in relation to water intended for human consumption. Annex III, Part B, Number 3 of the Drinking Water Directive lists the following PFAS:

- Perfluorobutanoic acid (PFBA),
- Perfluoropentanoic acid (PFPeA),
- Perfluorohexanoic acid (PFHxA),
- Perfluoroheptanoic acid (PFHpA),
- Perfluorooctanoic acid (PFOA),
- Perfluorononanoic acid (PFNA),
- Perfluorodecanoic acid (PFDA),
- Perfluorundecanoic acid (PFUnDA),
- Perfluorododecanoic acid (PFDoDA),
- Perfluorotridecanoic acid (PFTrDA),
- Perfluorobutane sulfonic acid (PFBS),
- Perfluoropentanesulfonic acid (PFPS),
- Perfluorohexane sulfonic acid (PFHxS),
- Perfluoroheptane sulfonic acid (PFHpS),
- Perfluorooctane sulfonic acid (PFOS),
- Perfluornonan sulfonic acid (PFNS),
- Perfluorodecane sulfonic acid (PFDS),
- Perfluorundecane sulfonic acid,
- Perfluorododecane sulfonic acid,
- Perfluorotridecane sulfonic acid.

Article 13 (7) of the Drinking Water Directive states that by January 12, 2024, the Commission shall establish technical guidance on analytical methods for monitoring PFAS under the parameters "total PFAS" and "sum of PFAS," including detection limits, parameter values, and sampling frequency.

In Germany, the new version of the Drinking Water Ordinance will apply from 2023, which, after a transitional period from 2026, will set a limit value for the sum PFAS-20 of 0.1 µg/l and an additional limit value of 20 ng/l for the substance group PFOA, PFNA, PFHxS and PFOS (PFAS-4) from 2028.

1.2.7 Cosmetic products

Regulation (EC) No 1223/2009 on cosmetic products (Cosmetics Regulation) prohibits and restricts the presence of various PFAS in cosmetic products. Article 14 (1) of the Cosmetics Regulation states that cosmetic products must not contain, among others, prohibited substances listed in Annex II of the Regulation or restricted substances that are not used in accordance with the restrictions laid down in Annex III of the Regulation.

Annex II of the Cosmetic Products Regulation lists the following PFAS:

- Perfluorooctanesulfonic acid; heptadecafluorooctane-1-sulfonic acid (PFOS), and a series of salts:
  - Potassium perfluorooctane sulfonate; potassium heptadecafluorooctane-1-sulfonate
  - Diethanolamine perfluorooctane sulfonate
  - Ammonium perfluorooctane sulfonate; ammonium heptadecafluorooctane sulfonate.
  - Lithium perfluorooctane sulfonate; lithium heptadecafluorooctane sulfonate.
- Trifluralin (ISO); α,α,α-trifluoro-2,6-dinitro-N,N-dipropyl-p-toluidine; 2,6-dinitro-N,N-dipropyl-4-trifluoromethylaniline; N,N-dipropyl-2,6-dinitro-4-trifluoromethylaniline.
- Ammonium pentadecafluorooctanoate
- Perfluorooctanoic acid (PFOA)
- Fluazinam (ISO); 3-chloro-N-[3-chloro-2,6-dinitro-4-(trifluoromethyl)phenyl]-5-(trifluoromethyl)pyridin-2- amine
- Tembotrione (ISO); 2-{2-chloro-4-(methylsulfonyl)-3- [(2,2,2-trifluoroethoxy)methyl]benzoyl}cyclohexane-1,3- dione
- Triflusulfuron methyl; methyl 2-({[4-(dimethylamino)-6-(2,2,2-trifluoroethoxy)-1,3,5-triazin-2-yl]carbamoyl }sulfamoyl)-3-methylbenzoate
- Perfluorodecanoic acid (PFDA) and its salts:
  - Ammonium nonadecafluorodecanoate
  - Sodium nonadecafluorodecanoate
- Flocoumafen (ISO); reaction mass of: cis-4-hydroxy-3-(1,2,3,4-tetrahydro-3-(4-(4-trifluoromethylbenzyloxy)phenyl)-1-naphthyl)cumarin and trans-4-hydroxy-3-(1,2,3,4-tetrahydro-3-(4-(4-trifluoromethylbenzyloxy)phenyl)-1-naphthyl)cumarin
- Perfluorononane-1 acid and its
  - Sodium and
  - Ammonium salts
- Triflumizole (ISO); (1E)-N-[4-chloro-2-(trifluoromethyl)phenyl]-1-(1H-imidazol-1-yl)-2-propoxye-thanimine

Annex III of the Cosmetic Products Regulation lists the following PFAS:

- -N-(2-hydroxyethyl)-2-nitro-4-trifluoromethylaniline

In Germany, the Food, Commodities and Feed Code (Lebensmittel- und Futtermittelgesetzbuch - LFGB) is the umbrella law for German food and feed law. It covers all production and processing stages along the food value chain and also applies to cosmetics and consumer goods. The LFGB does not contain any additional relevant national regulations.

1.2.8 Food contact material

Commission Regulation (EU) No 10/2011 contains restrictions on the use of various PFAS in plastic materials and articles intended to come into contact with food.

Article 5(1) of the Regulation stipulates that only the substances included in the Union list of approved substances in Annex I to the Regulation may be intentionally used in the manufacture of plastic layers in plastic materials and articles. Table 1 in Annex I contains the list of authorized substances. Column 10 of the list lists restrictions and specifications for the use of the listed substances. All controlled PFAS in Annex I of Regulation (EU) No. 10/2011 are listed in Annex A of this document.

At the national level, the relevant implementation takes place within the framework of the LFGB. Additionally relevant are the Federal Institute for Risk Assessment (BfR) recommendations on food contact materials, which, although they do not constitute legal standards, represent the current state of science and technology for the conditions under which consumer goods made of high polymer substances, such as silicones, paper and rubber, comply with the requirements of Section 31(1) LFGB and Article 3(1a) of Regulation (EC) No. 1935/2004 with regard to their safety to health. In the BFR Recommendation XXXVI. for food contact papers, PFAS are currently still permitted on a positive list.

It should also be mentioned that for the tolerable daily intake for the sum of PFOA, PFOS, PFHxS and PFNA, a value of 4.4 ng/kg body weight has currently been determined (EFSA, 2022).

1.2.9 Industrial emissions

The Industrial Emissions Directive 2010/75/EU (IED) is the successor of, among others, the IPPC Directive of 1996 and establishes rules for the integrated prevention and control of pollution from industrial activities. A key tool under the IED is the Best Available Techniques (BAT) Reference Documents, which contain rules on BAT in the areas of industrial installations of particular environmental concern and are updated every 8 years. One chapter of the BREFs is dedicated to the so-called BAT conclusions, which, since the entry into force of the IED, have been transformed into legally binding documents during the revision of each BREF and have to be transposed into national law. Thus, BAT, some of which are linked to BAT-associated emission levels, must be considered and implemented in permit conditions for industrial activities covered by the IED. BAT conclusions from BREFs that have not yet been updated since the introduction of the IED are still given non-binding recommendations, but it may be the case that these BAT will also become legally binding when these BREFs are updated in the near future.

Currently, a number of BAT contain binding as well as (still) non-binding BAT conclusions on PFAS, in particular on PFOS. In Table 15 two BAT conclusions are listed as examples, the STM BREF of 2006 being (still) non-binding, and the WT BREF of 2018 being a binding BAT conclusion.

Table 15: Examples of BAT conclusions for PFAS

BREFs	Example reference to PFAS
Surface Treatment of Metals and Plastics (STM) (2005)	For PFOS, it is BAT to minimize use by controlling addition and to minimize vapors by controlling through techniques such as floating surface isolation. However, occupational health can be an important factor. Anodizing can be phased out and alternative techniques are available for chromium(VI) and cyanide-free alkaline zinc plating (see Section 5.2.5.2 of the BREF for detailed BAT and Section 4.9.2 for detailed descriptions of each technique).
Waste treatment (WT) (2018)	BAT 7. BAT is to monitor emissions to water [...]. PFOS must be monitored at least once every 6 months at all waste treatment facilities (Monitoring frequency may be reduced if emission levels show sufficient stability).

Source: Own representation

It should also be noted that the conclusions on best available techniques of the revised BREF for the textile industry include various PFAS-related limits for emissions to water and applicable water purification techniques for PFOS and PFOA. For example, wastewater with a high pollutant load (e.g., PFAS) should be collected and pretreated separately to minimize emissions to water (BAT 19) (European Commission, 2022a).

Plants which, due to their nature or operation, are particularly likely to cause harmful effects on the environment or otherwise endanger, significantly disadvantage or significantly disturb the general public or the neighborhood, require a permit in accordance with the Federal Immission Control Act (BImSchG). The 4th BImSchV[7] specifies which individual installations require a permit.

1.2.10 Application to soils

1.2.10.1 Sewage sludge

Council Directive 86/278/EEC of 12 June 1986 on the protection of the environment, and in particular of the soil, when sewage sludge is used in agriculture regulates the use of municipal sewage sludge in agriculture so as to prevent harmful effects on soils, vegetation, animals and humans while promoting the proper use of sewage sludge.

In Germany, EU Directive 86/278/EEC is implemented by the "Ordinance on the Reorganization of Sewage Sludge Utilization" (Sewage Sludge Ordinance - AbfKlärV). The national sewage sludge ordinance reorganizes the recycling of municipal sewage sludge in Germany. The input of inorganic and organic pollutants to soils is to be minimized. In addition, the aim is to return valuable components of sewage sludge (phosphorus) to the economic cycle more comprehensively than before. At the same time, soil-related sewage sludge utilization will be significantly restricted from 2029 onwards in order to further reduce the input of pollutants into the soil.

With regard to PFAS (polyfluorinated compounds with the individual substances PFOS and PFOA), testing obligations § 5(2) apply in the case of soil-related recycling before the sewage sludge is handed over to the sewage sludge user, the mixture producer or the compost producer. These obligations are specified in § 32 by analysis methods.

The release of sewage sludge by the sewage sludge producer and the application or incorporation of the sewage sludge on or into the soil is only permitted if the tests according to § 5 (1) and (2) show that the limit values according to Annex 2 Table 1.4 Column 4 of the Fertilizer Ordinance and the limit values according to the Sewage Sludge Ordinance are not exceeded. From

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[7] Fourth Ordinance on the Implementation of the Federal Immission Control Act

2029, plants with a population equivalent of more than 100,000 are obliged to treat their sewage sludge thermally. The same applies to plants with a population equivalent of over 50,000 from 2032 onwards. (LAGA, 2020). According to the Sewage Sludge Ordinance, the phosphorus must be recovered from the sewage sludge and used primarily in the form of fertilizers (see chapter 1.2.10.2) and must be returned to the cycle.

With a few exceptions, industrial sewage sludge does not fall within the scope of the Sewage Sludge Ordinance. The sewage sludge from some industries is spread on soils as fertilizer in accordance with the Biowaste Ordinance. The sludge produced during the treatment of industrial wastewater has to mostly be disposed of thermally.

In addition, it should be mentioned that the Soil Protection Ordinance (BBodschV) has been amended and will be valid from August 2023. This contains test values for the soil-groundwater pathway, including for PFAS (BMUV, 2022a).

1.2.10.2 Fertilizer

The German Ordinance on the Placing on the Market of Fertilizers, Soil Additives, Cultivation Substrates and Plant Auxiliaries (Fertilizer Ordinance - DüMV) of December 01, 2012 (updated 2019) regulates the approval and labeling of fertilizers. This includes limits for the approval (§ 3 (3)), placing on the market (§ 4 (3)) and labeling (§ 6 (5)) for perfluorinated surfactants (PFT). For the sum of PFOA and PFOS, a limit of 0.1 mg/kg DM and a labeling of 0.05 mg/kg DM or more apply according to Annex 2 Table 1.4. Restrictions of PFT for application aids (8.2.11)[8] and foreign ingredients (8.3.8)[9] are also specified.

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[8] no perfluorinated surfactants

[9] no perfluorinated surfactants

1.3 Objective

The use quantities of (intermediate) products containing PFAS show a steadily increasing trend. PFAS can enter the environment during the use phase but also during the disposal phase. According to the documented increase in production and use of PFAS-containing feedstocks, the amount of PFAS in the waste streams should also increase. The research project "Investigation of the occurrence of PFAS (per- and polyfluorinated alkyl compounds) in waste streams" starts at this point.

This project pursues two strategies in order to obtain as comprehensive a picture as possible of the current situation in the waste streams. (i) To identify the quantities used on the basis of a literature search and (ii) to investigate the PFAS contamination of particularly affected waste streams using analytical methods. Based on these investigations, (iii) the load of the waste streams will be calculated and (iv) a risk assessment will be carried out from these data. Finally, (v) considerations on waste-relevant limit values for selected waste streams will be made.

(i) In the first section of this work, potentially PFAS-contaminated wastes and possible input pathways are to be identified for the Federal Republic of Germany on the basis of existing literature. This includes the following waste streams for the entire report:

- Contaminated sites
  - Point sources in soil: Airports, firefighting foams, sewage sludge, agricultural land.
- Specialty paper waste
- Old textiles
  - e.g. outdoor clothing, shoes
- Construction waste
  - Spot sampling of construction market products
- Sewage sludge
- Washing solutions of a thermal waste utilization

This project aims to provide an initial identification, quantification, and assessment of the occurrence of PFAS in these waste streams and to present a basis for further research priorities and action needs.

Since no concentrations for individual PFAS substances could be obtained from PFAS measurements of the waste streams sampled in this project, it was decided, in consultation with the Federal Environment Agency, to research and evaluate suitable literature values to achieve the objective of the study.

2 Approach

This chapter describes the approach for the individual work steps.

In order to achieve the project goals, first of all, based on a background research (see chapter 2.1and 3.1), relevant waste streams were identified, a sampling plan was developed, and targeted sampling and subsequent chemical-physical analysis were performed, in which the organic fluorine is recorded in the form of the sum parameter (EOF) (see chapter 2.2, 2.3, 2.4 and 3.2).

Based on the measurement results and the PFAS concentrations to be derived from them (see chapter 3.2), PFAS mass flows in the respective waste streams were to be calculated and, based on this, considerations of possible disposal routes and possible PFAS limit values in the area of waste legislation were to be made. However, no concentrations for individual PFAS substances could be obtained from the PFAS measurements of the samples, but only sum parameters (see chapter 3.2). Thus, the basis for calculating PFAS mass flows and for questions based on this was missing.

In order to be able to calculate the mass flows and derive the waste limit values, it was therefore decided in consultation with the Federal Environment Agency to research and evaluate suitable literature values. For this purpose, an additional literature search was carried out for the selected waste streams (paper, sewage sludge, soils and textiles) (see chapter 2.5 3.3). The values found were extracted, averaged and used to calculate the mass flows (see chapter 2.7 and 3.4) were determined.

To estimate the environmental relevance, the selected waste streams were placed in an environmental context (see chapter 2.8 and 3.5) and considerations of possible limit values (see chapter 2.9 and 3.6) have been made.

Based on the results, recommendations for the establishment of possible waste management limits were derived (see chapter 3.6) and instructions for the public and for waste management enforcement were formulated (see chapter 3.7 and 3.8).

2.1 Literature review on PFAS background (studies considered, data sources, data identification).

The objective of the first work package of the project was to identify and evaluate meaningful studies containing information on the release of PFAS from production or waste streams. In order to create as broad an overview as possible, national and international reports from ministries and their subordinate authorities at state and federal level and non-governmental organizations (NGOs), specialist journals, books and databases were used.

The main objectives of the research were defined as follows:

1. Identify industries that use a high amount of PFAS.
2. Identification of release pathways
3. On the basis of 1) and 2), an estimation of the relevant waste streams
4. Selection of waste for further investigation

For this purpose, studies with different geographic boundaries were included to provide a better overview here.

Various databases and search engines were used to achieve the above goal. A listing of the main search engines used is shown in Table 16.

Table 16: Databases and search engines used for the literature search conducted.

Database	Explanation	Link
ECHA	European Chemicals Agency	https://echa.europa.eu
Google	Internet search engine	https://google.com/
Google Scholar	Internet search engine for general literature search of scientific documents	https://scholar.google.com/
PubChem	Free database of chemical compounds	https://pubchem.ncbi.nlm.nih.gov/
PubMed	Database on biomedical literature	https://pubmed.ncbi.nlm.nih.gov/
ResearchGate	Database on the Internet for researchers from all fields of science	https://www.researchgate.net/
Web of Science	Internet search engine for general literature search of scientific documents	https://apps.webofknowledge.com/
United States Patent and Trademark Office	Patent database	https://www.uspto.gov/
German Patent and Trademark Office - Homepage	Patent database	https://www.dpma.de/recherche/
Thru.com	List of German industrial installations covered by the Industrial Emissions Directive	https://www.thru.de/
Federal Statistical Office	Statistics database	https://www.destatis.de/

In the databases used, the following keywords and the different operators ("and", "or" and "not") were mainly used to obtain literature on the given topic, using both the German and English terms:

PFAS; Perfluorinated; Polyfluorinated; PFOS; PFOA; Environment(al); Toxicology; Waste; Stream; Production; Sewage sludge

For the combination of "*PFAS*", "*Sewage*" and "*Sludge",* this results in a number of hits of 27 publications in the Web of Science database when these three terms are combined with the operator "*And" and "Topic" is* selected as the search query. The corresponding publications were then preselected on the basis of the abstract and, if they matched the search objective, this article was either downloaded or obtained via interlibrary loan. The databases of Thru.de (A database maintained by UBA that provides environmental information from industrial plants and diffuse sources) and the German Federal Statistical Office were used primarily to estimate the quantities released.

2.2 Sampling

2.2.1 Old textiles

For the investigations presented here, textiles from the corresponding waste stream were used. For this purpose, randomly selected garments were taken from various disposal sites in Germany. The garments were finely comminuted using a granulator, homogenized, and then the organic bound fluorine was extracted and analyzed.

2.2.2 Sewage sludge

The sewage sludge investigated originates from various municipal and industrial wastewater treatment plants in Germany. Special attention was paid to the paper industry. Accordingly, sewage sludge from eight industrial wastewater treatment plants (KW1-KW8), which are in direct connection with the paper industry, and nine further sewage sludge (KW9-KW17) from municipal wastewater treatment plants, in which elevated PFAS contents had already been detected in the past, were investigated. The samples were taken by the wastewater treatment plant operator and, upon receipt, were freeze-dried, ground and homogenized. Subsequently, the organic bound fluorine was extracted and analyzed.

2.2.3 Washing solutions from thermal treatment of hazardous waste

The investigated aqueous solutions originate from the scrubbers of a thermal hazardous waste treatment plant in Germany and were taken by the operator. A total of 14 samples were taken from two scrubbers (basic/acidic) over a period of one month. The organically bound fluorine was extracted directly from the aqueous solutions and analyzed.

2.2.4 Paper waste

Two fractions from a waste aper sorting plant in Germany were used to investigate the EOF contamination of wastepaper. On the one hand, the graphic paper fraction (newspapers, magazines, brochures) and on the other hand the packaging paper fraction were sampled. In the case of the paper fraction, a duplicate was taken to check whether there was a strong inhomogeneity in the paper fraction. In all cases, approximately 10 kg of material per sample was taken from the respective stream, finely crushed using a granulator, and homogenized using a Röhnrad mixer. Subsequently, the organic bound fluorine was extracted and analyzed.

2.2.5 Soils

The soils investigated in this study come from different federal states: Four soils are from an area with known PFAS contamination (B1, B3, B4, and B5). Two soils are without known prior contamination (B2 and B7). Two soils were taken from landfills (landfill class II) (B6 and B8). One soil is from a soil wash (B9). Since the samples were taken by federal state agencies not all information is known to the authors of this report. The samples were freeze-dried and ground prior to analysis. The organic bound fluorine was then extracted and analyzed.

2.2.6 Hardware store products

The DIY products from different manufacturers were purchased in a local hardware store and the products were examined directly without a prior aging and weathering process. In each case, products for outdoor use were selected, which are thus subject to an environmental influx after one use. Mainly liquid-applicable materials were investigated. The materials are listed in section 3.2.6. The organically bound fluorine was extracted from the pure product and analyzed.

2.3 Extraction and purification used

The extraction of the organically bound fluorine was performed as described by Simon et al. (2022). For this purpose, 1 g of the sample was weighed and 4.9 mL of methanol and 0.1 mL of acetic acid were added to the sample. The suspension was then placed in an ultrasonic bath for five minutes, centrifuged for ten minutes, and the supernatant was pipetted off. These steps were repeated five times to ensure complete extraction of the organically bound fluorine. The solvent of the combined extracts was finally evaporated in a nitrogen stream and the residue was reabsorbed in a defined volume of a water/methanol mixture (1:1 (*V/V*)). The samples were diluted so that the concentrations were in the range of the calibration line and thus in the linear range of the measurement method used. Where necessary, SPE (solid phase extraction, HLB cartridge) was performed as described by Gehrenkemper et al. (2021) described. This was performed to remove fluoride from the aqueous phase, such as in the scrubber samples.

2.4 Sample analysis used

To consider the loading, the EOF (extractable organic fluorine) was determined using HR-CS-GF-AAS (high-resolution continuum-source graphite furnace absorption spectroscopy). This method and sum parameter were chosen to provide the most comprehensive view of PFAS loading. The high resolution allows the in-situ generated gallium fluoride to be measured at 211,248 nm. The measurement routine used is described in the publication "Determination of organically bound fluorine sum parameters in river water samples-comparison of combustion ion chromatography (CIC) and high resolution-continuum source-graphite furnace molecular absorption spectrometry (HR-CS-GFMAS)" by Gehrenkemper et al. For the measurement, the sample is placed in a furnace together with the molecule former (gallium nitrate) and a modifier mix (palladium nitrate, magnesium nitrate), which runs a temperature ramp to dry the sample (80-110 °C), pyrolyze it (500 °C) and then measure it (1550 °C). The furnace is then cleaned at 2450 °C. The measurements were carried out as a triple determination.

2.5 PFAS concentrations from the literature

PFAS concentrations in four selected waste streams were researched. At the start of the project, it was planned to calculate the mass flows based on the PFAS concentrations obtained in the project, but concentrations of individual substances could not be obtained because the concentrations in the samples were too low. Therefore, it was decided to summarize measured values from selected literature. Waste streams of textiles, papers, soils and sewage sludges were selected. The focus was on analytical data from Germany.

For this purpose, literature was systematically searched. Individual Boolean operators were created for each waste stream, coordinated with UBA and applied in PubMed as well as in the EuropePMC database. The retrieved studies were combined and duplicates were deleted. The following operators were used for the search:

Textiles:

(PFAS OR perfluoroalkyl OR polyfluoroalkyl OR fluorotelomer* OR "fluoro telomer*") AND (textile* OR cloth* OR carpet* OR upholstery) AND Germany*

Individual studies found: 241

Sewage sludge:

(PFAS OR perfluoroalkyl OR polyfluoroalkyl OR fluorotelomer* OR "fluoro telomer*") AND (sludge OR "sewage sludge") AND Germany*

Individual studies found: 77

Paper:

ABSTRACT:(PFAS OR perfluoroalkyl OR polyfluoroalkyl OR fluorotelomer* OR "fluoro telomer*") AND (paper OR ''paper material'' OR ''paper packaging'' OR "food contact material" NOT "current paper" NOT "this paper") AND Germany*

Individual studies found: 217

Soils:

(PFAS OR perfluoroalkyl OR polyfluoroalkyl OR fluorotelomer* OR "fluoro telomer*") AND (soil* OR earth OR clay OR dirt OR topsoil) AND Germany*

Individual studies found: 392

For the paper measurement data, the "ABSTRACT" function of the EuropePMC database was additionally applied, since food contact material (FCM) is very often mentioned in the context of PFAS as an application example. The search was therefore narrowed down to a mention in the short description, on the one hand to increase the relevance of the hits, but also to bring the number of hits to a manageable level.

In a second step, the studies received were checked for their relevance. For this purpose, first the title and then the brief description were analyzed for their relevance to the project. The program "Distiller" was used for this purpose. After the relevance check, the following number of relevant studies were identified: Textiles, 16; Sewage Sludge, 18; Paper, 27; and Soils, 37. It should be noted here that some studies were found to be relevant in more than one category, resulting in duplications. The relevant studies were downloaded and subsequently analyzed.

On the one hand, information relevant to the project was noted and, on the other, existing PFAS measurement data for the four waste streams/products were transferred to an Excel spreadsheet. Furthermore, reports from public institutions (Federal Environment Agency, State Offices for the Environment, etc.) as well as data from the environmental sample database were researched and reviewed. In total, PFAS concentrations relevant to the project were extracted from 20 studies.

Ultimately, PFAS soil samples were provided in anonymized form by the Rastatt District Office (Rastatt, 2022b). This is a data set with ~8,000 measurements, which were also recorded. Approximately 3,000 solid measurements and approximately 5,000 eluate measurements were taken.

2.6 Average PFAS concentrations

Average and median values were calculated from the PFAS concentrations collected in the literature. For this purpose, the measurements of the PFAS individual substances were summed up into groups for each of the samples. All perfluorocarboxylic acids (PFCA), perfluorosulfonic acids (PFSA), perfluorophosphoric acids (PFPA), and precursors were summed. Likewise, the groups were summed to give a ∑PFAS value. From the PFAS groups and the ∑PFAS value, average and median values were then calculated across all individual samples per waste stream.

2.7 Statistical waste data and PFAS mass flows

For the four selected waste streams (textiles, sewage sludge, paper and soils), the next step was to research waste data in order to calculate mass flows. Important for this is the amount of waste generated and the further treatment (recycling, thermal waste treatment, landfilling, etc.).

Waste data were obtained from the German Federal Statistical Office and Eurostat. There, data on soils and sewage sludge could be obtained. Data for the paper waste stream were obtained from a statistical report of the paper industry. Data for the textile waste stream were obtained from the statistics of the Federal Statistical Office and Eurostat on the one hand and from UBA and industry reports on the other. A description of the individual waste streams can be found in chapter 3.4.

The waste data found were then multiplied by both the average and median values of the PFAS groups and the ∑PFAS value to calculate a PFAS load. The results can also be found in chapter 3.4 can be found.

2.8 Presentation of the environmental context es for the selected waste streams

After calculation, the calculated PFAS mass flows of the four selected waste streams are put into an environmental context. For this purpose, a risk assessment is carried out, in which the following questions are considered in more detail:

- Are the disposal and recovery processes applied suitable to destroy or irreversibly transform the PFAS contained?
- Can new PFAS be generated during these processes or are the existing PFAS transformed?
- Do disposal or recovery operations possibly cause an environmental or health hazard (i.e. relevant exposure of humans or the environment)? Relevant limit values are also considered here. Emission of significant amounts of PFAS to the environment is generally considered a risk.

Possible exposure pathways are considered and it is discussed which processes in particular can lead to a risk to humans and the environment, or what should happen so that exposure can be avoided. Knowledge gaps are also identified.

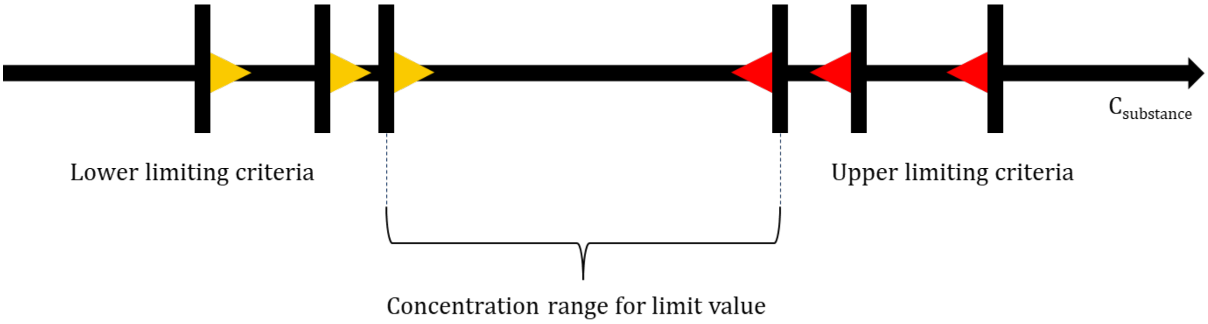
Emission and occupational exposure limits already exist for selected PFAS, which can be used for further consideration insofar as data on emissions and air concentrations are available.

2.9 Considerations for the derivation of limit values

Based on the calculated data, limit values for PFAS in the waste streams are calculated. For this purpose, the methodology for deriving POP waste limit values (Annex IV and V of the POP Regulation) is taken from Potrykus et al. (2015) is used.

The methodology limits the concentration range of a possible limit value of a substance downwards and upwards on the basis of various criteria. The following Figure 6 illustrates this approach:

Figure 6: Delimiting the concentration range of a substance for the derivation of limit values



Source: Own representation based on Potrykus et al. (2015)

2.9.1 Limiting criteria

Specific criteria are used to be able to define the concentration range. The lower limiting criteria are:

- **Analytical method**: The limit value should be analytically detectable and controllable
- **Background contamination**: The limit value should be above existing background contaminations of the substance in the environment
- **Disposal and recovery capacities**: The limit is to be set in such a way that there is sufficient availability for the (new) disposal routes required for recovery and disposal and their capacities
- **Economic impact**: The limit should be set so that the required disposal costs are economically justifiable

The upper limiting criteria are:

- **Limit values**: The limit value should not be contradictory to existing limit values
- **Potential environmental and health effects**: The limit value should be such that possible negative effects on the environment and health are avoided

2.9.1.1 Analysis method

A limit value must be analytically controllable. Therefore, appropriate measurement methods must be available whose limit of quantification is sufficiently low and costs are economical. A possible limit value must be above such typical limits of quantification and also in a range that is economically justifiable (very low limits of quantification are often possible, but such analyses cost significantly more).

The costs of the analyses often depend on the matrix to be examined, the effort and the number of substances to be measured. This is particularly important in the case of PFAS, since this is a group of substances and not a single substance. Decisive is finally the determination limit of usual samples of the considered matrix with usual effort. Costs of 500 € per single measurement are considered reasonable (analogous to (Potrykus et al., 2015)).

2.9.1.2 Background contamination

Possible limit values should also be above background contamination levels of the substance under consideration in the environment. If the limit value is below the background contamination, this would mean that virtually every sample would be above this limit value.

A distinction is made between typical background exposure and elevated exposures, e.g. near point sources. In general, an uncertainty factor of 10 is applied, since data on background exposure is limited. For this purpose, the highest background load found is multiplied by 10. (Potrykus et al., 2015). The limit value should not be below this value.

The values for the background load were determined on the one hand within the scope of the research for the PFAS concentrations, on the other hand data from other federal states could be obtained, which are used as a reference.

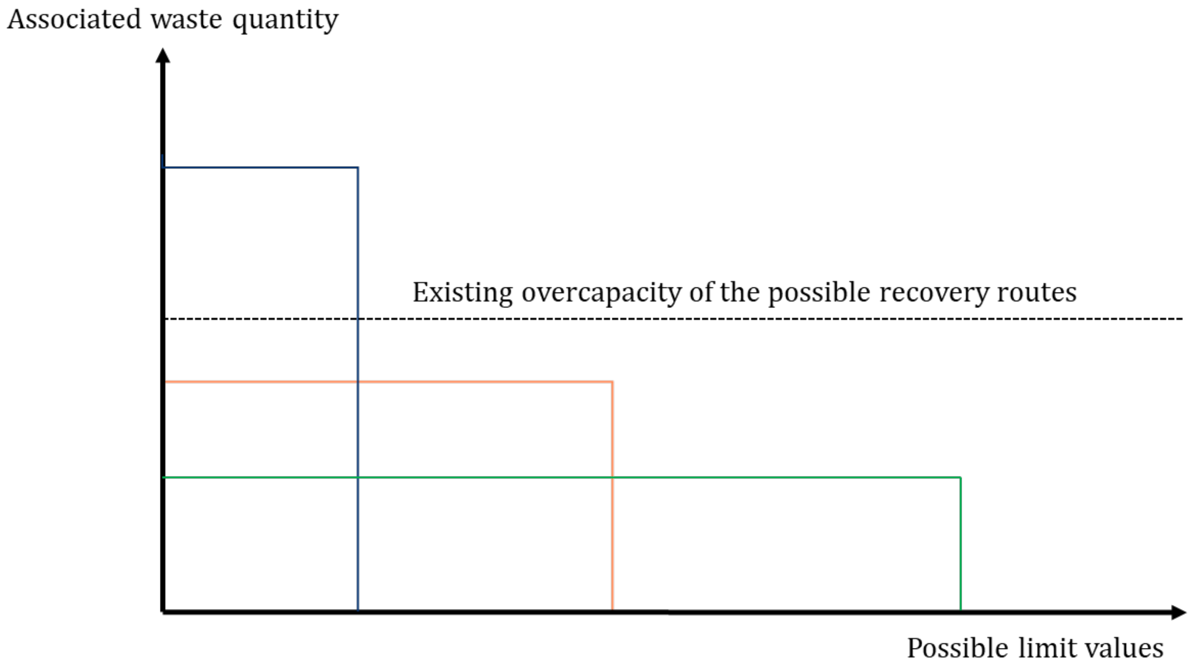
2.9.1.3 Disposal and recovery capacities

The limits should be set so that the necessary recycling capacity is realistically available for the waste above the limit.

In the case of PFAS, thermal treatment is the best method for destroying this class of substances. However, in certain cases, other methods can be used for decontamination, such as precipitation from the aqueous phase or remediation/decontamination of soils (see chapter 3.5).

For the limit value derivation, scenarios with possible limit values are first worked out and their effects on the treatment paths are analyzed. The limit value has a direct influence on the amount of waste to be disposed of. If the limit value is too low, a lot of waste has to be disposed of thermally and the available capacities are exceeded, which means that the waste can no longer be disposed of properly. Based on the available concentrations and waste quantities, possible limit values are worked out and the affected quantities are calculated. By graphically comparing the limit value and the associated waste quantity, suitable limit values can be read off. The following Figure 7 illustrates this relationship:

Figure 7: **Graphical representation of the determination of possible limit values based on the quantities of waste generated.**



Source: Own representation based on Potrykus et al. (2015)

2.9.1.4 Economic impact

The limits should be set so that the resulting economic impact is of a reasonable magnitude.

Additional costs arise if, for example, a waste may no longer be recycled but must be sent for thermal waste treatment.

If the limit value is exceeded, the waste concerned should be treated in such a way that the PFAS it contains are destroyed. For example, the POPs Regulation requires that contaminated waste above the limits listed in Annex IV "be disposed of or recovered in such a way that the POPs it contains are destroyed or irreversibly transformed". Studies show that thermal waste treatment can effectively destroy PFAS (see chapter 3.5), which is why this process is proposed for the destruction of PFAS.

For the economic impact, the price changes between old and new treatment should be considered. In a recent study commissioned by the BMUV, a cost range of 105-136€ for thermal waste treatment per ton of waste is given (Pohl et al., 2022). Due to limited capacities in Germany, prices have increased up to 200€/ton in 2021, but have eased in the turn of the year 2021/2022 of 2022 (Pohl et al., 2022). It can be assumed that due to the shortage of raw materials, in particular natural gas in 2022, as well as the inclusion of thermal waste treatment in the scope of the BEHG from 2023, the prices for thermal waste treatment will increase again. Potrykus et al. (2015) assumes 170€/ton of waste. Based on the current situation, this report also assumes 170€/t in disposal costs for thermal waste treatment.

For recycling give Potrykus et al. (2015) costs of 40€/t, which could be confirmed after internet research.

Potrykus et al. (2015) give a cost range of 60-220€/t for landfilling waste, which is in a similar range to the data obtained via internet research. An average value of 140€/t is given. For the further procedure, an average value of 150€/t for the landfilling of waste is considered realistic.

Thus, the following cost differences result for the treatment of waste above the limit in Table 17:

Table 17: Disposal costs for waste, as well as the price change if waste is above a potential limit value.

Current treatment	Treatment if the limit value is exceeded	Price difference per ton of waste
Recycling (40 €)	Thermal waste treatment (170 €)	130 €
Deposit in landfills (150 €)	Thermal waste treatment (170 €)	20 €

The project team proposes a cost increase of 1% in relation to the current disposal costs as economically justifiable. This value can be discussed and possibly adjusted in the future.

2.9.1.5 Limits

The limit values should be set in such a way that they do not conflict with other legal limit values. For this purpose, relevant national and international limit values are researched and presented. Based on these data, possible conflicts of a potential limit value can be identified and avoided.

2.9.1.6 Potential environmental and health impacts

Ultimately, the limit values should be set in such a way that harmful effects on humans and the environment can be avoided.

In general, hazardous substances can be emitted throughout the life cycle of a product, but this research project considers wastes, which places the focus on the disposal and recycling of PFAS-containing wastes.

PFAS have been shown to have harmful effects on humans and the environment, which has led to some already being listed as POPs (cf. chapter 1.2.3).

However, these effects can only be observed above a certain exposure concentration. The environmental concentration at which it is predicted that no adverse effects will occur is called the "predicted no effect concentration" (PNEC). Below this concentration, no negative (environmental) risks can be assumed.

Environmental risks can arise directly from the disposal of waste (e.g. the application of sewage sludge to arable soils as fertilizer). If PFAS concentrations are above the PNEC values, the negative effects can arise directly. On the other hand, humans and the environment may also be indirectly affected by PFAS. For example, PFAS may leak out after landfilling and enter the environment, or they may not be completely destroyed during thermal waste treatment and enter the environment via off-gas or ash. If the PNEC values are exceeded as a result, negative effects may occur.

For this project, PNEC values for PFAS are therefore researched and the precautionary principle is applied to the values found by considering the lowest PNEC values found for the derivation of the limit values.

2.10 Development of instructions for action

The aim is to derive recommendations for limit values and disposal routes on the basis of the results of the research project. On the one hand, these should ensure the greatest possible elimination of pollutants and, on the other hand, enable environmentally compatible treatment of waste and recycling processes in which risks to humans and the environment are to be avoided or largely reduced. This also includes advice on which disposal routes are suitable for destroying or irreversibly converting the PFAS contained in the waste. The basis for this is provided by the findings from the consideration of the environmental context (see chapter 2.8 and 3.5).

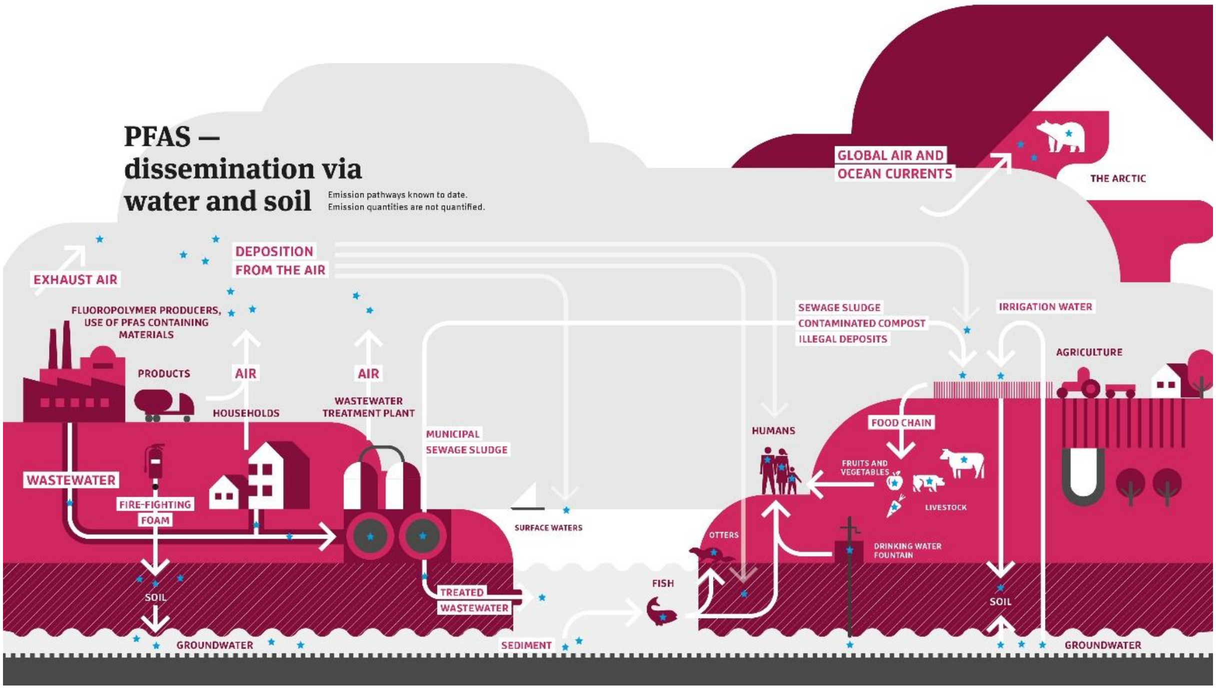
3 Results

3.1 Literature research on the substance group of PFAS

3.1.1 Production and use

The production volume of perfluorinated and polyfluorinated alkyl substances (PFAS) has followed a rapidly increasing trend in recent decades (Z. Wang et al., 2020). This trend toward greater production volume inevitably also increases the risk of release of these substances into the environment-both directly from production (*e.g.*, through industrial effluents) and through their use (*e.g.*, firefighting foams, products) or during the use of products or their disposal at the end of their useful life (*e.g.*, coated packaging) (Pabel et al., 2017). In this regard, each of these release pathways or the use of products associated with PFAS carries the risk of ingesting these substances and thus posing a direct risk to humans (Kingsley et al., 2019). This extensive network results in highly diffuse inputs to the environment, such as through landfills or wastewater treatment plants, making monitoring difficult. In addition to the multitude of exposure pathways, the high persistence and bioaccumulation properties of many of these substances lead to increased health risks for humans (Jain, 2013; Kingsley et al., 2018). These relationships are shown schematically in Figure 8.

Figure 8: **Schematic pathways of PFAS into waste streams, the environment, and human exposure.**



Source: (UBA, 2020a)

The flow chart illustrates the high pressure that lies on good downstream waste management, especially with regard to efficient degradation or retention of PFAS during wastewater treatment or in industrial wastewater treatment plants and in destroying these substances as far as possible during thermal waste treatment. Only by means of suitable pollutant sinks, such as thermal waste treatment, can an input into the environment be minimized and human exposure kept as low as possible. Here, however, it is also necessary to take a closer look at which parameters are required for complete thermal decomposition. A recent study from Australia by Coggan *et al.* (2019) shows that PFAS can pass through wastewater treatment plants largely unimpeded. Many other studies come to a similar conclusion, such as Clara *et al.* (2008, Austria) or Hamid *et al.* (2016, Canada). This can lead to the release of these substances via the treated wastewater into surface waters. Another part is transferred into the sewage sludge. In the recycling of sewage sludge, especially in the soil-related recycling process (Clara et al., 2008; Coggan et al., 2019; Hamid & Li, 2016), releases may also occur. The use of sewage sludge to is to be severely restricted in the future and, according to KlärV, will only apply to wastewater treatment plants with a population of less than 50,000 from 2032.

Once in the environment, removal of PFAS is extremely difficult to impossible and costly, so based on current knowledge, avoidance is the most promising strategy (German Federal Ministry for the Environment, Building and Nuclear Safety, 2017).

To get a rough overview of PFAS and tonnages per year imported or produced in the European Union (EU), the European Chemicals Agency (ECHA) database is particularly useful. (E. Database, 2021). To obtain the compounds relevant to this study, the database was searched using the keywords "*perfluoro*" and "*polyfluoro".* The substances found in this process, for which the tonnage range is indicated, are summarized Table 18.

Table 18: Substances contained in the ECHA database that can be extracted with the search term "perfluoro" or "polyfluoro".

Name	CAS number	Quantity range [t/year]
Perfluoro(methylcyclohexane)	355-02-2	10 - 100
Perfluoroethane	76-16-4	100 - 1 000
Perfluorooctane	307-34-6	0 - 10
1,1,1,2,2,3,3-heptafluoro-3-[(trifluorovinyl)oxy]propane	1623-05-8	100 - 1 000
1,1,1,2,4,5,5-nonafluoro-4-(trifluoromethyl)-3-pentanone	756-13-8	100 - 1 000
1,1,1,2,4,5,5-nonafluoro-4-(trifluoromethyl)-3-pentanone	756-13-8	1 000+
1,1,2,2,3,3,4,4-nonafluorobutane-1-sulphonic acid	375-73-5	0 - 10
3,3,4,4,5,5,6,6,6-nonafluorohexene	19430-93-4	100 - 1 000
Acetic acid, 2,2-difluoro-2-((2,2,4,5-tetrafluoro-5-(trifluoromethoxy)-1,3-dioxolan-4-yl)oxy)-, ammonium salt (1:1)	1190931-27-1	10 -- 100
Ammonium difluoro[1,1,2,2-tetrafluoro-2-(pentafluoroethoxy)ethoxy]	908020-52-0	10 - 100
Hexafluoropropene	116-15-4	10 000 - 100 000
Tetrafluoroethylene	116-14-3	10 000 - 100 000

Name	CAS number	Quantity range [t/year]
Trifluoro(pentafluoroethoxy)ethylene	10493-43-3	0 - 10
Trifluoro(trifluoromethoxy)ethylene	1187-93-5	100 - 1 000
Trifluoroacetic acid	76-05-1	1000 - 10 000
Trifluoroiodomethanes	2314-97-8	10 - 100
1,1,1,2,2,3,3,4,5,5,5-undecafluoro-4-(trifluoromethyl)pentanes	355-04-4	100 - 1 000
1,1,1,2,3,3-hexafluoro-2-(heptafluoropropoxy)-3-[(trifluorovinyl)oxy]propanes	1644-11-7	0 - 10
1,1,1,3,4,4,4-heptafluoro-3-(trifluoromethyl)butan-2-one	756-12-7	1 - 10
1,1,2,2,3,3,4,5,5,6-decafluoro-4,6-bis(trifluoromethyl)cyclohexanes	335-27-3	0 - 10
1-bromoheptadecafluorooctane	423-55-2	0 - 10
3,3,4,4,5,5,6,6,7,7,8,8-tridecafluorooctanesulphonic acid	27619-97-2	10 - 100
Perflunafene	306-94-5	0 - 10
Tetradecafluorohexane	355-42-0	10 - 100
Trifluoromethanesulphonic acid	1493-13-6	100 - 1000
1,1,2,3,4,4-hexafluorobuta-1,3-diene	685-63-2	10 - 100
Nonafluoro(trifluoromethyl)cyclopentane	1805-22-7	1 - 10
2,2,4-trifluoro-5-(trifluoromethoxy)-1,3-dioxoles	161611-74-1	1 - 10
2-Propenoic acid, γ-ω-perfluoro-C8-14-alkyl esters	85631-54-5	10 - 100
Reaction mass of perfluoro(dimethyl - N - butylamine) and perfluoro (methyl - di - N - propylamine) and perfluoro (dimethyl - N - propylamine) and 2,2,3,3,5,5,6,6, octafluoro-4-(trifluoromethyl)morpholine and perfluoro-N-pentane	-	10 - 100
(HFE-7100) Methyl nonafluorobutyl ether (40%) and Methyl nonafluoroisobutyl ether (60%) [AEGL-1].	163702-07-6	1 - 10
2H-tricosafluoro-5,8,11,14-tetrakis(trifluoromethyl)-3,6,9,12,15-pentaoxaoctadecane	37486-69-4	0 - 10
3-ethoxy-1,1,1,2,3,4,4,5,5,6,6-dodecafluoro-2-(trifluoromethyl)-hexanes	297730-93-9	10 - 100

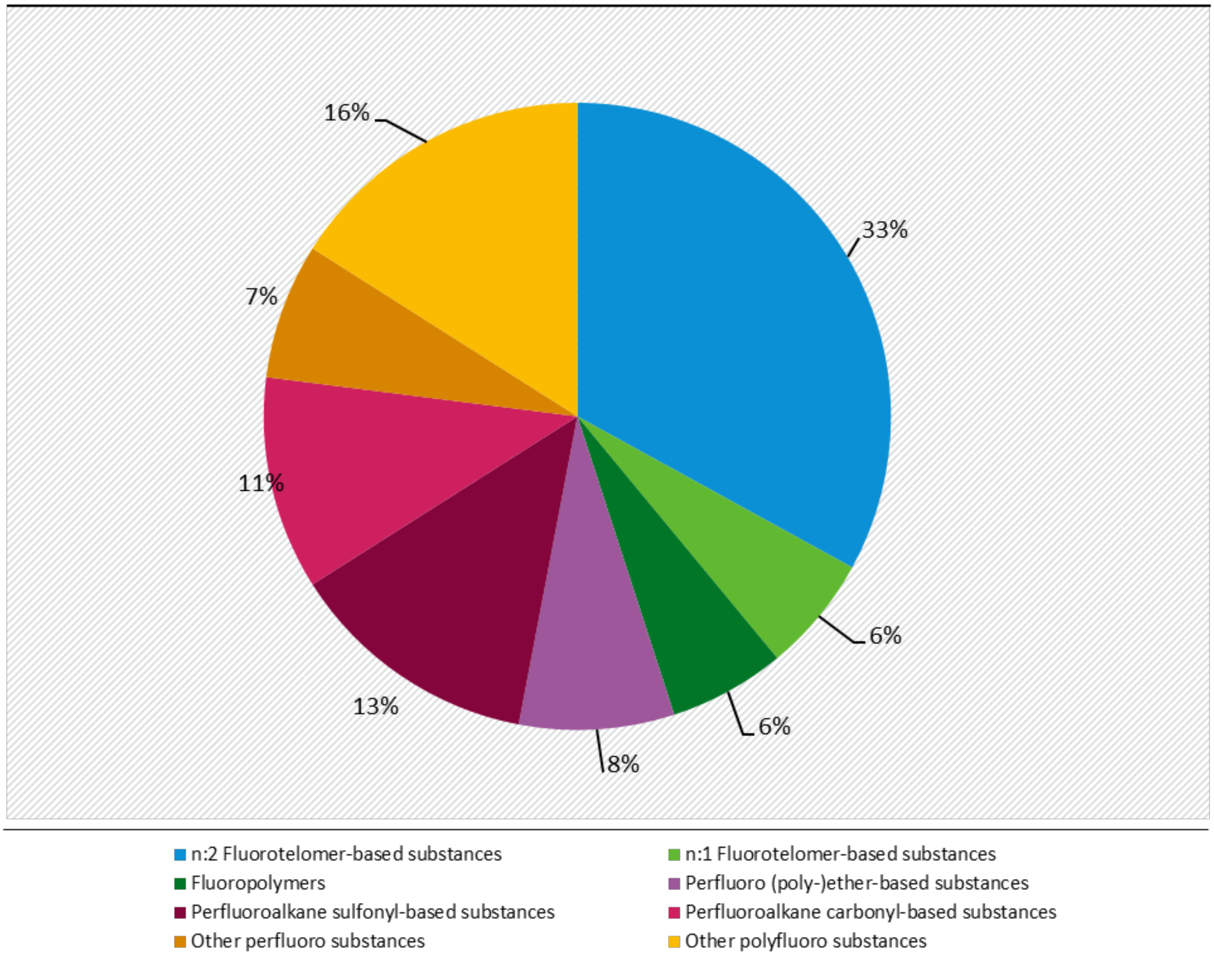
Name	CAS number	Quantity range [t/year]
Reaction mass of 1,1,2,2,3,3,3-heptafluoro-N-(heptafluoropropyl)-N-(pentafluoroethyl)propan-1-amine and 1,1,2,2,3,3,3-heptafluoro-N-(heptafluoropropyl)-N-(trifluoromethyl)propan-1-amine and 1,1,2,2,3,3,3-heptafluoro-N,N-bis(heptafluoropropyl)propan-1-amine	-	10 - 100
(E)-1,1,1,2,3,4,5,5-nonafluoro-4-(trifluoromethyl)pent-2-ene	3709-71-5	100 - 1 000
1,1,1,2,3,4,5,5,5-decafluoro-3-methoxy-4-(trifluoromethyl)pentane	132182-92-4	10+
1,1,2,2,3,3,4,4-nonafluoro-N-(2-hydroxyethyl)-N-methylbutane-1-sulphonamide	34454-97-2	100 - 1 000
3,3,4,4,5,5,6,6,7,7,8,8,8-tridecafluorooct-1-ene	25291-17-2	10 - 100
3,3,4,4,5,5,6,6,7,7,8,8-tridecafluorooctyl methacrylate	2144-53-8	100 - 1 000
Ammonium 2,3,3,3-tetrafluoro-2-(heptafluoropropoxy)propanoate	62037-80-3	10 - 100
Trichloro(3,3,4,5,5,6,6,7,7,8,8-tridecafluorooctyl)silanes	78560-45-9	10 - 100
Triethoxy(3,3,4,5,5,6,6,7,7,8,8-tridecafluorooctyl)silanes	51851-37-7	10 - 100
1,1,1,2,2,3,3,4,4,5,5,6,6,-tridecafluorotetradecane	-	10 - 100
Dichloromethyl(3,3,4,5,5,6,6-nonafluorohexyl)silanes	38436-16-7	0 - 10
N,N,N,-triethylethanaminium 1,1,2,2,3,3,4,4-nonafluorobutane-1-sulfonate	25628-08-4	0 - 10
Tetraethylammonium heptadecafluorooctanesulphonate	56773-42-3	0 - 10
3,3,4,4,5,5,6,6,7,7,8,8-tridecafluorooctyl acrylate	17527-29-6	100 - 1 000
3,3,4,4,5,5,6,6,7,7,8,8-dodecafluorodeca-1,9-diene	1800-91-5	0 - 10
2H-tricosafluoro-5,8,11,14-tetrakis(trifluoromethyl)-3,6,9,12,15-pentaoxaoctadecane	37486-69-4	0 - 10
1-[5-(2,6-difluorophenyl)-4,5-dihydro-1,2-oxazol-3-yl]ethanone	1173693-36-1	100 - 1 000

When adding up the quantity ranges of these substances, a value in the range of 10,000 - 100,000 t/year is reached. Since not all substances are listed in this database, the production volume of all PFAS can certainly be placed at the upper end of this scale. According to Janousek *et al.* (2019) it can be assumed that many more substitutes of short-chain PFAS are in circulation, which have not yet been registered. Another database for recording production volumes in the European region is the Substances in Products in the Nordic Countries (SPIN) database, which provides data for the countries Denmark, Finland, Norway, and Sweden (S. Database, 2021). The advantage of this database is that the scope of application of the chemicals is also given here.

3.1.2 Trends towards substitution

Due to the regulation or prohibition of the production and use of individual compounds from the PFAS substance class, a change in production towards shorter-chain substituents is observable (Wirth et al., 2019). For example, the production of PFOS at the largest PFOS producer 3M was discontinued from 3,300 t per year in 2000 to 2002 (Oliaei et al., 2013). Similarly, PFOS production at DuPont and the German chemical industry also ceased production of PFOS in 2002 (Stubleski et al., 2017; Wirth et al., 2019). According to a 2007 OECD report, PFAS production has shifted to shorter-chain compounds and, in this regard, mainly to compounds with a carbon chain length in the range of four to six carbon atoms (Oliaei et al., 2013; Stubleski et al., 2017). A similar trend was also observed in a study by the German Federal Environment Agency (UBA) (Wirth et al., 2019). One reason for this is the dependence of the stability of the compounds on the length of their carbon chain. Another reason for substitution is the progressive regulation and the tendency of these substances to be excreted from the organism more quickly when their chain length is shorter. Currently, more than 4,700 substances related to PFAS are registered in the Chemical Abstracts Service (CAS) Database with a corresponding number (Gaballah et al., 2020). The breakdown of these individual substances into the different substance classes are shown in Figure 9.

Figure 9: Overview of the percentage of registered PFAS in the individual substance classes.



Source: (OECD, 2018)

The graph shows the large proportion that n:1 and n:2 fluorotelomers (number of fully fluorinated carbons (n): Number of $-CH_2$ groups in the structure) account for compared to the total amount of PFAS. The great variety of registered substances shows how difficult it is to determine the load of PFAS. It also makes regulation more difficult, as the substitution of substances in the production process is relatively easy to realize due to the great abundance of substances.

3.1.3 Application range of per- and polyfluorinated compounds

The quantities used are extremely difficult to record due to the variety of applications and the numerous industries involved. Estimates for various branches assume the following quantities placed on the EU market. (German Federal Statistical Office, 2020; Wirth et al., 2019):

Intermediates: <10,000 t/year

Direct polymer production: <500 t/year

Surfactants C6: <500 t/year; C4: <200 t/year

Surfactants for firefighting: 1,500-3,000 t/year

Paper: <300 t/year

Textile treatment: <1,200 t/year

Surface treatment of hard materials: <1,000 t/year

These estimates are in good agreement with the industries identified by the U.S. Environmental Protection Agency (EPA) as major consumers of fluorotelomers. (U.S.E.P. Agency, 2009):

Textile/clothing industry

Carpet/Carpet Care Industry

Coating industry (paper, *etc.*)

Thus, the textile industry, the paper industry and the electroplating industry can be identified as the industrial sectors with the highest consumption. The fire extinguishing foams represent a special role, since the negative environmental impacts occur directly through their application.

Based on these figures, the initial assessment from the objective can be confirmed: the three named industries paper, textile/carpet and electroplating have the highest potential to be emitters of PFAS based on the quantities used. Due to the high production quantities and the partly short duration of use, *e.g.*, in the case of food contact papers, it can be assumed that these can also be found in the corresponding waste streams. In addition, during the production of these substances, they can enter the wastewater, so PFAS can occur in sewage algae here (Ahrens et al., 2010; Heydebreck et al., 2015).

3.1.3.1 Paper industry

For the paper industry, the Paper Industry Association for Germany states a total of 165 companies in 2018, which had a total production volume of 22 million tons. Of particular interest here is the production of water- and grease-repellent specialty papers. One type of papers that use PFAS are those that are in direct contact with food. Glüge *et al.* (2020) also describe other paper products such as tablecloths as an area in which fluorinated compounds are used. According to an estimate by UBA, special papers with these properties are produced in 5-10 paper mills in Germany. For Europe, the production volume of such papers is estimated at about 47,000 t for 2018 (UN Comtrade database)(United Nations, 2021).

The heavy use of PFAS in this industry can be explained both by their hydrophobic and lipophobic properties and by their very high resistance to decomposition, which is present even at high temperatures (Herzke et al., 2012; Schaider et al., 2017).

For example, studies by Trier *et al.* (2011), Gebbink *et al.* (2013) and Shoeib *et al.* (2016) detected elevated PFAS concentrations in food packaging made of paper or cardboard in their studies. In Germany, the BfR recommendation (Recommendation XXXVI. Papers, cartons and cardboards for food contact; and Recommendation XXXVI/2. Papers, cartons and cardboards for baking purposes 2019) applies to papers, cartons and cardboards for food contact, in which 21 PFAS compounds are listed with maximum permissible amounts. Currently, it is not known which substances from this list are actually used, so it can be assumed that all listed compounds of PFAS are used (Bokkers, 2018). The upper limits from this recommendation, as well as the data from the previously mentioned studies, suggest that PFAS are included here in the per mille to lower percent range (T. Held, 2015). Highly complex fluorinated polymers and perfluoropolyether dicarboxylic acids are mainly used here (T. Held, 2015). It can be assumed that PFAS are only used in the production of food contact papers with special properties. However, a contamination of the paper cycle takes place via the entry into the wastepaper cycle. Therefore, it is likely that there is contamination with PFAS in the material stream at a large number of paper manufacturing or converting plants. In this context, it is evident that the end product is highly relevant both during production and during disposal and recycling. Thus, sampling of the end consumer product, *e.g.*, packaging from the fast food industry, as well as of the production and its effluents and the recycling plants as possible emission pathways for PFAS is of interest here.

3.1.3.2 Textile industry

For the textile and clothing finishing industry, the Federal Statistical Office lists 45 establishments for 2019, with a production volume of 23,000 tons (Federal Statistical Office, 2020). Here, finishing is primarily used to make clothing waterproof and dirt-repellent (van der Veen et al., 2020). Other finishes in which these substances are used include flame retardancy, wrinkle resistance, and so on. Accordingly, the production of (outdoor) jackets is one of the main applications for PFAS in this sector. Other applications, which account for a very large proportion, are in the field of commercial clothing for emergency services such as police and firefighters and in hospital clothing (Gremmel et al., 2016; T. Held, 2015; Herzke et al., 2012; Peaslee et al., 2020). Here, a distinction can be made between two types of application. In the first case, impregnation takes place by means of so-called foulards during production. In the second, an application can be realized by means of appropriate impregnation sprays or as a washing additive (T. Held, 2015; Mumtaz et al., 2019).

A study by Supreeyasunthorn *et al.* (2016), in which washing water was investigated, comes to the conclusion that there is a gradual release of PFAS from the textiles tested, so that a release must be assumed here, particularly in the case of new or freshly impregnated clothing, which thus represents a direct input pathway into wastewater. A similar assessment was made in a 2014 study by the Federal Environment Agency (Thomas P Knepper et al., 2014).

Gremmel *et al.* come to a similar conclusion in their study from 2016. They were able to detect increased PFAS concentrations in textiles and also observed a strong dependence of the concentration with the "history" (age, storage conditions, *etc.*) of the product.

Fluorotelomers can be detected particularly strongly in this product group, which can be attributed to their strongly water- and grease-repellent properties.

Several interesting waste streams emerge from the data: As in the previous chapter on paper, direct wastewater or the consumer product at the end of its life cycle can provide an emission here. In the case of textiles, however, the slow release due to the repeated washing cycles and, if necessary, repeated impregnation is added here. This also makes municipal wastewater or wastewater from large laundries interesting for sampling.

3.1.3.3 Electroplating

For electroplating and metal finishing, 181 establishments are listed for 2018, but for which no data on production volume is available (Federal Statistical Office, 2020). However, the actual number is difficult to record, for example, the Zentralverband Oberflächentechnik e.V. (Central Association for Surface Technology) estimates the number of electroplating companies for 2017 at approx. 2400 (Willand et al., 2020). By using electroplating, material properties of a workpiece are improved by electrochemical deposition of metallic layers or an embellishment is made (T. Held, 2015). Two of the main processes in which PFAS, especially PFOSs, are or have been used are the chrome plating of metals and the pickling of plastics with process solutions containing chromium trioxide (Blepp et al., 2016). However, PFAS could also be found in other galvanic processes besides chrome plating, such as galvanizing or copper plating (T. Held, 2015). In these wet chemical surface finishing processes, PFAS are used as wetting agents. The reason for the use of wetting agents in this industry is occupational safety, as gas bubbles are formed at the anodes, which can then migrate to the surface, burst, and throw chromium trioxide, which is classified as a carcinogen, into the air. The wetting agent is used to reduce the size of the bubbles and thus reduce the ejection of chromium trioxide (Blepp et al., 2016; T. Held, 2015). For hard chromium plating, the largest amount of PFOS was used in Germany in terms of volume - approx.

50% (Blepp et al., 2016). However, the mainly used PFOS exhibits a long-lasting desorption process, so that a slow release of PFOS can be observed even after switching to other substances - the so-called bleeding of the plant (Blepp et al., 2016). Due to the restrictions on the use of PFOS, this substance has been largely substituted by 6:2 FTS (6:2-fluorotelomer sulfonic acid) in electroplating, so that the possibility of its release exists (Willand et al., 2020).

These figures show that these industries are strongly represented in the Federal Republic of Germany, both in terms of the number of companies and the quantity of PFAS used. It follows from this that, given the intensive use of PFAS by these industries as reported in the literature, release is possible. In particular, release during the production process into wastewater and as sludge from the industrial pretreatment plant is possible as an input pathway.

3.1.3.4 Building materials

In building materials, PFAS can be found in many different products, *e.g.*, paints and varnishes, to protect against soiling of house facades, and in sealants and adhesives (Janousek, Lebertz, et al., 2019). Janousek, Lebertz, et al. investigated the release of PFAS with respect to 29 target analytes from building materials in a 2019 study. Their study was able to show that there is a release of the target analytes from the studied building materials. In a study conducted by Knepper *et al*. (2020) the authors were able to show that PFAS are contained in the products (e.g. paints, coatings) of various building products according to the data provided. In the 23 building materials examined, PFAS could be detected, for example, in three of four coatings examined (Knepper & Janousek, 2020).

No further data could be found on the release from this product group, so that an estimate of the relevance of this group compared to the previous three (paper, textiles, electroplating) cannot be conclusively clarified. A possible strategy here would be to sample soil near a newly constructed building after a rain event. Furthermore, a long-term observation, i.e., sampling prior to the construction/rehabilitation of a structure and subsequent repeated sampling at the same location, would be of interest to determine the release. Since the release is a direct consequence of the PFAS content in the building products, these can also be measured directly accordingly (after a review of the safety data sheets, as was carried out in the study by Knepper *et al.*) or the amount of PFAS in building rubble can be determined in order to record an exposure situation here.

3.1.4 Entry paths

Due to the highly explosive nature of this class of compounds and the high quantities used, a large number of studies have been carried out to investigate their fate and effects in the environment and on humans.

3.1.4.1 Surface water

In addition to the input of PFAS to the environment through wastewater, there is also contamination of surface water through the application of PFAS-contaminated substances such as sewage sludge and through the use of PFAS-containing fertilizers or firefighting foams (Houtz et al., 2013; McCarthy et al., 2017; Nickerson et al., 2020; Pulkrabová et al., 2019). In a study from the United Kingdom, Paul *et al.* (2009) the global total amount released (1970 - 2002), i.e., both directly through production and indirectly through households, of PFOS to be 450-2,700 t running through waste streams. In this study, it is stated that this amount is mainly released to the aquatic environment (Paul et al., 2009).

Some studies refer to rivers, since here the adjacent industry and wastewater treatment plants partly discharge these substances due to inadequate treatment, which can lead to an increased release: In a 2007 study by McLachlan *et al. in* which 14 different rivers were sampled, concentrations for PFHxA of 0-32 ng/L, PFHpA of 0.2-6.6 ng/L, for PFOA of 0-200 ng/L, and for PFNA of 0-1.50 ng/L were measured using HPLC-ESI-MS/MS. The highest load was found in the Po River. Based on the data obtained, releases of the substances were calculated to be 2.8 t/yr (PFHxA), 0.86 t/yr (PFHpA), 14.3 t/yr (PFOA), and 0.26 t/yr (PFNA), with the high concentrations in the Po River attributed to local industry (e.g., textile industry) as a possible polluter (McLachlan et al., 2007). A river system that has been and continues to be studied many times because of its economic importance is the Rhine. In a 2019 publication by Janousek, Mayer, et al. the Rhine was sampled over five years in a monthly rhythm and the composition especially of PFAS was followed. This shows a decreasing trend of the tested substances. Most clearly, this trend can be observed from the year 2016 to the year 2017. Furthermore, a strongly decreasing PFOS and PFOA concentration can be observed. While the sum of the analyzed PFAS was still up to 80 ng/L in 2014, it decreased to below 10 ng/L in 2018. However, observe Janousek, Mayer, *et al.* in this study observed an increase in the concentration of mainly short-chain substances, so that on the one hand a clear trend towards substitution can be identified here based on the effectiveness of the legislation banning certain substances in river systems (Janousek, Mayer, et al., 2019). A similar conclusion was reached in a study conducted in the German state of Bavaria on the Alz River. On this river, PFOA was produced until 2003 and in 2006 a PFOA concentration of 5-8 µg/L could be detected in the river water. In 2016, PFOA could no longer be detected (detection limit of the method used in the study 0.02 µg/L), which can be attributed to the fact that PFOA has hardly been used since 2016 (Bavarian State Office for the Environment, 2020). Instead of PFOA, the fluorine-containing substitute ADONA (ammonium salt of perfluoro-4,8-dioxa-3H-nonanoic acid) is now used (Bavarian State Office for the Environment, 2020).

A study carried out in 2017 by Metzger *et al.* came to a similar conclusion, in which the Rhine was also sampled and the EOF was recorded here. Samples were taken at 14 sites and concentrations between 50 and 300 ng/L were detected. The results of this study speak for local point sources where a large PFAS load is discharged to rivers (Metzger et al., 2019).

Studies from the year 2017 by Heydebreck (2017) of samples from treated wastewater discharged into the Rhine River show the crucial role of wastewater treatment plants: in the study, PFAS concentrations (sum of 12 substances) of up to 4 290 ng/L were measured, while comparative measurements in the river in the immediate vicinity showed about 80 ng/L due to dilutions (Heydebreck, 2017).

A similar finding was reached in a study from Baden-Baden, in which 10 wastewaters from wastewater treatment plants from this region were examined - here, an emission of 540 g of organic fluorine per day was determined in total for all 10 examined wastewater treatment plants from the municipal sector on the basis of the measured data. In this study, in addition to target analytes, the AOF of the samples was also determined. The studies conducted in this way showed that only about 2% of the total load could be explained by the analysis for known substances and 98% of the organic fluorine originated from unknown compounds (Rastatt, 2018). A similar trend is also shown for wastewater treatment plants in Halle (Westphalia), whose PFAS concentration in the effluent exceeds that of the Weser River by a factor of 4, which can be attributed primarily to the high dilution.

A 2017 study/compilation of data by Shafique *et al.* provides an overview of PFAS contamination in drinking water worldwide. For European sampling sites, PFAS concentrations were found to range from 2 ng/L (Faroe Islands) to 33 ng/L (France). Similar high levels could also be measured in Brazil (36 ng/L) (Shafique et al., 2017). In India (14 ng/L) and Australia (13 ng/L), the load is significantly lower at the sites studied. Concentrations are significantly higher in Kenya, where concentrations of over 100 ng/L were measured. The studies also show that the proportion of PFBS as a substitute for PFOS in the samples is a larger proportion (Shafique et al., 2017). From this, due to the increasing use of substitutes, it is clear that extended monitoring is needed. Another point that stands out in the individual studies is that there is no uniform choice of substances/parameters, so that it is difficult to compare the totals formed and, if only a few substances are selected, the total load cannot be represented. This again speaks in favor of a uniform sum parameter with, if necessary, downstream single substance analysis.

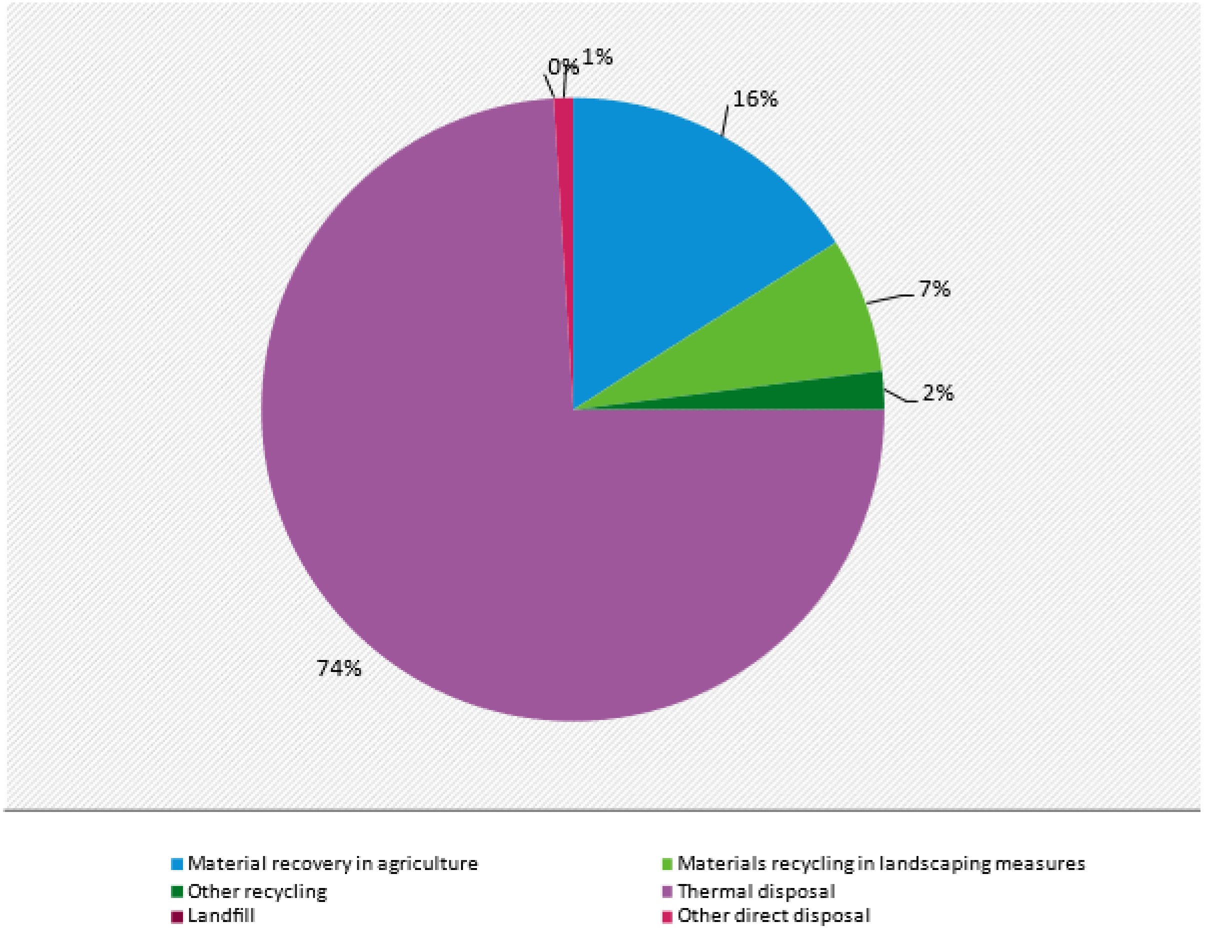
3.1.4.2 Sewage sludge and treatment plant effluents

Investigations of municipal and industrial wastewater treatment plant effluents on the Rhine and in the region of Baden-Baden and Halle (Westphalia) showed that some of them had greatly increased PFAS concentrations (cf. chapter 3.1.4.1). Closely related to the effluent of a wastewater treatment plant is the sewage sludge (Hamid & Li, 2016; Saez et al., 2008; Yan et al., 2012). This, in turn, is closely related to soils and surface water when it is used soil-related, e.g., as fertilizer. For this reason, sewage sludge is increasingly the subject of studies (Alder & van der Voet, 2015; Campo et al., 2014; Clara et al., 2008; Coggan et al., 2019; Hamid & Li, 2016; Llorca et al., 2011; Milinovic et al., 2016; Ruan et al., 2015; Sindiku et al., 2013). A limit value for application as fertilizer to soils has already been established (100µg/kg for the sum of PFOS and PFOA, see chapter 1.2.10.2). This limit value applies both to utilization as fertilizer and to landscaping measures.

3.1.4.2.1 Waste treatment and sewage sludge disposal

The disposal of sewage sludge is divided into six categories by the Federal Statistical Office (Federal Statistical Office, 2020): material recycling in agriculture; material recycling in landscaping measures; other material recycling; thermal disposal; landfilling; other direct disposal. For the 1.7 million tons generated in 2018, the breakdown into these categories are shown in Figure 10. According to the Landfill Ordinance (as of 2011), direct disposal of sewage sludge to landfills is only permitted after thermal pretreatment - accordingly, this value for 2018 is 0 tons.

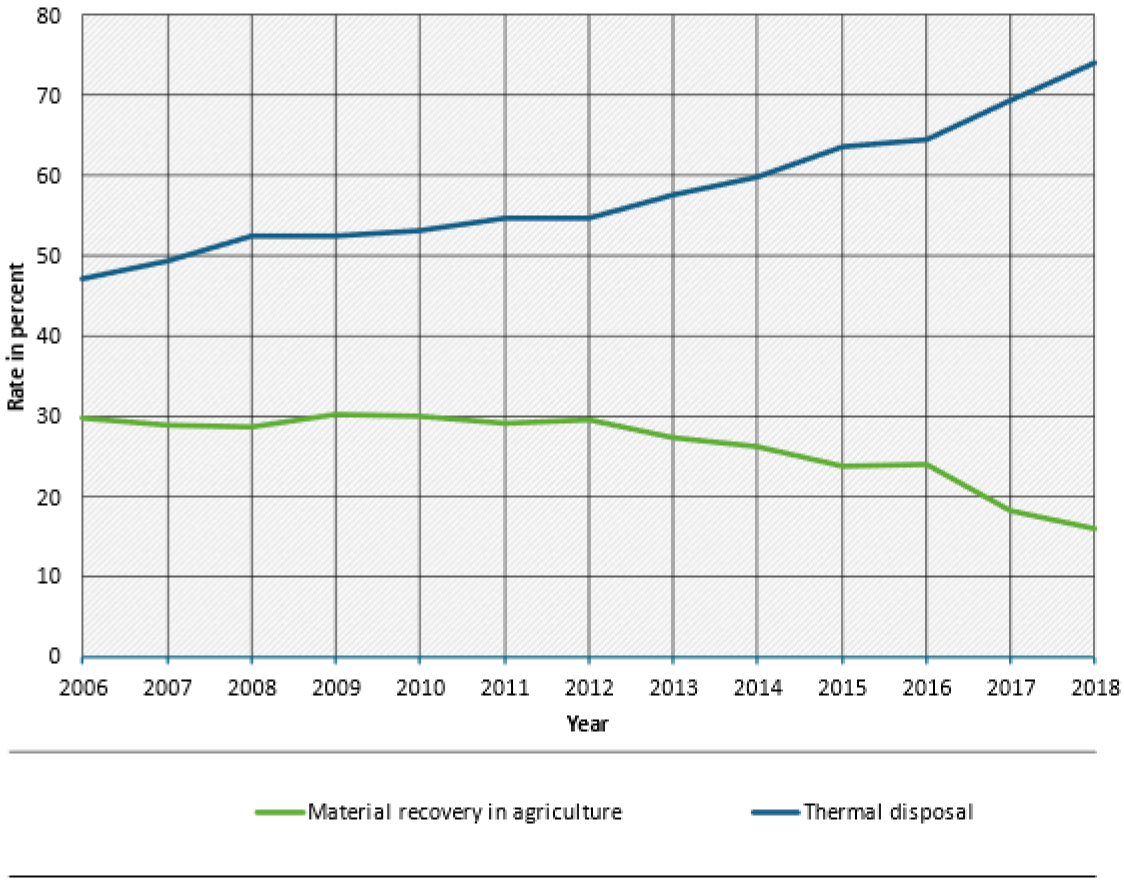
Figure 10: **Percentage recycling or disposal of municipal sewage sludge.**



Source: (Destatis, 2020a)

In addition to recycling in agriculture, sewage sludge is mainly disposed of thermally. Together, these two types of utilization and disposal accounted for 90% of the 1.7 million tons of sewage sludge generated in 2018. The trend of sewage sludge utilization in these two variants in 2006-2018 is shown in Figure 11.

Figure 11: Percentage recycling of municipal sewage sludge.



Source: (Destatis, 2020a)

Figure 11 shows that agricultural use of sewage sludge has declined sharply from 2006 to 2018, while thermal treatment has increased sharply over the same period. This may be due, in part, to the fact that increasingly strong monitoring of compliance (State Office of Natural Resources, 2015) is taking place and higher requirements are being imposed under legislation (Ministry of Environment Nature and Consumer Protection of the State of North Rhine-Westphalia, 2018). If necessary, this may also be a consequence of the "*PFAS scandals*", in which sewage sludge is also suspected to have played a role - since 2012, an ever greater decline in material recycling has been observable, which could also be a result of the higher pressure from farm manure.

3.1.4.2.2 Sewage sludge load

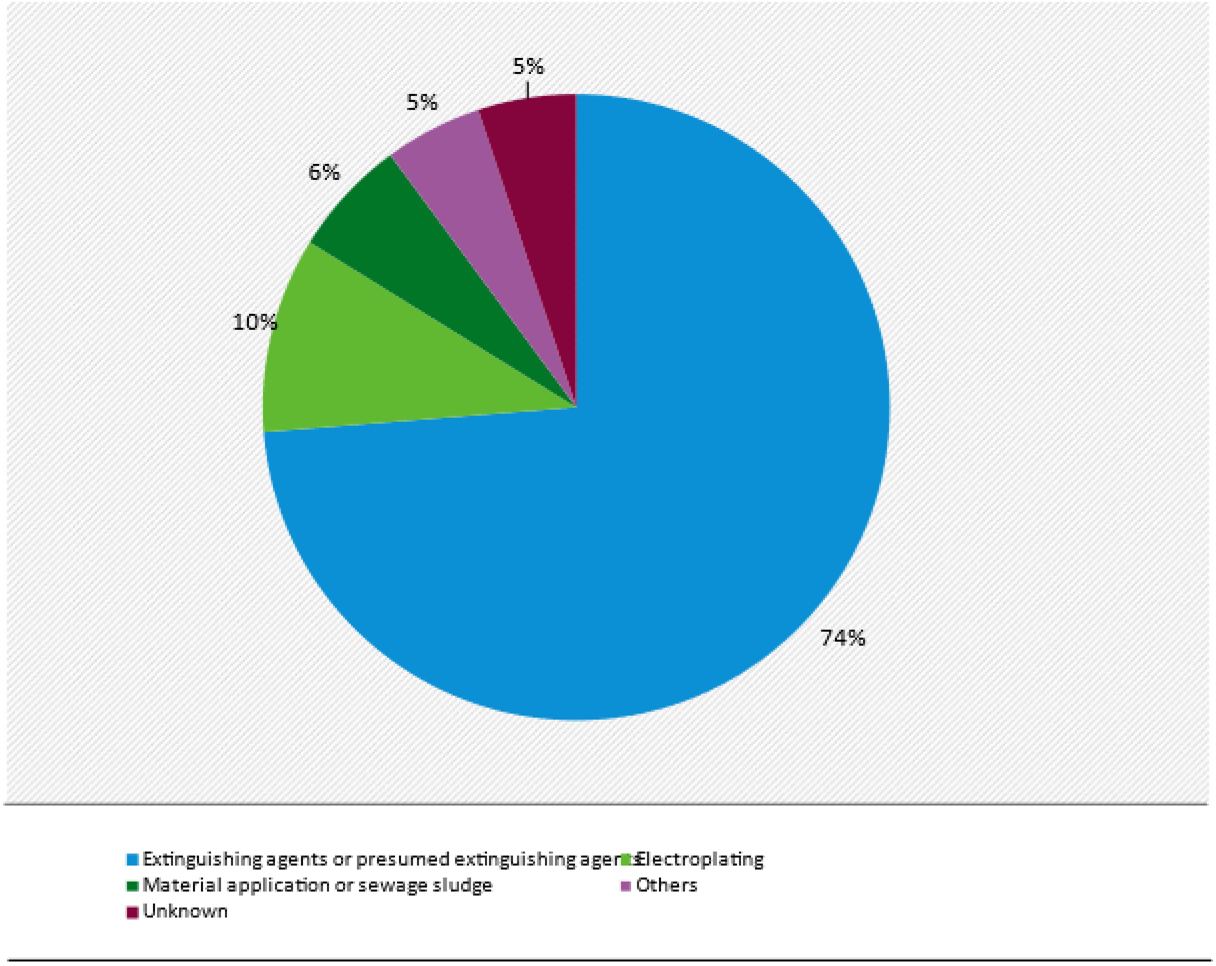
Due to cases of damage in relation to contaminated soils, some federal states have also increased the implementation of sampling campaigns and have increasingly investigated fluorinated compounds in sewage sludge. One federal state that has requested and evaluated a very large series of measurements (sewage sludge) from its wastewater treatment plants here is Bavaria (Ulrich et al., 2016). For this purpose, sewage sludges from 685 to 784 wastewater treatment plants per year were evaluated in the years 2008 to 2013. The results (sum of the concentration of 11 individual substances) were divided into ranges smaller than 10 µg/kg DM; 10-125 µg/kg DM; 125-500 µg/kg DM and more than 500 µg/kg DM. It can be seen that the proportion in the highest group decreased from eleven to one WWTP over the six years, while the lowest group increased from 229 to 451 WWTPs. This result can be explained mainly by the decreased concentration of PFOS, which was reduced from a mean concentration of 46-50 µg/kg DM to 12-19 µg/kg DM. A similar trend can be observed in North Rhine-Westphalia. Between 2006 and 2009, sewage sludge from 27 wastewater treatment plants was examined. Here a significant reduction of PFOS and PFOA can be shown in the given time period. During this period, a decrease in the values from over 6,000 µg/kg to below 2,000 µg/kg was observed. (Landesamt für Natur, 2011).

3.1.4.2.3 Soil

The reasons for PFAS contamination in soils are manifold. The main reasons for soil contamination are the application of contaminated materials, such as sewage sludge, or the use of substances containing PFAS for firefighting, such as fire-fighting foams. An event in which the application of contaminated sewage sludge polluting the soil can occur, for example, if sewage sludge has been used as fertilizer or in landscaping, which can lead to contamination of the soil as well as the groundwater.

NRW has been keeping statistics on the recording of PFAS damage cases in relation to soils since 2011. (State Office for Nature, 2019). These collected cases were further investigated by the state and an intensive search for further possible input pathways is currently underway. The percentage distribution of these damage cases correlates with possible causes and is shown in Figure 12.

Figure 12: PFAS damage cases in NRW.



Source: (State Office for Nature, 2019)

In the contamination of reported soils in NRW, firefighting operations with extinguishing agents containing PFAS or suspected extinguishing agents account for 73% and thus three quarters of all cases are attributed to them (State Office for Nature, 2019). Electroplating follows with 10% and material application or sewage sludge with 6%. This statistic clearly shows how frequent and ubiquitous the contamination of soils with PFS is and that a risk to drinking water can directly follow from this. In addition, there is the possible danger of direct contamination of food and animal feed grown on appropriately contaminated agricultural land.

3.1.5 Analytical methods

High-performance analytical methods are needed to detect fluorine compounds from the PFAS group. Against the background of a trend towards decreasing PFAS limits, the analytical methods must also become more and more sensitive in order to make these small amounts, which are usually in the ng/L range, detectable. This is the only way to monitor compliance with the limits specified by the regulations.

Due to the great diversity of PFAS, comprehensive analysis is difficult. Fluorine is monoisotopic (only the stable ^{19}F isotope exists), which makes it difficult to study by organic mass spectrometry, which usually uses the isotopic pattern of individual elements to identify compounds.

As it stands, the analytics can be broadly divided into three different parameters: i) total fluorine content; ii) sum parameters related to organically bound fluorine; and iii) target analytics. (Al Amin et al., 2020; Cousins et al., 2020; Jahnke & Berger, 2009; Miaz et al., 2020; Schultes et al., 2018). In this context, the most common procedure in current routine analysis is the acquisition of target analytes (Backe et al., 2013; Chiesa et al., 2018; He et al., 2015; Jamari et al., 2019; Navarro et al., 2011). In this regard, to determine the presence of fluorinated compounds in sewage sludge, compost and soils, DIN 38414-14 is frequently used (DIN 38414-14, 2011) or the AbfKlärV is used. This DIN describes a method that uses high-performance liquid chromatography (HPLC) and mass spectrometric detection (MS). This method includes 12 target analytes, which are divided into 10 acids of different carbon length. Since only a small spectrum of PFAS loading is mapped here, it can lead to a false assessment of loading (Spaan et al., 2020). This could be shown in the context of various studies, which found a sometimes very large difference between the concentration that could be identified by means of target analytes and the concentration that was determined with a sum parameter. For example, in an article published in 2011, Loi *et al.* were able to show that only between 0.01% and 80% of the organic fluorine found could be explained in a large number of the samples studied (Loi et al., 2011). This difference may have several causes. Possible causes could be the extended product range or intermediates or metabolites of PFAS, such as PFOS precursors that have been biotransformed (biologically converted). This complicates above all the (eco-)toxicological evaluation of a sample, since a large number of different species are present and not just the parent compound. The problem of analytical detection of individual compounds will be further exacerbated in the future, as a large number of production processes have been converted to comply with the new legislation (Koch et al., 2019).

For this reason, there is an increasing focus on sum parameter analysis in order to be able to map the contamination with organically bound fluorine as comprehensively as possible (Kotthoff & Bücking, 2018). Here, mainly the two parameters extractable organic fluorine (EOF) and adsorbable organic fluorine (AOF) have become established (Akhdhar et al., 2020; von Abercron et al., 2019; Willach et al., 2016; Yeung et al., 2013). In EOF, the organic compounds are usually extracted from the solid using nonpolar solvents, such as hexane, while the fluoride remains in the solid. This can still be combined with solid phase extraction (SPE), which separates any extracted fluoride from the organic compounds. In AOF, the organic compounds are adsorbed onto activated carbon and then burned and collected in an aqueous solution. Ion chromatography, usually used in combination with AOF, in the form of combustion ion chromatography (CIC) is then used to analyze the fluorine content. Alternatively, High Resolution-Continuum Source-Graphite Furnace Molecular Absorption Spectrometry (HR-CS-GFMAS) can be used, mostly in combination with EOF. Because these methods do not separate species and sum over all bound fluorine, they are suitable for identifying hotspots. HR-CS-GFMAS has limits of quantification in the lower µg/L range. (Gleisner et al., 2011; Krüger et al., 2012; Ozbek & Akman, 2013), whereas CIC operates in the mid µg/L range. (Miyake, Kato, et al., 2007; Miyake, Yamashita, Rostkowski, et al., 2007; Miyake, Yamashita, So, et al., 2007). Due to the upstream extraction, the methods are suitable for both solids and aqueous samples and thus for most common environmental matrices. In addition, enrichment can be used to further reduce the absolute detection limit, for example (Metzger et al., 2019). Gehrenkemper et al. (2021) were able to show here that comparable results are obtained when using the two methods AOF and EOF, and that here especially the limits of quantification and the scatter are different. To evaluate the (eco-)toxicological relevance of a finding in detail, an HPLC-MS/MS method can subsequently be used, which then determines, for example, the proportion of the 12 substances listed in DIN 38414 (DIN 38414-14, 2011) that can be quantified.

Another method for determining the PFAS loading situation in a sample is the TOP assay, in which precursor compounds are oxidized with the aid of hydroxyl radicals to form the persistent perfluorocarboxylic acids (see chapter 1.1, PFCA). These acids can then be measured, and the load of precursor compounds can be estimated from the difference in perfluoroalkyl acid concentration before and after the TOP assay. Thus, the PFAS load can be represented much more accurately than with "conventional" PFAS single compound analysis (Houtz & Sedlak, 2012). However, this technique is not yet widely used in routine analysis.

3.2 Analysis results

3.2.1 Textiles

The results of the EOF analysis (method: chapter 2.3 and 2.4) of the 13 textile samples are shown in Table 19.

Table 19: EOF content of the textile samples tested.

Sample (short name)	EOF content [µg/kg]	Standard deviation [µg/kg]	Sample (image)
WJ01	55	5	
WJ02	122	9	
WJ03	64	4	
WJ04	1163	100	

Sample (short name)	EOF content [µg/kg]	Standard deviation [µg/kg]	Sample (image)
WJ05	109	5	
WJ06	163	14	
WJ07	37	4	
WJ08	295	12	
WJ09	54	5	
WJ10	31	3	
TO1	<LOD	<LOD	
TO2	195	15	
TE1	83	8	

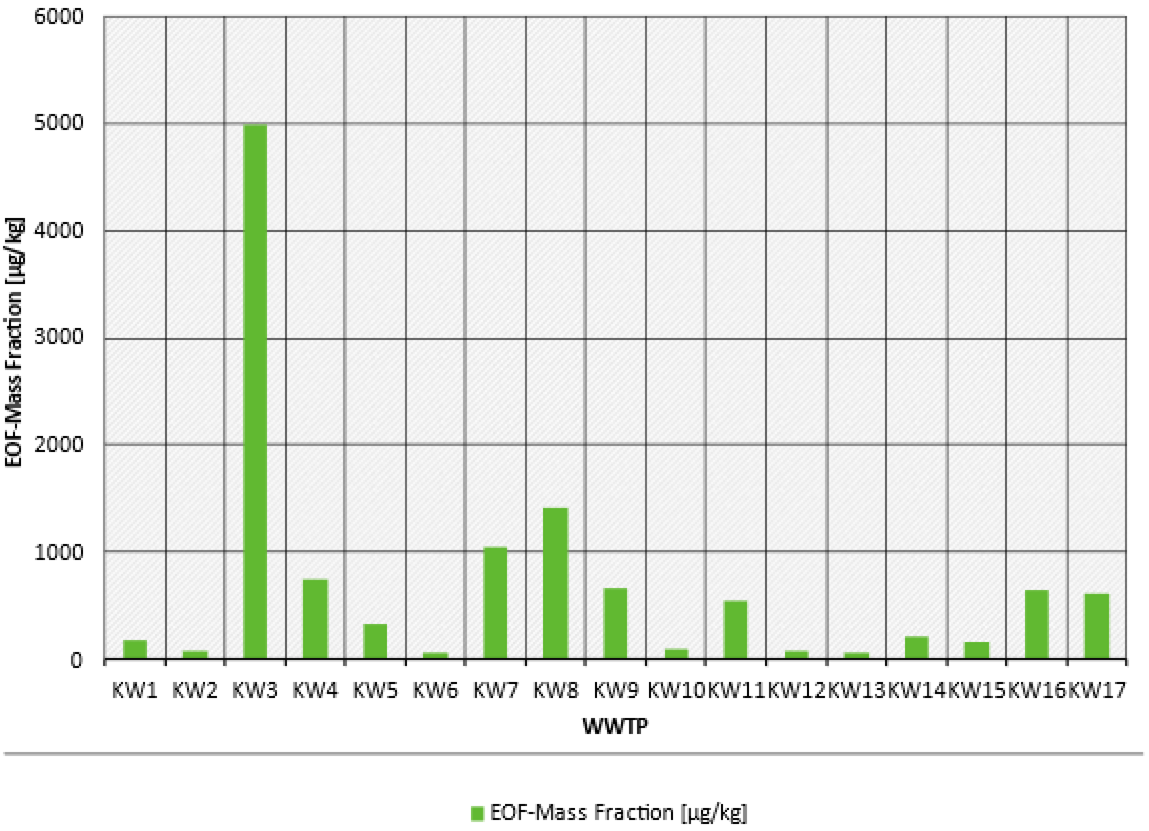
Source: Own representation

The EOF contents obtained from these analyses show that organically bound fluorine is detectable in almost all samples examined. Only in one sample (TO1, cf. Table 19), no organically bound fluorine could be detected with the method used. In the other samples examined, there was a wide variation in content (from approx. 50 µg/kg to approx. 1000 µg/kg) between the individual samples examined. One reason for this may be the different condition of the investigated samples. No further information is available on the lifetime of the product and its treatment, such as the frequency of washing or the number of re-impregnations. Accordingly, only the statement that organically bound fluorine is present in the waste stream can be made. However, with up to 1 mg/kg organically bound fluorine related to individual parts from this stream, it also shows that this load cannot be considered negligible.

3.2.2 Sewage sludge

The investigated sewage sludge originates from various municipal and industrial wastewater treatment plants. In the case of industrial wastewater treatment plants, special attention was paid to the upstream paper industry. Accordingly, sewage sludge from eight wastewater treatment plants (KW1-KW8), which are directly connected to the paper industry, and nine further sewage sludges (KW9-KW17) from municipal wastewater treatment plants, where elevated PFAS contents had already been detected in the past, were investigated. The results from the EOF measurements are shown in Figure 13.

Figure 13: EOF contents measured in the investigated sewage sludge samples - KW1-KW8 originate from wastewater treatment plants of the paper industry, samples KW9-KW17 originate from municipal wastewater treatment plants.



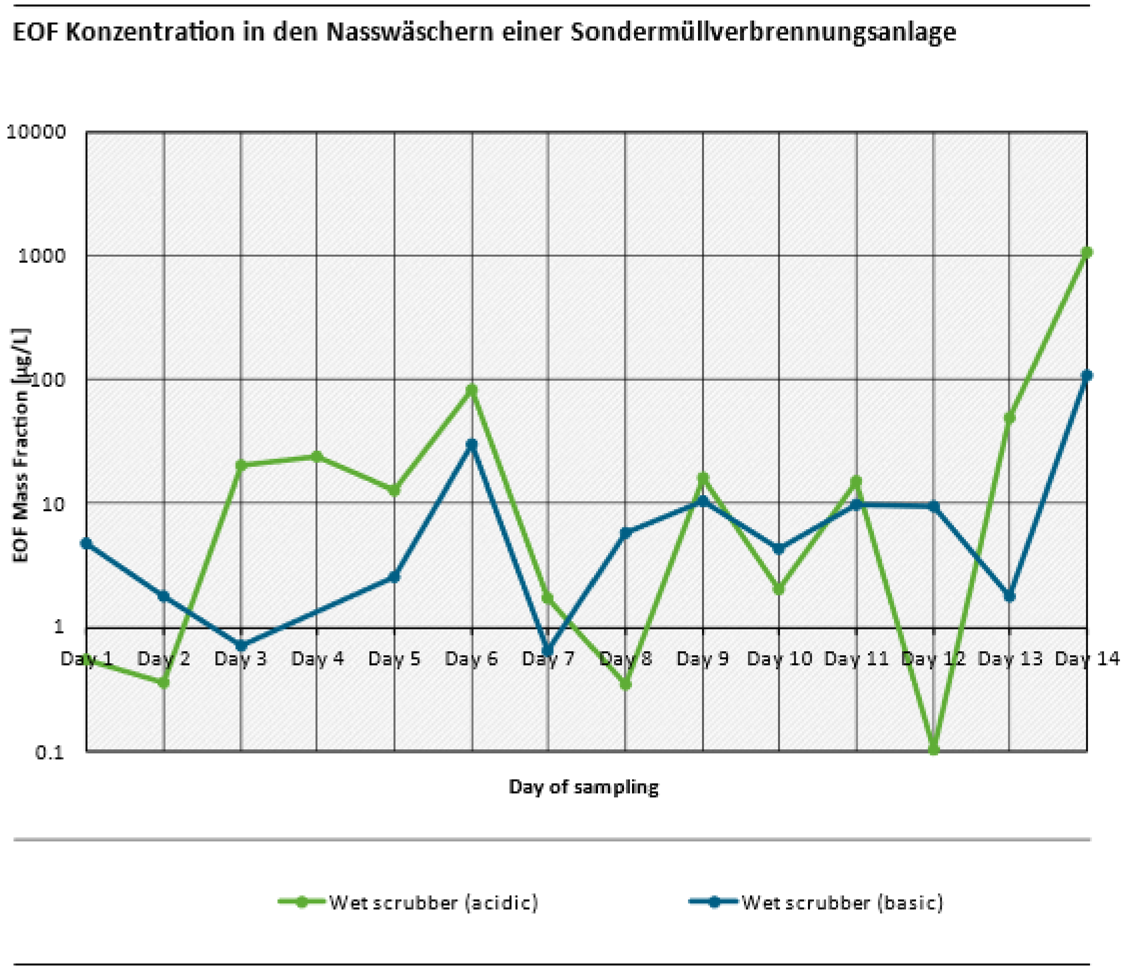
Source: Own representation

In previous studies on PFAS in sewage sludges, mainly target substances were investigated (above all PFOS and PFOA), whose mass fraction has decreased considerably over the years. The investigations carried out here, which are aimed at an overall consideration with regard to fluorinated compounds, clearly show that in some cases a significantly increased EOF content can be detected in the investigated sewage sludges. In particular, the investigated sewage sludge from KW3 (industrial wastewater treatment plant) shows an EOF load in the range of 5000 µg/kg. But also, in the municipal sewage treatment plants a high EOF load (up to approx. 0.75 mg/kg) can be determined. This clearly shows that the detection of individual target substances can lead to an underestimation of the load and thus a misjudgement for a hazard assessment is possible.

3.2.1 Washing solutions of a thermal treatment of hazardous waste

The investigated aqueous solutions originate from the scrubbers of a thermal hazardous waste treatment plant. A total of 14 samples were taken from two scrubbers (basic/acidic) over a period of one month. The EOF concentration profile obtained for the period of one month is shown in Figure 14.

Figure 14: Measured EOF concentrations in scrubbers of a thermal hazardous waste treatment plant - samples were taken over a period of one month.



Source: Own representation

Using the data from Figure 14 the remaining organically bound fluorine content after thermal waste treatment can be followed over a period of 14 days in the corresponding scrubbers. Since a large number of unknown substances can be formed after thermal treatment, the organically bound fluorine content was investigated here. The data clearly show that there was no complete turnover of fluorinated compounds during the incineration process and that they can be detected in the scrubbers. However, there is no clear trend between the two scrubbers (acidic/basic). There appears to be a strong dependence from the day-dependent recycled material to the organically bound fluorine found. This gives an indication that the amount of PFAS introduced as well as its type has an influence on the concentration found. Thus, on some days a higher concentration can be found in the acid scrubber and on others in basic scrubbers. However, it is also evident here that further investigations must be carried out. Especially in the last measurement, concentrations in the range between 100 µg/L and 1 mg/L can be found. Here the advantage of the EOF method as a fast-screening tool becomes apparent. The data obtained in this way must be backed up accordingly with target measurements in order to be able to make an estimate here of the risk emanating from this waste stream. This is also supported by publications such as (Longendyke et al., 2022) which underline that there is still a need for research to understand the processes that occur during thermal waste utilization and to use appropriate processes here.

3.2.2 Paper

Two fractions (graphic paper fraction/packaging paper fraction) from a wastepaper sorting plant were used to investigate the EOF load of wastepaper. The results of the EOF investigation are shown in Table 20.

Table 20: EOF content of the paper samples tested.

Sample (short name)	EOF content [µg/kg]	Standard deviation [µg/kg]	Sample (image)
Paper 1	132	9	
Paper 2	118	5	
Packaging papers 1	239	27	

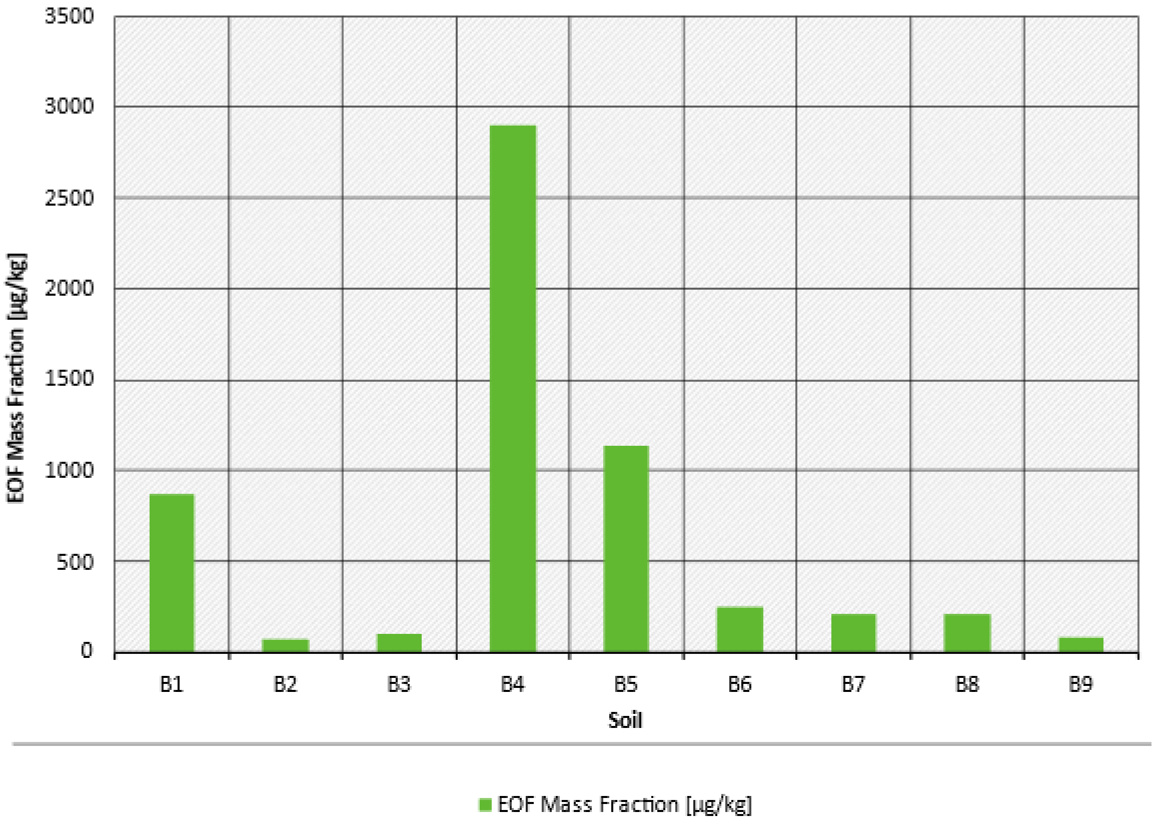
Source: Own representation

The EOF measurements for paper show that there does not appear to be a significant difference between the two samples taken for the day selected. It is generally seen that the two randomly taken samples have approximately the same EOF content, indicating an average EOF load for the day of investigation. The packaging paper studied, has a higher EOF value than the paper fraction. These data need to be further investigated for an evaluation in terms of (eco)toxicology by means of target methods. But it also shows the importance that sum parameters have: The EOF screening of the samples allows to identify increased sub-quantities and thus to preselect only a part for the significantly more expensive target methods while keeping costs at a minimum.

3.2.3 Soils

The soils investigated in this study come from different federal states and are classified in the following categories: Four soils are from an area of known contamination (B1, B3, B4, and B5). Two soils are without known prior contamination (B2 and B7). Two soils were taken from landfills (B6 and B8). One soil is from a soil wash (B9). The data from the EOF measurements are shown in Figure 15.

Figure 15: Measured EOF contents in the investigated soils.



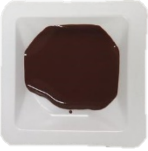
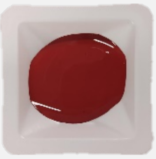
Source: Own representation

The pre-polluted soils show an EOF load of up to 3 mg/kg. The remaining soils all show an EOF load (< 0.25 mg/kg), but this does not differ that much from their sampling location. This indicates that there is a measurable background level of fluorinated organic compounds, and that further consideration is needed here - especially in light of the disposal and quantities of the soil under consideration playing a role in this.

3.2.4 Hardware store products

The measured EOF contents of the eight DIY products investigated are shown in Table 21 summarized.

Table 21: EOF content of the investigated DIY products.

Sample (short name)	EOF content [µg/kg]	Standard deviation [µg/kg]	Sample (image)
Roof/Facade Waterproofing	<LOD	<LOD	
Facade paint (White)	173	14	
Facade protection	107	17	
Sealing compound	<LOD	<LOD	
Hybrid glaze (mahogany)	51	3	
Universal sealant	<LOD	<LOD	
Deep primer	81	7	
Solid color (red)	60	48	

Source: Own representation

The data obtained suggest that the use of additional fluorinated compounds is dispensed with for sealing materials. In the case of the other outdoor DIY products investigated, which are offered in liquid form, a fairly constant EOF value of approx. 100 µg/L can be observed. In view of the quantities used, this result suggests that aging and the release to be expected with it must be investigated further. In particular, the release into the soil and the possible increased contamination of the groundwater must be mentioned here.

3.3 PFAS concentrations in selected waste fractions from further literature research

Since no measured values for individual PFAS could be obtained from the samples measured in this project, a further literature search was conducted on PFAS measurement data in four waste streams. The selected waste streams were paper, textiles, soils and sewage sludge. For all waste streams PFAS literature data with focus on German samples were searched, extracted and mean and median values were calculated from the obtained data. (for methodology see chapter 2.5 and 2.6). The results are presented in this chapter. However, some general limitations should be considered when looking at the data.

The major limitation arises from the varying number of PFAS analyzed. Each study places a different focus, with most analyzing C4-C10 carboxylic and sulfonic acids. The greatest differences are in the precursors. Some studies look only at PFCAs and PFSAs and measure no precursors, while other studies have measured up to 40 individual precursors. In general, when more compounds are measured, more positive hits are obtained, resulting in higher concentrations measured, compared to studies measuring only a few compounds. This may give the impression that PFAS concentrations are lower in studies that measured fewer PFAS. This is especially noticeable in the median values, as these often correspond to the value "0" for the precursors, because they were not measured in many studies.

Furthermore, it should be noted that, except for the soil samples, no or very few background measurements were taken or identified. In most cases, specific suspect cases are analyzed, resulting in positive hits for most of the measurements. At the same time, there are PFAS-free products and wastes, but these are mostly not sampled and measured. Consequently, it can be assumed that the identified values are higher than the real occurring average values.

Finally, it should be mentioned that for the calculation of the ∑PFAS median values, the median of the ∑PFAS values of each measurement has been formed instead of forming a sum of the median values of the individual PFAS groups (PFAS, PFCA, precursors, PFPA). This results in deviating values, which can be recognized, for example, in the PFAS concentrations in the textiles (cf. Table 25).

The following PFAS in Table 22 were measured at least once in the studies analyzed here:

Table 22: PFAS compounds found in the literature search.

PFAS	Name (CAS)
PFCAs	
TFAA	Trifluoroacetic acid (76-05-1)
PFPrA	Perfluoropropanoic acid (422-64-0)
PFBA	Perfluorobutanoic acid (375-22-4)
PFPeA	Perfluoropentanoic acid (2706-90-3)
PFHxA	Perfluorohexanoic acid (307-24-4)
PFHpA	Perfluoroheptanoic acid (375-85-9)
PFOA	Perfluorooctanoic acid (335-67-1)
PFNA	Perfluorononanoic acid (375-95-1)

PFAS	Name (CAS)
PFDA	Perfluorodecanoic acid (335-76-2)
PFUnDA	Perfluoroundecanoic acid (4234-23-5)
PFDoDA	Perfluorododecanoic acid (307-55-1)
PFTrDA	Perfluorotridecanoic acid (72629-94-8)
PFTeDA	Perfluorotetradecanoic acid (376-06-7)
PFHxDA	Perfluorohexane decanoic acid (67905-19-5)
PFODA	Perfluorooctanedecanoic acid (16517-11-6)
PFSAs	
PFBS	Perfluorobutanesulfonic acid (59933-66-3)
PFPS	Perfluoropentanesulfonic acid (2706-91-4)
PFHxS	Perfluorohexane sulfonic acid (355-46-4)
PFHpS	Perfluoroheptane sulfonic acid (375-92-8)
PFOS	Perfluorooctane sulfonic acid (1763-23-1)
PFNS	Perfluoronon sulfonic acid (68259-12-1)
PFDS	Perfluorodecane sulfonic acid (335-77-3)
PFUnDS	Perfluorundecane sulfonic acid (749786-16-1)
PFDoDS	Perfluorododecane sulfonic acid (79780-39-5)
PFTrDS	Perfluorotridecansulfonic acid (791563-89-8)
Precursors	
4:2 FTS	4:2 Fluorotelomer sulfonic acid (757124-72-4)
6:2 FTS	6:2 Fluorotelomer sulfonic acid (27619-97-2)
8:2 FTS	8:2 Fluorotelomer sulfonic acid (39108-34-4)
6:2 PAP	Mono[2-(perfluorohexyl)ethyl] phosphate (57678-01-1)
8:2 PAP	Perfluordecylphosphate (57678-03-2)
6:2 diPAP	6:2 Fluorotelomer phosphate diester (57677-95-9)
8:2 diPAP	8:2 Fluorotelomer phosphate diester (678-41-1)
6:2/8:2 diPAP	6:2/8:2 fluorotelomer phosphate diester (943913-15-3)
8:2/10:2 diPAP	8:2/10:2 fluorotelomer phosphate diester (1158182-60-5)
10:2 diPAP	10:2 Fluorotelomer phosphate diester (1895-26-7)
diSAmPAP	di-substituted polyfluoroalkyl phosphate esters of N-ethyl perfluorooctane sulfonamidoethanol (/)

PFAS	Name (CAS)
SAmPAP	Perfluorooctane sulfonamidoethanol-based phosphate esters (/)
4:2 FTOH	2-(Perfluorobutyl)ethyl alcohol, 4:2 fluorotelomer alcohol (2043-47-2)
6:2 FTOH	2-(Perfluorohexyl)ethyl alcohol, 6:2 fluorotelomer alcohol (647-42-7)
8:2 FTOH	2-(Perfluorooctyl)ethyl alcohol, 8:2 fluorotelomer alcohol (678-39-7)
10:2 FTOH	2-(Perfluorodecyl)ethyl alcohol, 10:2 fluorotelomer alcohol (865-86-1)
5:3 FTCA	4,4,5,5,6,6,7,7,8,8,8-Undecafluorooctanoic acid (914637-49-3)
7:3 FTCA	2H,2H,3H,3H-Perfluorodecanoic acid (812-70-4)
ADONA	Ammonium-2,2,3-trifluoro-3-(1,1,2,2,3,3-hexafluoro-3-(trifluoromethoxy)propoxy)propanoate (958445-44-8)
H-DONA	Ammonium-2,2,3-trifluoro-3-(1,1,2,2,3,3-hexafluoro-3-(trifluoromethoxy)propoxy)propanoate (958445-44-8)
PFOSA	Perfluorooctane sulfonamide (754-91-6)
FOSAA	Perfluorooctanesulfonamidoacetic acid (2806-24-8)
EtFOSAA	N-Ethylperfluorooctanesulfonamidoacetic acid (2991-50-6)
N-MeFOSA	Heptadecafluoro-N-methyloctanesulfonamide (31506-32-8).
N-EtFOSA	Sulfluramide (4151-50-2)
N-MeFOSAA	N-methylperfluoro-1-octanesulfonamidoacetic acid (2355-31-9)
N-EtFOSAA	N-Ethylperfluorooctanesulfonamidoacetic acid (2991-50-6)
PFECHS	Perfluoro-p-ethylcyclohexylsulfonic acid (646-83-3)
7H-PFHpA	7H-Perfluoroheptanoic acid (1546-95-8)
9Cl-PF3ONS	9-Chlorohexadecafluoro-3-oxanonane-1-sulfonic acid (756426-58-1)
11Cl-PF3OUdS	11-Chloroeicosafluor-3-oxaundecan-1-sulfonic acid (763051-92-9)
HFPO-DA	Undecafluoro-2-methyl-3-oxahexanoic acid (13252-13-6)
HPFHpA	7H-Dodecafluoroheptanoic acid (1546-95-8)

PFAS	Name (CAS)
H2PFDA	2H,2H-Perfluorodecanoic acid (27854-31-5)
H4PFUnDA	2H,2H,3H,3H-Perfluoroundecanoic acid (34598-33-9)
PFPA	
Cl-PFHxPA	6-Chloroperfluorohexylphosphoric acid (-)

3.3.1 Textiles

As described in chapter 3.1.3.2 PFAS are frequently used as impregnating agents on textiles. This primarily affects outdoor textiles such as jackets, coats and gloves, as well as carpets and vehicle seats. PFAS are also used in the treatment of leather. For the following products in Table 23 PFAS measured values could be found from five studies. (Gremmel et al., 2016; Janousek, Lebertz, et al., 2019; Müller & Schlummer, 2011; Norwegian Environment Agency, 2021):

Table 23: Number of textile samples found.

Sample	Number of individual samples
Carpets	17
Awnings and protective films	14
Gloves	4
Leather	14
Vehicle seats (e.g. cars, boats, buses)	8
Outdoor materials (e.g. jackets, coats, pants)	21
Work clothes	1
Textiles in maritime applications	5
Total	**84**

Particularly high PFAS concentrations were found in awnings (max. 4407 µg/kg ∑PFAS) (Janousek, Lebertz, et al., 2019) and in outdoor jackets (max. 4687 µg/kg ∑PFAS) were found, which originate from the use of fluorotelomer alcohols (Gremmel et al., 2016). No PFAS were detected in some samples (9).

For textile measurements, the unit µg/m^2 is often used, which meant that these values had to be converted into µg/kg. For this purpose, typical weights in kg/m^2 of the materials concerned (carpets, vehicle seats, leather, awnings, gloves and outdoor jackets) were researched by means of Internet research and then averaged. The averaged values in Table 24 were then used to convert PFAS concentrations from µg/m^2 to µg/kg.

Table 24: Conversion factors for textile samples which are given in the unit µg/m² to convert them into µg/kg.

Textile type	Conversion factor [kg/m²]
Carpets	2.302
Awnings and protective films	0.322
Gloves	0.762
Leather	0.554
Vehicle seats (e.g. cars, boats, buses)	0.352
Outdoor materials (e.g. jackets, coats, pants)	0.238
Work clothes	-
Textiles in maritime applications	-

Mean and median values were then calculated from all PFAS values, which are shown in the following Table 25 is shown below:

Table 25: PFAS mean and median values in the textile samples found. All data in µg/kg.

Substance	Mean value	Median
∑PFCA	87.56	8.36
∑PFSA	20.65	0.00
∑PFPA	0.00	0.00
∑Precursors	280.70	3.46
∑PFAS	**388.91**	**74.05**

It can be seen that the median value is significantly lower than the mean value, which is due to the fact that the different studies place different emphases. This can be seen in particular in the case of precursors, as some studies do not measure these at all, which means that a value of 0 is assumed here and the median shifts accordingly.

The PFAS values averaged here for the textiles (Table 25) agree well with the textiles measured in this study (see chapter 3.2.1). The EOF levels of the measured textiles are in the range of <LOD-1163 µg/kg, with most values in the high double-digit to low triple-digit µg/kg range.

In a recent report by. Wood, (2020) concentrations of 0.025-0.05% are reported for leather (250-500 mg/kg), 0.03% for synthetic carpets (300 mg/kg), and 2-3% for textiles and furniture (20,000-30,000 mg/kg). These concentrations are above the concentrations calculated here in Table 25which may be due to the fact that the concentrations of Wood (2020) are based on the average PFOS concentrations in textiles from UNEP (2017) are based.

Similar concentrations are also given in the overall PFAS restriction process. These vary between <0.1% and 7% depending on the application. Some applications consist of 100% PFAS such as PTFE membranes (Annex XV, 2023a). In general, the concentrations here are also higher than the calculated values in Table 25which may be due to the fact that the restriction procedure covers all PFAS (including polymers) and not only the short-chain acids and some precursors as in this project.

3.3.2 Sewage sludge

Wastewater is treated in wastewater treatment plants; the treated wastewater is discharged into surface waters. A basic distinction is made between municipal wastewater treatment plants, which treat wastewater from households and businesses, and industrial wastewater treatment plants. The latter usually treat only their own industrial wastewater. Some municipal wastewater treatment plants receive both municipal and industrial wastewater (Becker et al., 2008). A few industrial wastewater treatment plants also treat municipal wastewater. For industrial WWTPs, a distinction can be made between direct and indirect dischargers. Most wastewater treatment plants are direct dischargers, as they discharge treated wastewater directly into surface waters. Indirect dischargers are, for example, companies that discharge their wastewater untreated or pretreated into the public sewer system and thus into the municipal wastewater treatment plant. Their wastewater is therefore discharged "indirectly" into water bodies.

Major PFAS point sources include wastewater from industries that manufacture or use PFAS in their processes, such as the paper, textile, metal coating, and semiconductor industries (Mueller & Yingling, 2017). However, because PFAS are also used in a variety of consumer products and household applications, they are found in many municipal wastewater treatment plants, for example, through leaching from textiles.

Since primary and secondary wastewater treatment processes (biological and mechanical treatment) are not efficient enough to remove PFAS from wastewater, most of them accumulate in sewage sludge, especially the long-chain PFAS (W. Zhang et al., 2022). Analyses have shown that this is especially the case for long-chain PFAS, which tend to accumulate in sewage sludge due to the high intermolecular forces between PFAS molecules and solid particles. A study by Stahl et al. 2018 illustrates this problem. In their study, long-chain PFAS accounted for 85.9% and short-chain PFAS accounted for 14.1% of the total concentration in sewage sludge, indicating that sewage sludge can serve as a sink for long-chain PFAS (Stahl et al., 2018). When applied to land, it can then pose a risk to terrestrial but also aquatic environments. Since not all PFAS accumulate in the sewage sludge phase, they can also still be found in the effluent of wastewater treatment plants (Campo et al., 2014; Coggan et al., 2019; Fuertes et al., 2017).

In order to obtain an overview of PFAS concentrations in German sewage sludge, a large number of sewage sludge samples from the literature were analyzed (see also chapter 2.5).

A total of six publications were found that analyzed PFAS in German sewage sludges (Becker et al., 2008; Frömel et al., 2016; Sachsen-Anhalt, 2008; Stahl et al., 2018; Toshovski et al., 2020; Ulrich et al., 2016), including scientific articles and reports from local government agencies. These studies examined a variety of different WWTPs, including municipal WWTPs as well as WWTPs receiving municipal wastewater and wastewater from local businesses. Purely industrial WWTPs were excluded from this review because they usually cannot recycle their sludge under the Sewage Sludge Ordinance. Some are recycled according to the Biowaste Ordinance, but the majority is sent to thermal waste treatment. Generally, such plants have higher pollutant concentrations (including PFAS) than municipal plants, especially if they can be assigned to industries that use PFAS (e.g., paper or textile industry) (Frömel et al., 2016) (see also chapter 3.2.2).

A total of 71 individual samples were extracted. Based on the data obtained, the following mean and median values were calculated in Table 26.

Table 26: PFAS mean and median values in the sewage sludge samples found. All data in µg/kg.

Substance	Mean value	Median
∑PFCA	18.03	16.60
∑PFSA	48.80	31.30
∑PFPA	0.00	0.00
∑Precursors	0.36	0.00
∑PFAS	**67.18**	**47.40**

The highest PFAS value measured (417.8 µg/kg) came from the sewage sludge of a municipal wastewater treatment plant in Saxony-Anhalt (Saxony-Anhalt, 2008). Since in most studies no precursor substances were measured (value of "0") or only one substance was identified per sample, the median value is significantly lower than the mean value. If measured values from industrial sewage sludge were included, it can be assumed that PFAS concentrations would increase. The values shown here, in Table 26, therefore probably underestimate the real values.

A long-term study (5 years from 2008-2013) showed that PFAS concentrations in sewage sludge decreased overall for both regulated and non-regulated substances (Ulrich et al., 2016). However, it remains an open question whether the reason could be that industry is replacing long-chain PFAS with short-chain PFAS, which are less prone to pass into the sludge phase. The results of this study should be confirmed by further measurements.

The data shown in Table 26 PFAS mean and median values calculated can be compared with the EOF values measured in this study in chapter 3.2.2. While EOF values up to 5,000 µg/kg could be detected in industrial WWTPs, the PFAS concentrations from the municipal WWTPs (CW 9-17) are mostly in a range below 500 µg/kg (mean 338 µg/kg). The calculated values from Table 26 are therefore slightly below these values.

As part of the total PFAS restriction process, mass flows and concentrations for PFAS in waste have been published. There, the following PFAS concentrations (Table 27) in municipal and industrial sewage sludges are given. (Annex XV, 2023b):

Table 27: PFAS concentrations in sewage sludge in the PFAS restriction dossier. (Annex XV, 2023b). All values in µg/kg.

Substance	Mean value	Median
PFCA	51	2
PFSA	16	2
PFPA	0.00	0.00
Precursors	47	35
∑PFAS	**114**	**39**

It can be seen that the values in Table 27 and Table 26 are very similar, which probably suggests that these values reflect reality relatively reliably.

3.3.3 Paper

As described in chapter 3.1.3.2 PFAS are used in papers, especially food contact papers (including food packaging), to impart water- and grease-repellent properties to the paper. Since papers are largely recycled in Germany (see chapter 3.4.3), these substances can also be found in other papers, such as paper/cardboard packaging that is not intended for food contact (Müller & Schlummer, 2011). This could also be confirmed by the analyses in this project, as EOF values in the range of more than 100 µg/kg could be detected (see chapter 3.2.2).

In general, the term food contact papers is very broad. This includes, for example, burger papers, paper bags at the bakery, baking paper, paper plates, paper straws and pizza boxes, but also paper and cardboard packaging of, for example, frozen goods. Depending on the application, more or less PFAS are used. For example, fewer PFAS are usually used in packaging than in paper bowls, as these require special water- and grease-repellent properties.

Such papers (and other food contact materials) are regulated via Regulation EC No. 1935/2004 "on materials and articles intended to come into contact with food". This regulation is valid for "materials and articles [...] which, as a finished product

a) are intended to come into contact with food, or

b) are already in contact with food and are intended for that purpose, or

c) reasonably foreseeable to come into contact with food or release their constituents into food under normal or foreseeable use."

This definition thus covers a wider range of possible contact papers, from direct contact papers such as burger papers to paper and cardboard packaging. Under c), equipment such as coffee machines are also included, as they come into contact with food during use.

In total, three relevant studies with 47 individual paper samples could be identified (Kotthoff et al., 2015; Müller & Schlummer, 2011; Strakova et al., 2021). All samples but one were food contact paper. The one sample is a paper packaging, which was not used for food. All identified samples are listed Table 28:

Table 28: Number and type of paper samples found

Sample	Number of individual samples
Baking paper/sandwich paper/burger paper	21
Paper baking pan	14
Paper and cardboard packaging with food contact	6
Paper and cardboard packaging without food contact	1
Soup bowls from sugar cane	2
Doughnut bag / bakery bag	2
Pizza box	1
Total	**47**

This results in the following mean and median values in Table 29:

Table 29: PFAS mean and median values in the paper samples found. All data in µg/kg.

Substance	Mean value	Median
∑PFCA	155.87	0.60
∑PFSA	1.48	0.70
∑PFPA	0.00	0.00
∑Precursors	116.47	0.00
∑PFAS	**273.81**	**10.40**

The highest PFAS value was measured in a paper bowl for soups (3437 µg/kg) (Strakova et al., 2021), which, similar to textiles, originates from the use of fluorotelomer alcohols. This is to be expected as such bowls require particularly high water and fat resistance in contrast to, for example, cardboard packaging for frozen goods. For these values, too, the median is significantly lower than the mean, since some studies did not measure any (or only a few) precursors and often only one substance per sample could be identified.

The calculated values in Table 29 PFAS can be compared with the EOF values of the paper measurements in chapter 3.2.2. It should be mentioned that the comparability of the measurement data is not direct, since most of the papers considered in this chapter are food contact papers, while the EOF measurements were made on wastepapers from the wastepaper collection. Nevertheless, the values are very similar. The calculated PFAS concentrations from Table 29 are ~274 µg/kg, while the EOF values from the wastepaper and waste cardboard measurements range from 118-239 µg/kg (see Table 20).

Under the total PFAS restriction procedure, fluorine concentrations in food contact papers of 537 mg/kg (mean) and 1,200 mg/kg (maximum) are reported. These values refer only to fluorine, but PFAS molecules also consist of other atoms such as carbon and oxygen. The preparers of the dossier have therefore assumed that fluorine accounts for about 50% of the total weight of PFAS molecules (cf. PFHxA ~66% fluorine content and side-chain fluorinated PFAS ~12.8% fluorine content) (Annex XV, 2023a). This results in PFAS concentrations of 1,074 mg/kg (mean) and 2,400 mg/kg (maximum) in food contact papers. These values are significantly higher than those given in Table 29 which may be due to the fact that all PFAS are considered in the restriction dossier (acids, polymers, side-chain fluorinated polymers, etc.), whereas only the short-chain acids and some precursors are considered here.

3.3.4 Soils

In soils, PFAS are not intentionally used in most cases, but they also find application in e.g. plant protection products (European Commission, 2020). In most cases, however, PFAS only occur as contaminants in soils. For example, PFAS can enter the soil through the application of PFAS-contaminated sewage sludge or paper waste. This most likely happened in the Rastatt district by 2008, leading to serious consequences for humans, the environment, and the water supply (Rastatt, 2022a). As a consequence, drinking water has to be treated in this region. A similar case can be found in Gendorf. Here, the manufacture of PFOA and fluoropolymers and the associated emissions into the air, among other things, and subsequent deposition on the surrounding soils have polluted the environment with PFAS (Bavarian State Office for the Environment, 2020).

Furthermore, PFAS can be emitted during their production or during the production of PFAS-containing products, and they have also been found in leachate from landfills (Busch et al., 2010). In nature, PFAS can then enter soils through dry deposition and precipitation, for example. There, PFAS are persistent and thus accumulate, but they can also leach into groundwater. From the soil, PFAS can then be taken up by plants and ultimately by animals and humans.

The soil data set includes by far the most samples in this project with ~8,000 individual measurements. Data were taken from four scientific publications (Felizeter et al., 2020; Göckener et al., 2021; Janda et al., 2019; Kotthoff et al., 2020), two state reports (Bayerisches Landesamt für Umwelt, 2011; LUBW, 2016) and the environmental sample bank extracted (Environmental Sample Bank, 2022). The samples of the environmental sample bank are always taken at the same sites and cover urban sites as well as arable soils and forests. The country reports focus on soils from the respective federal state. The environmental sample bank comprises 27 individual samples and the data of 109 samples could be extracted from the reports and publications. Urban, forest and arable soils as well as compost soils are considered.

The data from the Rastatt district office includes the most data with ~8,000 individual measurements (Rastatt, 2022b). This included ~3,000 solid measurements and ~5,000 eluate measurements. Most samples were taken in suspected cases, but some background measurements were also taken. Many of the suspect cases were found to be uncontaminated with PFAS, placing these measurements below the detection limit. The LOD for these measurements was 1 µg/kg.

The eluate measurements of the samples from Rastatt are 2:1 shake eluates. This means that two liters of water are added to one kilogram of soil and the mixture is then shaken. During this process, the PFAS dissolve from the solid and pass into the liquid phase. The tendency to go into solution is higher the shorter the molecule and the more charges it has. Since the measurements of the eluates are in µg/L, a conversion factor of 2L/kg was applied to convert all measured values to µg/kg. It is thereby assumed for this project that all PFAS pass into the liquid phase. In reality, especially the shorter-chain PFAS can be washed out, whereas the longer-chain PFAS remain attached to the surfaces of particles. Therefore, the approach chosen here is associated with a certain uncertainty.

It should be mentioned that PFAS mostly accumulate in the upper layers of the soils, as they bind more strongly to solids with increasing chain length. Generally, the deeper the soil samples are taken, the lower the PFAS concentrations are as well, with longer-chain PFAS (≥C8) more likely to be found in the upper part and shorter-chain PFAS (≤C7) more likely to be found in the deeper part of the soils (e.g., due to diffuse inputs) (Bavarian State Office for the Environment, 2011; Brusseau et al., 2020). In older damage cases, PFAS concentrations may also be higher in the deeper soil layers than in the upper ones, as PFAS are leached over time by rain into deeper soil levels and eventually into groundwater. In specific emission cases (e.g., from firefighting foam), the PFAS profile may also be entirely different, with only specific PFAS. Since no specific waste data depending on soil depth could be identified in the further project, the depth is not considered further and all PFAS samples are given the same weighting.

Due to the large amount of data, the data from Rastatt has the greatest weighting and therefore does not necessarily reflect the situation in other regions of Germany. A total of three different mean and median values were calculated. The first calculation in Table 30 includes all values from the reports, the scientific studies and the environmental sample bank, as well as the solid samples from the district of Rastatt. The second calculation in Table 31 again includes all values from the publications and the environmental sample bank, as well as the eluate samples from the district of Rastatt. The last calculation in Table 32 includes all values.

Table 30: PFAS mean and median values in the identified soil samples. Eluate samples from Rastatt were not included. All data in µg/kg.

Substance	Mean value	Median
∑PFCA	25.80	0.00
∑PFSA	8.19	0.00
∑PFPA	0.00*	0.00
∑Precursors	5.72	0.00
∑PFAS	**39.71**	**0.00**

* The concentration for PFPA is not zero here, but statistically irrelevant.

In many of the suspected cases, PFAS concentrations (Table 30) were below the LOD, resulting in the median also having a value of zero.

Table 31: PFAS mean and median values in the identified soil samples. Solid samples from Rastatt were not included. All data in µg/kg.

Substance	Mean value	Median
∑PFCA	3.71	0.20
∑PFSA	0.73	0.03
∑PFPA	0.00	0.00
∑Precursors	0.03	0.00
∑PFAS	**4.47**	**0.27**

For the eluate samples in Table 31 the average PFAS concentrations are significantly lower than for the solid samples, which can be attributed to the fact that not all PFAS have passed into the aqueous phase. However, PFAS could be detected in more samples because the LOD is lower than for the solid samples, resulting in values >0 for the median.

Table 32: PFAS mean and median values in the identified soil samples. All samples from Rastatt were included. All data in µg/kg.

Substance	Mean value	Median
∑PFCA	11.90	0.14
∑PFSA	3.50	0.00
∑PFPA	0.00*	0.00
∑Precursors	2.14	0.00
∑PFAS	**17.53**	**0.21**

* The concentration for PFPA is not zero here, but statistically irrelevant.

The mean and median values over all measured values in Table 32 lie, as expected, between the values in Table 30 and Table 31. Since no PFAS could be detected in many measured values, many of the median values are also zero. Since the majority of the data originates from the district of Rastatt, these data are not necessarily representative for all of Germany.

In addition, comparative values could be obtained from North Rhine-Westphalia. In uncontaminated arable soils, ∑ PFAS values of up to ~7.6 µg/kg could be detected. However, most values were below 1 µg/kg. These concentrations are largely composed of PFOS, PFOA, and PFBA. Higher concentrations were measured in top soils than in subsoils. In selectively contaminated soils, concentrations ranging from a few µg/kg to over 10,000 µg/kg were detected. (LANUV, 2022b). The calculated mean value of ~20 µg/kg in Table 32 agrees well with these values, if it is considered that this value includes both polluted and unpolluted soil.

Lastly, the PFAS concentrations calculated here found in Table 32 can be compared with the measured EOF values (Figure 15). It is assumed that the EOF values are composed of PFAS only. In the measured soils, EOF values of up to 3 mg/kg were detected (1,249 µg/kg mean). In the uncontaminated soils, the EOF values varied between 73-209 µg/kg and were thus higher than the calculated values shown in Table 32.

3.4 Waste data and material flows of PFAS in selected waste streams.

This chapter presents the waste data found for each waste stream and resulting PFAS mass flow. The methodology is described in chapter 2.7 described.

3.4.1 Textiles

3.4.1.1 Waste data

Textiles represent a complex waste stream as they have a wide range of applications. A large proportion of the textiles used are used in clothing, which are collected separately in used clothing containers (compared to textiles such as carpets, car seats, curtain rails, etc. see below). For this data, a statistics report from the Bundesverband Sekundärrohstoffe und Entsorgung e.V. was used, which reports data from 2018 (BVSE, 2020). Table 33 presents the data found.

Table 33: Waste data on separately collected used clothing in 2018 from. (BVSE, 2020). All data in tons.

Emergence	Reuse [62%]	Reuse (cleaning rags) [14%]	Recovery (recycling) (material recovery) [12%].	Recycling (RDF/thermal) [8%]	Waste for disposal [4%]
1.271.242	788.170	177.974	152.549	101.699	50.850

Furthermore, textiles are used as home textiles, work clothes and in vehicles. Home textiles (such as carpets and blankets) are often disposed of in household waste, as are some old textiles such as shoes and cleaning rags. According to Dornbusch et al. (2020) about 3.5% of household waste consists of textiles, which corresponds to a total amount of about 370,000 t per year. This amount is composed of ~40% clothing and ~60% other textiles (curtains, towels, bedding, etc.). The textiles disposed of in this way are mostly of poor quality and can therefore only be reused to a limited extent. It is assumed for the further procedure that these textiles are thermally treated.

Finally, this report considers the waste stream of textiles, which is collected separately under EWC code 07.6 and CN code W076. The former is reported by Destatis and the latter by Eurostat. Both consist of, among others, industrial waste from fiber production, used clothing, textile packaging and leather waste, but the definition of the two streams differs in the exact wording. However, due to the similar waste, tonnages and almost identical numbers, it is assumed that they are the same waste stream. The Eurostat data additionally includes data on the treatment of this waste, which is why it is used in the rest of the project (Eurostat, 2022). The data from the Federal Statistical Office only show the origin of the waste, but not the treatment. Table 34 presents the Eurostat data found.

Table 34: Waste data on separately collected used textiles in 2018 (CN code W076). All data in tons.

Total waste	Disposal - landfill, incineration and others (D1-D12)	Recovery - energy recovery (R1)	Recovery - Recycling	Recovery - backfilling
312.433	0	81.649	227.976	0

3.4.1.2 Material flow

For the material flows of PFAS, the mean and median concentrations from chapter 3.1.1 were multiplied by the waste quantities. This results in the following mass flows in Table 35 and Table 36 for the respective PFAS groups and individual textile streams:

Table 35: PFAS material flows in separately collected used clothing in 2018. The concentrations for the perfluorocarboxylic, -sulfone and -phosphoric acids, as well as for the precursors are shown. A summed value for all PFAS has also been calculated. All values were calculated based on the mean and median values in chapter 3.3 calculated.

Substance	Volume [t]	Reuse [62%] [t]	Further use (cleaning rags) [14%] [t]	Recovery (recycling) [12%] [t]	Recycling (RDF/thermal) [8%] [t]	Waste for disposal [4%] [t]
	1,271,242	788,170	177,974	152,549	101,699	50,850
PFAS quantities from mean value [kg].						
∑PFCA	111.32	69.02	15.58	13.36	8.91	4.45
∑PFSA	26.25	16.28	3.68	3.15	2.10	1.05
∑PFPA	0.00	0.00	0.00	0.00	0.00	0.00
∑Precursors	356.83	221.24	49.96	42.82	28.55	14.27
∑PFAS	**494.40**	**306.53**	**69.22**	**59.33**	**39.55**	**19.78**
PFAS quantities from median [kg].						
∑PFCA	10.63	6.59	1.49	1.28	0.85	0.43
∑PFSA	0.00	0.00	0.00	0.00	0.00	0.00
∑PFPA	0.00	0.00	0.00	0.00	0.00	0.00
∑Precursors	4.40	2.73	0.62	0.53	0.35	0.18
∑PFAS	**94.14**	**58.37**	**13.18**	**11.30**	**7.53**	**3.77**

Table 36: PFAS material flows in separately collected used textiles in 2018 (CN code W076). Shown are the concentrations for the perfluorocarboxylic, -sulfonic and -phosphoric acids, as well as for the precursors. A sum value for all PFAS has also been calculated. All values were calculated based on the mean and median values in chapter 3.3 calculated.

Substance	Total waste [t]	Disposal - landfill, incineration and other (D1-D12) [t].	Utilization - energy recovery (R1) [t].	Recovery - recycling [t]
	312,433	0	81,649	227,976
PFAS quantities from mean value [kg].				
∑PFCA	27.36	0	7.15	19.96
∑PFSA	6.45	0	1.69	4.71
∑PFPA	0.00	0	0.00	0.00
∑Precursors	87.70	0	22.92	63.99
∑PFAS	**121.51**	**0**	**31.75**	**88.66**
PFAS quantities from median [kg].				
∑PFCA	2.61	0	0.68	1.91
∑PFSA	0.00	0	0.00	0.00
∑PFPA	0.00	0	0.00	0.00
∑Precursors	1.08	0	0.28	0.79
∑PFAS	**23.14**	**0**	**6.05**	**16.88**

In addition, there are the 370,000 tons of textiles from household waste that are assumed to be fully incinerated. The PFAS quantities are as shown in the following Table 37:

Table 37: PFAS material flows to waste data on separately collected used textiles in 2018 (CN code W076). Shown are the concentrations for the perfluorocarboxylic, -sulfonic and -phosphoric acids, as well as for the precursors. A sum value for all PFAS has also been calculated. All values were calculated based on the mean and median values in chapter 3.3 calculated.

Substance	Textile waste in household waste [t].
	370,000
PFAS quantities from mean value [kg].	
∑PFCA	27.36
∑PFSA	6.45
∑PFPA	0.00
∑Precursors	87.70

Substance	Textile waste in household waste [t].
∑PFAS	**121.51**
PFAS quantities from median [kg].	
∑PFCA	2.61
∑PFSA	0.00
∑PFPA	0.00
∑Precursors	1.08
∑PFAS	**23.14**

Overall, this results in the following mass flows in Table 38 (divided into reuse/recycling, energy recovery and other disposal):

Table 38: PFAS material flows in textiles in 2018. Concentrations for perfluorocarboxylic, -sulfone and -phosphoric acids as well as for precursors are shown. A summed value for all PFAS has also been calculated. All values were calculated based on the mean and median values in chapter 3.3.

Substance	Total textile waste [t]	Recycling (69%) [t]	Thermal treatment (28.4%) [t].	Other disposal (2.6%) [t].
	1,950,.867	1,346,669	553,348	50,850
PFAS quantities from mean value [kg].				
∑PFCA	170.83	117.92	48.45	4.45
∑PFSA	40.29	27.81	11.43	1.05
∑PFPA	0.00	0.00	0.00	0.00
∑Precursors	547.60	378.00	155.32	14.27
∑PFAS	**758.71**	**523.74**	**215.20**	**19.78**
PFAS quantities from median [kg].				
∑PFCA	16.31	11.26	4.63	0.43
∑PFSA	0.00	0.00	0.00	0.00
∑PFPA	0.00	0.00	0.00	0.00
∑Precursors	6.76	4.67	1.92	0.18
∑PFAS	**144.47**	**99.73**	**40.98**	**3.77**

Thus, the total PFAS load is ~760 kg and ~145 kg, respectively (Table 38), depending on whether the mean or median is considered. Of this, 69% is recycled or otherwise reused (524 kg and 100 kg, respectively), assuming that the PFAS are not destroyed. Approximately 28% of the textiles are recycled for energy, most of which destroys the PFAS. The remaining approx. 3% is disposed of elsewhere (e.g., landfilling).

It should be mentioned here that this calculation assumes that all textiles in the waste stream contain PFAS, which is not the case in reality. The actual PFAS load is likely to be lower. In general, it is assumed that the mean overestimates the load because outliers with high concentrations strongly influence the mean. The median probably underestimates the PFAS load because, for example, sulfonic acids are not included. Due to many measurements in which they were not detected or measured, these show a value of 0 µg/kg in the median.

3.4.2 Sewage sludge

3.4.2.1 Waste data

Statistical data from Destatis, 2020a show that most of the sewage sludge from municipal wastewater treatment plants in Germany is disposed of by thermal treatment, while another part is disposed of by soil-based recycling. 77% of the total amount of sewage sludge was disposed of by thermal treatment (e.g., by thermal waste treatment in mono- and co-incineration plants), while 22% was recycled by application in agriculture and landscaping. The data can be found in Table 39.

Table 39: Sewage sludge disposal from public wastewater treatment in 2020 (Destatis, 2020a). All values in tons.

Total direct sewage sludge disposal	Thereof								
	Material recovery				Thermal disposal				Other direct disposal
	Total	In agriculture	For landscaping measures	Other recycling	Total	Mono incineration	Co-incineration	Unknown	
1.740.556	388,886	259,851	25,181	103,854	1,334,994	507,929	795,819	31,246	16,676

3.4.2.2 Material flow

To calculate the material flows of PFAS, the mean and median concentrations from chapter 3.3.2 were multiplied by the waste quantities. This resulted in the concentrations shown in Table 40 for the respective PFAS groups.

Table 40: **PFAS material flows in sewage sludge in 2020. The concentrations for the perfluorocarboxylic, -sulfone and -phosphoric acids, as well as for the precursors are shown. A summed value for all PFAS has also been calculated. All values were calculated based on the mean and median values in chapter 3.3.**

Substance	**Total direct sewage sludge disposal [t].**	**Thereof**								
		Material recovery				**Thermal disposal**				**Other direct disposal [t]**
		Total [t]	**In agriculture [t]**	**For landscaping measures [t].**	**Other recycling [t]**	**Total [t]**	**Mono-incineration [t]**	**Co-incineration [t]**	**Unknown [t]**	
	1,740,556	388,886	259,851	25,181	103,854	1,334,994	507,929	795,819	31,246	16,676
PFAS quantity from mean value [kg].										
∑PFCA	31.38	7.01	4.68	0.45	1.87	24.06	9.16	14.35	0.56	0.30
∑PFSA	84.94	18.98	12.68	1.23	5.07	65.14	24.79	38.83	1.52	0.81
∑PFPA	-	-	-	-	-	-	-	-	-	-
∑Precursors	0.63	0.14	0.09	0.01	0.04	0.48	0.18	0.29	0.01	0.01
∑PFAS	**116.94**	**26.13**	**17.46**	**1.69**	**6.98**	**89.69**	**34.13**	**53.47**	**2.10**	**1.12**
PFAS quantity from median [kg].										
∑PFCA	28.89	6.46	4.31	0.42	1.72	22.16	8.43	13.21	0.52	0.28
∑PFSA	54.48	12.17	8.13	0.79	3.25	41.79	15.90	24.91	0.98	0.52
∑PFPA	-	-	-	-	-	-	-	-	-	-

Substance	**Total direct sewage sludge disposal [t].**	**Thereof**								
		Material recovery				**Thermal disposal**				**Other direct disposal [t]**
		Total [t]	**In agriculture [t]**	**For landscaping measures [t].**	**Other recycling [t]**	**Total [t]**	**Mono-incineration [t]**	**Co-incineration [t]**	**Unknown [t]**	
∑Precursors	-	-	-	-	-	-	-	-	-	-
∑PFAS	**82.50**	**18.43**	**12.32**	**1.19**	**4.92**	**63.28**	**24.08**	**37.72**	**1.48**	**0.79**

From Table 40 the total PFAS load is 117 kg or ~83 kg, depending on whether the mean or median value is considered. Since 77% (89.7 kg or 63.3 kg) of the total amount of sewage sludge is incinerated, it is assumed that most of the PFAS is destroyed. Approximately 22% (26.1 kg and 18.4 kg, respectively) will be recycled through agricultural and other uses, assuming that PFAS will not be destroyed. The remaining 1% (1.1 kg or 0.79 kg) is disposed of by other means (e.g., composting, anaerobic digestion). As described in chapter 3.4.4 mentioned, the application of contaminated sewage sludge to arable land introduces PFAS into the soil, from where they can eventually be taken up by plants or humans.

The median values are also significantly lower than the mean values here, as precursor substances were often not taken into account. The reason for this are the many measurements where PFAS were not detected or had a value of 0 µg/kg. Therefore, it can be assumed that the median value probably underestimates the PFAS load.

3.4.3 Paper

3.4.3.1 Waste data

As described in chapter 3.1.3.1 PFAS are used in particular in food contact papers, but they also find application in folding cartons, tablecloths, wallpapers, and printer inks, among others (Glüge et al., 2020). They can therefore be found in many different paper and packaging products.

In general, life contact paper has a very short lifespan and is usually disposed of in the same year (Conversio, 2018). A general definition of which type of papers belong to food contact papers can be found in chapter 3.3.3 can be found.

According to the Federal Environment Agency, however, many paper-based food contact papers do not belong in paper waste; instead, they should be disposed of in packaging or household waste. These include, for example, coffee-to-go cups, wet-strength or greaseproof impregnated papers and cardboard such as burger wrappers and muffin tins, and soiled, residue-emptied paper packaging such as pizza boxes or cardboard trays for oven meals (UBA, 2020c). Depending on whether the waste is packaging, e.g. a burger wrapper, then it must be disposed of in packaging waste, but if the waste is not packaging e.g. a paper drinking straw, then it must be disposed of in household waste. Egg cartons, on the other hand, belong in paper waste (UBA, 2020c).

Plastic packaging should be disposed of empty in the yellow bag/bin.

However, it cannot be ruled out that this type of paper is also disposed of in the wastepaper. A proportion of food contact paper in the separately collected wastepaper could not be determined from the literature. In general, contaminated paper is not suitable for recycling (Paper for Recycling Council, 2022).

According to Dornbusch et al. (2020) 21.5% of household waste in Germany is recovered paper (6.6 kg per inhabitant per year), which corresponds to an annual volume of approximately 550,000 tons. This is composed of 40.5% recovered paper and packaging, 38% printer products, 14.1% other recovered paper and 7.4% recovered paper in the range of 10 - 40 mm. Furthermore, in 2017, approximately 66 kg of recovered paper per inhabitant was collected through the separate collection systems in Germany. This corresponds to approx. 5,500,000 tons in total.

Wastepaper waste volumes are also reported on Eurostat under CN code W072 (Eurostat, 2022). This code includes, among others, recycled "waste and wastepaper" or cardboard, old and unsold newspapers, telephone directories and brochures, waste and residual paper or cardboard, mainly wood pulp and sorted and unsorted recovered "waste and wastepaper" or cardboard. A total of ~4,523,000 tons of this was generated in Germany in 2018, with approximately 98% recycled.

The Federal Statistical Office also reports data on wastepaper. The 2020 statistics report indicates a quantity of wastepaper in separately collected household-typical municipal waste (EAV codes 15 01 01 and 20 01 01) of 6,866,000 tons, 99% of which was recycled. The remainder was thermally recycled. Furthermore, EWC code 07.2 indicates a wastepaper quantity of 7,165,00 tons for the year 2020. This code includes paper and cardboard waste from the sorting of the separate collection (EAV codes 15 01 01, 19 12 01 and 20 01 01). Due to the overlap of the EAV codes, it is assumed that these are the same wastes (Destatis, 2020b).

The paper industry also publishes annual statistics, which also include data on recovered paper. It is reported that in 2020 a wastepaper volume of 14,468,000 tons was generated in Germany. Europe-wide, there was a recycling rate of 73.9% in the same year. Information on the treatment of recovered paper was not mentioned in the report (The Paper Industry, 2022).

Finally, a report from the paper industry regarding the industry's residues was also viewed. Here, it is mentioned that in 2020, approximately 3,700,000 tons of residues were generated during paper production. These consist of bark and wood waste, deinking sludge, fiber waste, sludge from wastewater treatment and incineration residues, among other things. Of this, approx. 60 % is thermally recycled. Approx. 20% is recycled, which includes, for example, use in the brick and cement industry. 1.2 % is landfilled and approx. 2 % is biologically recycled. Other recovery routes are reported as 6-7%, and another 6% was sent to other building material recovery in 2020 (The Paper Industry, 2020).

In general, no conclusive data can be found regarding the wastepaper waste stream. It is unknown which relation the found data have to each other and often it is also unknown which wastes are considered in which data. For the further procedure, the wastepaper volume of The paper industry (2022) for 2020 is used, as it is assumed that this data includes all individual wastepaper waste streams. However, since the report does not present any treatment of the waste, a material recovery rate of 80% is assumed, following the data found. Energy recovery is assumed for the remaining 20%, except for the reported 1.2% of paper waste from paper production (44,400 tons, (The Paper Industry, 2020)), which is landfilled. This results in the following waste stream in Table 41:

Table 41: Waste data on recovered paper in Germany. Recycling rates are based on assumptions and data from (The paper industry, 2020, 2022). All values in tons.

Wastepaper volume	Recycling [80%]	Energy recovery [19.69%]	Landfill [0.31%]
14,468,000	11,574,400	2,849,200	44,400

3.4.3.2 Material flow

For the PFAS material flows, the mean and median concentrations from chapter 3.3.3 were multiplied by the waste quantities. This results in the following mass flows for the respective PFAS groups in Table 42:

Table 42: **PFAS material flows in recovered paper in 2020. The concentrations for the perfluorocarboxylic, -sulfone and -phosphoric acids, and for the precursors are shown. A summed value for all PFAS has also been calculated. All values were calculated based on the mean and median values in chapter 3.3 calculated.**

Substance	Wastepaper volume [t]	Recycling [80%]	Energy recovery [19.69%]	Landfill [0.31%]
	14,468,000	11,574,400	2,849,200	44,400
PFAS quantities from mean value [kg].				
∑PFCA	2,255.06	1,804.05	444.09	6.92
∑PFSA	21,36	17,09	4,21	0,07
∑PFPA				
∑Precursors	1,685.03	1,348.03	331.84	5.17
∑PFAS	**3,961.46**	**3,169.17**	**780.13**	**12.16**
PFAS quantities from median [kg].				
∑PFCA	8.68	6.94	1.71	0.03
∑PFSA	10.13	8.10	1.99	0.03
∑PFPA				
∑Precursors				
∑PFAS	**150.47**	**120.37**	**29.63**	**0.46**

Overall, according to Table 42 a PFAS load of 3,961 or 150 kg. As with the other waste streams, it is assumed that the average value overestimates the PFAS load, since not all papers are PFAS-contaminated. The median probably underestimates the load because precursors are not included.

Since a large part of the recovered paper is recycled, the PFAS are also recycled (~3.2 tons or ~120 kg). However, it can be assumed that a portion of the PFAS also accumulates in the wastewater or fiber sludge from the recycling facility and is eventually sent to a wastewater or sludge treatment facility. Such fiber sludges are predominantly incinerated, and it can be assumed that the PFAS are destroyed. It is assumed that only about 20% of the PFAS are thermally recycled and destroyed in the process. 0.46-12 kg end up in German landfills.

It should generally be noted that the PFAS load presented here is higher than the actual load, as only food contact papers were considered in the PFAS concentrations. However, for many papers in the wastepaper stream it can be assumed that they are often not contaminated with PFAS (e.g. newspapers, books, etc.), but are included by the calculation. Since no food contact paper was identified in the wastepaper stream, this calculation overestimates the PFAS load.

3.4.4 Soils

3.4.4.1 Waste data

Data on waste soils were found at Eurostat under EWC code W126. (Eurostat, 2022). The code includes soils (including excavated soil from contaminated sites), stones and dredged material.

Soil material at the place of origin (including contaminated soils that are not excavated) as well as uncontaminated soil material and other naturally occurring materials excavated during construction works, provided that it is ensured that the materials are used for construction purposes in their natural state at the place where they were excavated, are not covered by the concept of waste soils.

The data from 2018 are presented in Table 43.

Table 43: Soil disposal in Germany in 2018. All data in tons. (Eurostat, 2022)

Waste total	Elimination			Recovery		
	Landfill	Incineration	Other	Energy recovery	Recycling	Backfilling
128,858,796	19,193,317	20,697	52,310	10,763	20,830,599	88,751,111

Of the 128.9 million tons of waste soils in Germany from Table 43 14.95% was disposed of, while 85.05% was recycled. Most of the disposed waste was landfilled (14.89 %), *i.e.* it was deposited in/on a landfill/special landfill or permanently stored in a container, e.g. in a mine. Only a small fraction was incinerated (0.02%) or, if the contamination is in the form of liquid or sludge waste, biodegraded (0.04%) (Eurostat, 2022).

In terms of recovery methods, most of the soil (68.87%) was recovered by backfilling, which means that suitable non-hazardous waste was used for recultivation of excavated areas, as is often the case in construction, or for technical purposes in landscaping. 1.85% of the total soil waste generated in 2018 was hazardous waste, the rest was non-hazardous waste.

16.17% was recovered by recycling, i.e. by reprocessing organic material until it is ready to be used as originally intended. Only a small part of the contaminated soil (0.01%) was used in thermal waste treatment plants for energy production.

3.4.4.2 Material flow

To calculate the material flows of PFAS, the mean and median concentrations from chapter 3.3.4 were multiplied by the waste quantities. This results in the following mass flows for the respective PFAS groups, which are shown in the following tables (Table 44, Table 45, Table 46).

Table 44: PFAS material fluxes in soils in 2018. Eluate samples from Rastatt were not included. Shown are the concentrations for the perfluorocarboxylic, -sulfone and -phosphoric acids, as well as for the precursors. A sum value for all PFAS has also been calculated. All values were calculated based on the mean and median values in chapter 3.3 calculated.

Substance	Total waste [t]	Elimination			Recovery		
		Landfill [t]	Incineration [t]	Other [t]	Energy recovery [t]	Recycling [t]	Backfilling [t]
	128,858,796	19,193,317	20,697	52,310	10,763	20,830,599	88,751,111
PFAS quantity from mean value [kg].							
∑PFCA	3,324.39	495.16	0.53	1.35	0.28	537.40	2,289.67
∑PFSA	1.055.30	157.18	0.17	0.43	0.09	170.59	726.83

Sub-stance	**Total waste [t]**	**Elimination**			**Recovery**		
		Landfill [t]	**Incineration [t]**	**Other [t]**	**Energy recovery [t]**	**Recycling [t]**	**Backfilling [t]**
∑PFPA	0.11	0.02	0.00	0.00	0.00	0.02	0.08
∑Precursors	736.94	109.77	0.12	0.30	0.06	119.13	275.58
∑PFAS	**5,116.74**	**762.13**	**0.82**	**2.08**	**0.43**	**827.14**	**3,524.14**
PFAS quantity from median [kg].							
∑PFCA	0.00	0.00	0.00	0.00	0.00	0.00	0.00
∑PFSA	0.00	0.00	0.00	0.00	0.00	0.00	0.00
∑PFPA	0.00	0.00	0.00	0.00	0.00	0.00	0.00
∑Precursors	0.00	0.00	0.00	0.00	0.00	0.00	0.00
∑PFAS	0.00	0.00	0.00	0.00	0.00	0.00	0.00

If PFAS were measured only in soil samples (Table 44), the total PFAS load was 5,116 kg or 0 kg, depending on whether the mean or median value is considered. The median values are 0 because most of the samples found show concentrations below the LOD (i.e., a value of 0), so the median is also 0.

Table 45: PFAS material fluxes in soils in 2018. Sediment samples from Rastatt were not included.

Sub-stance	**Total waste [t]**	**Elimination**			**Recovery**		
		Landfill [t]	**Incineration [t]**	**Other [t]**	**Energy recovery [t]**	**Recycling [t]**	**Backfilling [t]**
	128,858,796	19,193,317	20,697	52,310	10,763	20,830,599	88,751,111
PFAS quantity from mean value [kg].							
∑PFCA	477.77	71.16	0.08	0.19	0.04	77.23	329.06
∑PFSA	94.02	14.00	0.02	0.04	0.01	15.20	64.75
∑PFPA	0.00	0.00	0.00	0.00	0.00	0.00	0.00
∑Precursors	3.77	0.56	0.00	0.00	0.00	0.61	2.60
∑PFAS	**575.56**	**85.73**	**0.09**	**0.23**	**0.05**	**93.04**	**396.41**
PFAS quantity from median [kg].							
∑PFCA	26.29	3.92	0.00	0.01	0.00	4.25	18.11
∑PFSA	3.61	0.54	0.00	0.00	0.00	0.58	2.49
∑PFPA	0.00	0.00	0.00	0.00	0.00	0.00	0.00

Sub-stance	**Total waste [t]**	**Elimination**			**Recovery**		
		Landfill [t]	**Incine-ration [t]**	**Other [t]**	**Energy recovery [t]**	**Recycling [t]**	**Backfilling [t]**
∑Precur-sors	0.00	0.00	0.00	0.00	0.00	0.00	0.00
∑PFAS	**34.53**	**5.14**	**0.01**	**0.01**	**0.00**	**5.58**	**23.79**

In eluate samples in Table 45 the total PFAS load is 575 kg or 35 kg, depending on whether the mean or median value is considered.

Table 46: PFAS material fluxes in soils in 2018. All samples from Rastatt were included.

Sub-stance	**Total waste [t]**	**Elimination**			**Recovery**		
		Landfill [t]	**Incinera-tion [t]**	**Other [t]**	**Energy recovery [t]**	**Recycling [t]**	**Backfilling [t]**
	128,858,796	19,193,317	20,697	52,310	10,763	20,830,599	88,751,111
PFAS quantity from mean value [kg].							
∑PFCA	1,533.09	228.35	0.25	0.62	0.13	24.83	1,055.91
∑PFSA	450.39	67.08	0.07	0.18	0.04	72.81	310.20
∑PFPA	0.04	0.01	0.00	0.00	0.00	0.01	0.03
∑Precursors	275.58	41.05	0.04	0.11	0.02	44.55	189.80
∑PFAS	**2,260.37**	**336.68**	**0.36**	**0.92**	**0.19**	**365.40**	**1,556.82**
PFAS quantity from median [kg].							
∑PFCA	17.85	2.66	0.00	0.01	0.00	2.89	12.29
∑PFSA	0.00	0.00	0.00	0.00	0.00	0.00	0.00
∑PFPA	0.00	0.00	0.00	0.00	0.00	0.00	0.00
∑Precursors	0.00	0.00	0.00	0.00	0.00	0.00	0.00
∑PFAS	**26.54**	**3.95**	**0.0043**	**0.01**	**0.0022**	**4.29**	**18.28**

Taking into account all samples analyzed, both solids and eluate, the PFAS loading in Table 46 total ~2,260 kg when the mean value is considered and ~26 kg when the median value is considered. In this case, the PFAS load is between the values in Table 44 and Table 45.

Only small amounts of PFAS are disposed of by thermal waste treatment (0.36 kg and 0.0043 kg, respectively), including energy recovery (0.19 kg and 0.0022 kg, respectively), leaving a large amount of PFAS in the contaminated soil to be reused. A large proportion of soils in Germany are backfilled (~70%), leaving PFAS in nature to be taken up by plants and animals.

These values are loaded with a high uncertainty, which is due to the methodology applied and the fact that a concentration has been derived for both polluted and unpolluted soils. An uncertainty of a factor of 10 is considered realistic, since the concentrations calculated here (see chapter 3.3.4) are in agreement with the concentrations in Table 57 and from (LANUV, 2022b) sufficiently correspond.

In general, reliable data for PFAS contamination in soils in Germany are needed in order to calculate realistic mass flows. Background levels in soils are being determined in a research project of the UBA. Results are expected in 2025/2026.

3.4.5 Summary

The following tables, Table 47 and Table 48, highlight the annual PFAS mass flows in the waste streams considered. A distinction is made between the type of treatment, which allows the annual amount of PFAS recycled, landfilled and backfilled to be identified.

For the soil waste stream, measurements of both the solid and the eluate are considered.
It should be taken into account that the values are loaded with many uncertainties (see chapter 3.3) and should therefore only be regarded as an estimate.

Table 47: Summary of calculated PFAS mass flows from the mean of the selected waste streams. All figures in kg.

Waste stream	Total PFAS quantity	Amount of PFAS recycled/materially recovered	Amount of PFAS recycled for energy	Quantity PFAS deposited/other
Textiles	758.71	523.74	215.20	19.78
Sewage sludge	116.94	26.13	89.69	1.12
Paper	3,961.46	3,169.17	780.13	12.16
Soils	2,260.37	1,922.22	0.55	337.60
Total	**7,097.48**	**5,641.26**	**1,085.57**	**370.66**

If the mean value in Table 47 is considered, the waste streams analyzed contain a total of~7,100 kg of PFAS. More than half of this (~56%) comes from paper waste, while only ~1.6% comes from municipal sewage sludge. The high PFAS load in the paper samples is not considered realistic, as only PFAS readings from food contact paper were found. However, many papers in the paper waste stream are not or only partially contaminated with PFAS (e.g., newspapers, books, etc.), which probably overestimates the PFAS load. The high PFAS amounts in the soil samples can be attributed to the comparatively very large amounts of waste soil collected (~130,000,000 t compared to ~17,000,000 t of wastepaper).

Table 48: Summary of calculated PFAS mass flows from median of selected waste streams. All figures in kg.

Waste stream	Total PFAS quantity	Amount of PFAS recycled/materially recovered	Amount of PFAS recycled for energy	Quantity PFAS deposited/other
Textiles	144.47	99.73	40.98	3.77
Sewage sludge	82.50	18.43	63.28	0.79

Waste stream	Total PFAS quantity	Amount of PFAS recycled/materially recovered	Amount of PFAS recycled for energy	Quantity PFAS deposited/other
Wastepaper	150.47	120.37	29.63	0.46
Soils	26.54	22.57	0.01	3.96
Total	**403.98**	**261.1**	**133.9**	**8.98**

If the median values in Table 48 are considered, the waste streams investigated contain a total of~ 400 kg PFAS. Here, too, the median values are significantly lower than the mean values. The reasons for this have already been discussed in chapter 3.3 discussed.

In the following procedure, the mean values are considered, since they represent the worst-case scenario in the context of this project.

3.5 Presentation of the environmental context for the selected waste streams.

In this chapter, the environmental context and risks related to the four waste streams are discussed. For this purpose, the questions from chapter 2.8 are used as a basis.

Disregarding the limit values in the legal background chapter (1.2), the following environmentally relevant limit values for PFAS apply in Germany.

TA-Luft

In chapter 5.2.7 of the TA-Luft, exhaust gas limits are specified for carcinogenic and reproduction-toxic substances. Carcinogenic substances are divided into three classes, with individual compounds mentioned by name. PFAS are not mentioned. Substances that are not mentioned should be assigned to the three classes based on their potency. If the potency is unknown, they should be assigned to Class 1 as a precaution. The emission limit value for Class I is 0.15 g/h as mass flow or 0.05 mg/m^3 as mass concentration. Some PFAS such as PFOS and PFOA are classified as potentially carcinogenic (H351). Thus, they do not fall under the scope of carcinogenic substances, but under regular organic substances of Class I under chapter 5.2.5. Here, a waste gas mass flow of 0.1 kg/h or a mass concentration of 20 mg/m^3 applies. (TA-Luft, 2022). If the carcinogenicity of a PFAS is proven and it is classified as such, the stricter limit value for carcinogenic substances applies.

PFOS and PFOA are additionally classified as reprotoxic substances (H360D), which means that a waste gas limit value as a mass flow of 2.5 g/h or as a mass concentration of 1 mg/m^3 must not be exceeded (TA-Luft, 2022).

Since many PFAS are not yet classified, but it can be assumed that they have similar properties, the strictest relevant limit value should always be applied as a precaution.

TRGS 900

Furthermore, the Technical Rules for Hazardous Substances (TRGS) 900 regulates occupational exposure limits for various compounds. Of the PFAS, only PFOS is mentioned, for which an occupational exposure limit of 0.01 mg/m^3 is prescribed. (TRGS 900, 2022). Since no other PFAS are mentioned, this limit value is taken as the value for all PFAS for the environmental assessment.

MAK Commission

The Senate Commission for the Testing of Hazardous Substances (MAK Commission) advises the German Research Foundation as well as the federal and state governments, parliaments and authorities on health protection issues when handling hazardous substances, especially in occupational health and safety. Within the scope of its activities, it also proposes maximum workplace concentrations (MAK values), which, however, are not legally binding. For PFOA and PFOS, a MAK value of 0.05 mg/m^3 and 0.01 mg/m^3, respectively, is proposed. (DFG, 2022). The TLV for PFOS has already been implemented in TRGS 900 as a legally binding occupational exposure limit value (OEL), but the TLV for PFOA has not yet been included in TRGS as a legally binding OEL.

3.5.1 Textiles

Table 49: Environmental context of PFAS in the textile waste stream for the treatment routes of material recovery, thermal disposal, and landfill/other.

Waste treatment	Quantity of the substance (kg/a)	Possible risks related to the PFAS content
Material recovery	524	Almost 70% of the collected textile waste is recycled, which means that 524 kg of PFAS are not destroyed and can therefore pose a risk to humans and the environment. Often, the collected textile waste is further processed into cleaning rags or insulation materials (Celep, 2022). The wide variety of materials in the waste stream makes it difficult to find suitable and viable recycling methods. This is especially true for garments that contain many components such as labels, threads, buttons, zippers or a mixture of different types of materials. Currently, there are two main textile recycling processes, one is mechanical recycling processes and the other is chemical recycling processes. In mechanical recycling processes, textile waste is mechanically stressed and shredded. Since this produces fibers with shorter length and deteriorated properties, blending with new fibers is required (Damayanti et al., 2021). This can lead to PFAS contamination of the new fibers if the recycled textiles were also treated with PFAS. Chemical recycling processes, on the other hand, where chemical solvents are used to dissolve the fibers, do not harm the fibers and no new materials need to be added. Likewise, pollutants such as additives and applied chemicals (such as PFAS) can be largely separated in this process (CEFIC, 2022). Chemical recycling processes are not expected to result in the proliferation of PFAS; however, chemical recycling processes currently account for less than 1% of textile recycling (McKinsey & Company, 2022). As both methods require pure waste streams, there is a high probability that personnel will come into contact with the waste during sorting and separation of the waste (manual sorting of the waste depending on the quality and color of the fabric, manual removal of unwanted parts). In addition, they may come into contact with the waste when loading the machines, posing an exposure risk for PFAS. There is a risk of personnel coming into contact with the fabric contaminated with PFAS. Although dermal ingestion of PFAS has been identified as a potential transmission route for PFAS, scientific data on the risk it could pose are lacking (Ragnarsdóttir et al., 2022). However, a study that investigated possible routes of human exposure to PFAS, including dermal ingestion, concluded that food and beverage ingestion is the more important route of human exposure to PFAS than dermal ingestion (Poothong et al., 2020). After shredding, the fibers are washed to remove dirt and pollutants. As a result, the PFAS contained can get into the wastewater. The limit values in Annex 38 of the Wastewater Ordinance generally apply here, but no limit values for PFAS are listed there. Since wastewater treatment plants do not degrade or destroy PFAS efficiently enough, they usu-

Waste treatment	Quantity of the substance (kg/a)	Possible risks related to the PFAS content
		ally end up in the sewage sludge (shorter PFAS tend to end up in the water phase, while longer-chain PFAS tend to accumulate in the solid phase). As already discussed in chapter 3.1.4.2.2, the use of sewage sludge as fertilizer on agricultural land can lead to contamination of the soil and ultimately also of the groundwater. Since PFAS are usually not destroyed during recycling processes, they can pose a serious risk to workers involved in recycling processes. Due to the higher concentration of PFAS in the working environment, workers are more likely to be exposed to them than the general population, especially if the textiles have to be manually separated into different fiber mixtures before recycling, or are shredded during recycling. In this process, compounds with high vapor pressure in particular pass into the gas phase. A study by Schlummer et al. (2013) was able to detect various FTOH in the air of a carpet store, two offices, two sports stores and two outdoor stores. The measured concentrations could be attributed to the use of PFAS in the textiles present there (carpets in offices and in the carpet store, outdoor and sports textiles in the sports and outdoor stores). A maximum concentration of 285.8 ng/m³ (8:2 FTOH) could be measured, which is clearly below the TRGS 900 limit value of 0.01 mg/m³ for PFOS (since no limit value for FTOH is available, the limit value of PFOS is used as a reference value). Fluorotelomer alcohols are also a very volatile class of compounds, which means that PFCA and PFSA concentrations can be assumed to be in an even lower range. As elevated concentrations of airborne PFAS could be encountered in a recycling facility of appropriate textile wastes (e.g. carpets), consideration could be given to taking personal protective measures to minimize the risk of PFAS and avoid their inhalation and possible skin contact. This could include, for example, wearing respirators and gloves. Appropriate air measurements should be taken to determine the need for such protective measures. Generally, high PFAS concentration has been found in textiles that are more exposed to outdoor environment and adverse weather conditions. To name a few: Samples of awning cloths (Janousek, Lebertz, et al., 2019) As well as work clothing and outdoor jackets (Gremmel et al., 2016) all showed PFAS concentrations above 1000 µg/kg. To mitigate the risk of such products (which are highly likely to be significantly contaminated with PFAS), they could be separated from other textile waste and treated with an appropriate waste treatment method, e.g. thermal.
Thermal treatment	215.20	In total, 26.4% of the textile waste is recycled for energy, which corresponds to a PFAS load of approx. 215 kg. According to the Industrial Emissions Directive (2010/75/EU) (IED), European waste incinerators must maintain a minimum temperature of 850°C and a minimum residence time of 2 seconds. Temperatures of at least 1,100°C are required for the thermal treatment of hazardous waste. In Germany, these regulations are implemented via the 17th BImSchV. During thermal treatment of PFAS, in the optimum case, CO_2, water and HF or fluoride salts are formed, but destruction is not always complete under the above conditions. Aleksandrov et al. (2019) studied the thermal waste treatment of PTFE (Teflon) under conditions similar to those in thermal waste treatment plants and analyzed the waste gas for perfluoro acids (perfluorocarboxylic acids and perfluorosulfonic acids). Only negligible amounts of perfluoro acids were

Waste treatment	Quantity of the substance (kg/a)	Possible risks related to the PFAS content
		detected, suggesting that the off-gas is not a significant source of perfluoro acids. However, no short-chain gases, such as tetrafluoromethane (CF_4), trifluoromethane (CHF_3), hexafluoroethane (C_2F_2), etc., were measured. Some studies were able to detect the presence of such compounds in the exhaust gas after thermal utilization of perfluorinated substances (Geertinger et al., 2019; Huber et al., 2009; Taylor et al., 2014; Taylor & Yamada, 2003). In the literature, a temperature of 1,400°C has been reported for the destruction of CF_4 (Tsang et al., 1998; US EPA, 2020), which is not reached by European waste incinerators. Thus, there is a risk that such compounds will be emitted through the exhaust gas during thermal recycling of PFAS. In order to ensure that all PFAS are destroyed, it is recommended to perform measurements of short-chain F-gases in the exhaust gases of waste incineration plants. Furthermore, perfluoro acids were measured in the ash in some studies, but the concentrations are in the pg/g to ng/g range in these cases (Rijkswaterstaat, 2020; Sandblom, 2014; Wohlin, 2020). In Europe, the ashes are often used in the construction sector, e.g., for road construction or as an aggregate in cement (Blasenbauer et al., 2020; EEA, 2006). In Germany, 70% of the ashes are deposited in landfills (Blasenbauer et al., 2020), whereby PFAS can be eluted and found in the leachate. In the case of backfilling measures, it is assumed that the PFAS are also not destroyed. As part of the total PFAS restriction process, a mass flow for PFAS in ash from waste incinerators was calculated, obtaining a total mass flow of only less than 100 kg for all of Europe (Annex XV, 2023b), suggesting that PFAS are largely destroyed and that ashes from thermal recycling are not a significant source of PFAS. Generally, destruction rates of >99.95% are reported. (Geertinger et al., 2019; Ministry of Japan, 2013; Taylor et al., 2014). which suggests that, although municipal waste incinerators are not capable of completely destroying PFAS, they largely destroy them and, therefore, no significant amounts of PFAS are emitted from waste incinerators. It is therefore recommended that PFAS-contaminated fractions of textile waste be fed into thermal waste treatment wherever possible, as this largely destroys the PFAS they contain.
Deposit in landfills/other	19.78	About 2.6% of collected textile waste is disposed of in other ways. Since in BVSE (2020) however, no information is given on the disposal method, it is unknown what happens to these textiles. Textiles are not allowed to be landfilled in Germany because they do not comply with the allocation values of the Landfill Ordinance (loss on ignition mostly too high). Therefore, no risk assessment can be carried out.

3.5.2 Sewage sludge

Table 50: Environmental context of PFAS in the municipal sewage sludge waste stream for the treatment routes of material recovery, thermal disposal, and landfill/other.

Waste treatment	Quantity of the substance (kg/a)	Possible risks related to the PFAS content
Material recovery	26.13	In 2020, about 22 % of the sewage sludge from municipal wastewater treatment plants in Germany was recycled (soil-related). Since PFAS are not degraded in the wastewater treatment plants and ultimately end up partially in the sewage sludge, according to the values analyzed in this study, the amount of 26 kg/a PFAS is included in the material recycling routes (see chapter 3.4.1.2). In general, it is assumed that PFAS are not destroyed during the recycling of sewage sludge and that this disposal route therefore represents a risk. A number of studies show that PFAS have been detected in sewage sludge intended for application to soils as well as in treated soils (after soil cleaning) (Bolan et al., 2021). Since transfer from the contaminants to the soil can occur, there is still a risk that they can be taken up by plants or eventually enter the human food chain. Bioaccumulation of PFAA has been noted, for example, in vegetable crops such as lettuce or tomato planted on soils treated with sewage sludge (Blaine et al., 2013; Scheurer et al., 2021). In addition, PFAS can be translocated deeper into the soil with stormwater and eventually leach into groundwater. The risk of groundwater contamination is particularly high for short-chain PFCAs (<C8), which have a greater potential to be leached and thus be transported deep into the soil or leach into groundwater (Hamid & Li, 2016). However, the long-chain PFAA can be found more frequently in sewage sludge, while short-chain PFAA bind less to solids than long-chain compounds due to their short chains (Stahl et al., 2018). The largest share of material recycling is in agriculture with approx. 15 % of the total sewage sludge volume, while approx. 1 % is used in landscaping measures (e.g. recultivation) and approx. 6 % in other material recycling (e.g. humification or composting) (see chapter 3.4.2). As a rule, further sludge treatment processes are carried out before the sewage sludge is used in agriculture or landscaping. These include thickening or dewatering, composting, fermentation, and/or heat treatment, which, however, only occur at temperatures up to about 200°C (LfU Bavaria, 2003). According to studies, various sludge treatment processes are not, or only minimally, suitable to remove PFAS, or in some processes led to an increased PFAS concentration in the resulting sewage sludge, in particular due to the degradation of precursor substances (Lakshminarasimman et al., 2021; W. Zhang et al., 2022). Direct contact of personnel with sewage sludge during treatment processes is considered unlikely. However, studies show that emissions into the air from sewage treatment plants, especially FTOHs, have been (Ahrens et al., 2011; Frömel et al., 2016). It can be assumed that personnel may be exposed to these emissions in the wastewater treatment plant as well as in further treatment processes, but the measured FTOH concentrations are far below the TRGS 900

Waste treatment	Quantity of the substance (kg/a)	Possible risks related to the PFAS content
		limit value of 0.01 mg/m³ for PFOS (since no limit value for FTOH is available, the limit value of PFOS is used as a reference value). The utilization of sewage sludge, sewage sludge mixtures or sewage sludge compost as a substance according to fertilizer law is regulated by the specifications of the AbfKlärV and restricts this to soils of agriculture and landscaping (LfU Bavaria, 2022). A large number of agricultural areas are already excluded by the AbfKlärV such as permanent grassland, fruit and vegetable cultivation areas, arable fodder cultivation areas, ecological farming or areas in water protection areas, nature conservation areas and national parks (LfU Bavaria, 2019). Potential areas of application include crops such as cereals, oilseeds and energy crops, or limited use in the cultivation of sugar beets and corn if the sewage sludge is incorporated before sowing. In such crops, the PFAS contained in the sewage sludge can then be recovered as they have been taken up by the plants. Since 2017, the DüMV contains a limit value for PFAS (Annex 2 Tab. 1.4 No. 1.4.9) of 100 µg/kg DM as well as a labeling threshold from 50 µg/kg DM, which is defined as the sum value of PFOS and PFOA. If the limit value is exceeded, the sewage sludge may not be utilized as fertilizer and therefore may not be applied to soils (LfL, 2021). According to the Sewage Sludge Ordinance, sewage sludge must generally be regularly tested for PFOS and PFOA. Nine of the total of 71 samples considered in this study are above the DüMV limit of 100 µg/kg for PFOS and PFOA and a further eleven are above the value for mandatory labeling of 50 µg/kg for PFOS and PFOA. Most of the studies considered examined sewage sludge from municipal wastewater treatment plants, of which eight of the samples were above the DüMV limit. Becker et al. (2008) (2008), sewage treatment plants that treat only wastewater from households have significantly lower PFOS and PFOA concentrations. The UBA study by Frömel et al. (2016) highlights the difference between industrial and municipal wastewater treatment plants, because there all strongly increased concentrations of sewage sludge with sum values of 294 - 354 µg/kg for PFOS and PFOA could be assigned to an industrial wastewater treatment plant. The data obtained from the LANUV (2022a) were also analyzed for their PFOS and PFOA concentrations. Only sewage sludge intended for use as fertilizer was analyzed. Out of 1362 individual samples, only 26 were above the limit of 50 µg/kg and two samples were above the limit of 100 µg/kg for the sum of PFOS and PFOA, which therefore must not be used as fertilizer. Calculations by Stahl et al. (2018) according to which the mass of sewage sludge applied to agricultural and landscaping soils in Germany in 2015 resulted in the release of 9.3 kg of PFOA and PFOS into the environment. In contrast, this study calculated that approximately 26 kg of PFAA enters the environment through the recycling of PFAS in agriculture and landscaping. However, this value includes more PFAS than just PFOS and PFOA.

Waste treatment	Quantity of the substance (kg/a)	Possible risks related to the PFAS content
		Some federal states, such as Bavaria, had already decided before the amended AbfKlärV came into force that the spreading of sewage sludge in agriculture should be ended in the medium term (LfL, 2021). According to the 2017 AbfKlärV, the use of sewage sludge on soils in Germany will only be possible for sewage sludge from small wastewater treatment plants (< 100,000 p.e. from 2029, < 50,000 p.e. from 2032). The application is only possible if the sewage sludge complies with the requirements of the AbKlärV and the fertilizer law. Thus, it is obligatory to determine the PFAS concentration, to feed contaminated sewage sludge to energy recovery and thus to minimize the risk to soils, plants, groundwater and humans.
Thermal treatment	89.69	In total, 77% of municipal sewage sludge in Germany is thermally treated, which corresponds to a PFAS load of approx. 90 kg. The environmental context of waste incineration is described in chapter 3.5.1 described.
Deposit in landfills/other	1.12	Approx. 1% of the sewage sludge is otherwise recycled, which includes, for example, delivery to drying plants and unknown disposal routes. In total, these disposal routes result in a load of approx. 1 kg PFAS. Since it is assumed that these disposal routes do not destroy the sewage sludge but recycle it, reference is made to the above chapters regarding recycling for the environmental context and risks. The landfilling of sewage sludge is prohibited in Germany.

3.5.3 Paper

Table 51: Environmental context of PFAS in the paper waste stream for the treatment routes of recycling, thermal disposal, and landfill/other.

Waste treatment	Quantity of the substance (kg/a)	Possible risks related to the PFAS content
Material recovery	3,169.17	It is assumed that approx. 80% of the wastepaper volume is recycled (see chapter 3.4.3.2), which corresponds to a quantity of 3,170 kg PFAS in this project. At the recycling plant, the paper is first sorted and separated to obtain different paper categories (e.g. cardboard, magazine paper, computer paper, etc.). If sorting is done manually, there is a risk of personnel coming into contact with PFAS-contaminated paper. Although dermal ingestion of PFAS has been identified as a potential transmission route for PFAS, scientific data on the risk it may pose are lacking (Ragnarsdóttir et al., 2022). However, a study that investigated possible routes of human exposure to PFAS, including dermal ingestion, concluded that food and beverage ingestion is the more important route of

Waste treatment	Quantity of the substance (kg/a)	Possible risks related to the PFAS content
		human exposure to PFAS than dermal ingestion (Poothong et al., 2020). Since there are PFAS approved for use in food contact papers, the risk of adverse health exposure from dermal contact with PFAS in the waste phase of papers is considered low. After sorting, the paper is shredded and finally mixed with water and chemicals to produce paper fiber. The shredding process generates dust, which in turn creates a higher risk of inhalation of small contaminated particles by workers. In addition to physical skin contact, the possibility of PFAS inhalation should not be ignored. In a study by. Strakova et al. (2021) semi-volatile precursors (e.g., FTOH) could be detected most frequently in the analyzed paper-based food packaging. FTOH have a higher vapor pressure than perfluoro acids (perfluorocarboxylic acids and perfluorosulfonic acids) and thus can be detected more often in the gas phase (Ahrens et al., 2011; Schlummer et al., 2013). Schlummer et al. (2013) measured FTOH in a kitchen where fluorine-coated papers are handled and were able to measure detectable amounts of FTOH, but all measured values were below 0.5 ng/m³, which means that the occupational limit value of TRGS 900 of 0.01 mg/m³ for PFOS is not exceeded (since no limit value for FTOH is available, the limit value of PFOS is used as a reference value). It can be assumed that PFAS concentrations in paper recycling plants and plants using recycled paper could be higher, but this should be checked by measurements. PFAS can also end up in wastewater, as large amounts of water are used in recycling. This is particularly the case with short-chain PFAS. Since these are very mobile and have a low adsorption potential, they can easily enter water (Brendel et al., 2018). Long-chain PFAS, on the other hand, tend to bind more to solid particles (Knutsen et al., 2019). Here, the limit values in Annex 28 of the Wastewater Ordinance generally apply, but no limit values for PFAS are listed there, only a requirement to minimize the quantities used. Strakova et al. (2021) were able to detect PFAS in both compostable takeaway foods (e.g., sugarcane husks) and fast-food chain takeaway packaging (e.g., pizza box, burger paper, doughnut bag). Since these products are usually contaminated with oil and food residues after use, they should be disposed of in the yellow garbage can (as light packaging) and not in the wastepaper garbage can. If they are mistakenly disposed of in the wastepaper garbage can, this can lead to further contamination of new products. Similarly, PFAS from compostable materials can contaminate biowaste and future compost for soil fertilization. Therefore, papers potentially containing PFAS should not be composted. This was most likely the case in Rastatt, where PFAS-contaminated paper sludge and compost were applied to cropland soils over several years, resulting in significantly higher PFAS concentrations in these soils (Rastatt, 2022a). In general, unapproved PFAS, through the use of recycled papers, but also virgin papers, can come into contact with the food when used in food packaging and eventually migrate from the paper into the food. A study by (Fengler et al., 2011) confirmed that PFAS, particularly FTOH, can migrate from food packaging into food at certain temperatures. To minimize the risks, highly contaminated paper waste or paper waste with a high risk of containing PFAS could be separated from the rest of the paper waste and treated with an appropriate waste treatment process, e.g. thermal waste treatment.

Waste treatment	Quantity of the substance (kg/a)	Possible risks related to the PFAS content
Thermal treatment	780.13	A total of 19.3% of the paper waste is recycled for energy, which corresponds to a PFAS load of approx. 780 kg (see chapter 3.4.3.2). The environmental context of waste incineration is described in chapter 3.5.1 described.
Landfill/other	12.16	Paper waste is landfilled only to a small extent (0.3%) due to the system (The Paper Industry, 2020), which corresponds to a PFAS load of about 12 kg. However, the majority of paper waste can be thermally treated. Nevertheless, risks cannot be excluded. PFAS can be detected in leachate from landfills (Rijkswaterstaat, 2020), suggesting that they are washed out by rain and weather and enter the leachate. Perfluoroalkyl acids can also enter leachate through the decomposition of side-chain fluorinated polymers. These polymers have a carbon main chain and PFAS as a side chain, which are typically linked to the main chain via ester or amide bonds. These bonds are easily cleaved, allowing the side chain PFAS to slowly separate and leach out over time (OECD, 2022). The leachate collects on the bottom of the landfill. Many modern landfills are equipped with collection systems to collect the leachate that accumulates. This is then either treated on site or discharged directly to a wastewater treatment plant, but as mentioned earlier, these are not capable of destroying PFAS (W. Zhang et al., 2022), nor is on-site treatment of landfill leachate. (Travar et al., 2020). In the U.S., it has been shown that WWTPs receiving leachate from landfills have up to three times higher PFAS concentrations in influent than WWTPs not receiving leachate. PFAS concentrations in the leachate were 10 times higher than in the influent of the WWTPs (Masoner et al., 2020). In this context, short-chain PFAS tend to accumulate in leachate, while long-chain PFAS are more likely to bind to solids (Knutsen et al., 2019). Therefore, both untreated and treated leachate pose a risk. For example, wastewater contaminated with PFAS can enter surface waters and eventually enter the food chain. Likewise, leachate can leach into the soils of the landfill and contaminate them with the pollutants it contains. Landfills and wastewater treatment plants can also emit PFAS to air, particularly semi-volatile precursors (e.g., FTOH, FOSE, FOSA) that are commonly used in the textile industry (Ahrens et al., 2011; B. Wang et al., 2020). Once airborne, PFAS can travel long distances and deposit on the ground, which is why some PFAS, PFOS, PFOA, and PFHxS have been identified as persistent organic pollutants (POPs). Near their release, they can also be inhaled by local people. In Canada, FTOH, FOSA, and FOSE could be detected in and around landfills and wastewater treatment plants. Concentrations at landfills could reach up to 17 ng/m³ (8:2 FTOH) for individual substances and up to 12 ng/m³ (6:2 FTOH) at wastewater treatment plants in this context (Ahrens et al., 2011). However, both values are well below the TRGS 900 limit of 0.01 mg/m³ for PFOS (since no limit for FTOH is available, the limit of PFOS is used as a comparative value). The highest PFOS (171 pg/m³) and PFOA (47.3 pg/m³) concentrations were measured in a wastewater treatment plant, but these values are also well below the TRGS 900 limit value. To minimize risks (e.g., direct discharge of PFAS from the landfill to the environment, inhalation of PFAS at the landfill), regular maintenance of leachate collection systems is recommended. Precipitation and adsorption are effective methods to remove PFAS from the liquid phase and subsequently destroy them thermally (D. T. Held & Reinhard, 2020).

Waste treatment	Quantity of the substance (kg/a)	Possible risks related to the PFAS content
		In general, paper waste contaminated with PFAS should not be landfilled because the PFAS are not destroyed. Thermal waste treatment is the recommended form of disposal.

3.5.4 Soils

Table 52: Environmental context of PFAS in the soils waste stream for the treatment routes of material recovery, thermal disposal, and landfill/other.

Waste treatment	Quantity of the substance (kg/a)	Possible risks related to the PFAS content
Material recovery	1,922.22	About 80 % of the generated soil waste is recycled, mostly by backfilling, i.e. suitable non-hazardous waste is used for recultivation of excavated areas or for landscaping. In early 2022, the BMUV published a guideline for PFAS assessment, listing limit values for PFAS in soils (BMUV, 2022b). Here, soils are divided into three categories depending on the PFAS concentration in a 2:1 shake eluate (a total of 7 PFAS are mentioned). (1) At low concentrations, the soils may be utilized without restriction. (2) At elevated concentrations, the contaminated soils may be used in open paving to a limited extent only in areas with already elevated PFAS levels. (3) In the case of soils with highly elevated PFAS concentrations, the soils may only be installed in technical structures with defined safety measures on a restricted basis. The aim of this restriction is to avoid groundwater contamination with PFAS. (BMUV, 2022b). In the eluate samples obtained from Rastatt, 3244 samples are below the concentrations of the first category, 284 are above the concentrations of the first category but below those of the second category, 770 are above the concentrations of the second category but below those of the third category, and 726 samples are above the limits of the third category (a total of 5004 samples). According to the guide, this means that about 70% of the samples can be used without restrictions. Generally, backfilling can be done with the same soil that was excavated or with imported material, depending on site requirements (Designing Buildings, 2022). If PFAS-contaminated soil material is used, the raw material may be mixed with PFAS. The excess soil material during construction activities is usually temporarily stored and later used for other purposes. In this way, PFAS-contaminated soil material can be transported and distributed from one site to another. The PFAS in the soil then mostly bind to the solid particles, but this process is reversible, which means that the PFAS are washed out and can enter groundwater, for example. They can also be taken up by plants and subsequently by humans and animals. For both processes (uptake and leaching) the soil type is of particular importance, as some soils bind PFAS better than others. However, there is a lack of concrete studies on this (Scheurer et al., 2021).

Waste treatment	Quantity of the substance (kg/a)	Possible risks related to the PFAS content
		During excavation and backfilling of soil material, PFAS, especially volatile PFAS such as FTOH, can pass into the gas phase and be inhaled. Likewise, skin contact may occur. Consideration might therefore be given to wearing appropriate protective equipment (e.g., protective mask and gloves) during such operations. Appropriate air measurements should be taken to determine the need for such protective measures. In general, it is recommended that PFAS-contaminated soils be remediated before they are reused. Procedures for remediation of contaminated soils can be carried out by in-situ and ex-situ remediation, including biological, chemical, physical or thermal soil remediation. However, due to its high chemical and thermal stability and extreme persistence, PFAS removal can be challenging. For example, special solutions, additives, or materials can be added to the soil to leach out or immobilize the PFAS (Bolan et al., 2021). None of these methods are available on a large scale or result in the complete destruction of PFAS. More common remediation methods include soil washing, immobilization of PFAS, and thermal desorption, but data on the effectiveness of these methods are often lacking, so remediation is currently focused on pump-and-treat and soil replacement as the main procedures (D. T. Held & Reinhard, 2020). PFAS in soils can be effectively destroyed thermally by high temperatures (>1,200 °C) (UBA, 2020b), however, chemical treatment processes can also destroy PFAS (Bolan et al., 2021). Biologically, PFAS cannot be destroyed, only a degradation of the precursors to the analogous perfluoro acids takes place.
Thermal treatment	0.55	Only a very small proportion of the soils generated as waste in Germany are thermally treated (0.03%), which corresponds to a PFAS load of only about 0.5 kg. The environmental context of waste incineration is described in chapter 3.5.1 described.
Deposit in landfills/other	337.60	In Germany, the landfilling of PFAS-containing soils is possible in principle (UBA, 2020b). Approx. 15% of the soil material generated as waste is deposited in landfills, which, according to the calculations in this project, corresponds to a load of approx. 337 kg PFAS. After being deposited in a landfill, (hazardous) substances can leach out due to leaching by rain. The rainwater eventually collects on the bottom of the landfill. In the Netherlands, leachate from landfills where contaminated soil material was deposited was analyzed for PFAS. Concentrations of individual substances of up to 16 µg/L (PFBS) were detected. (Rijkswaterstaat, 2020), illustrating that landfills are not a suitable method to effectively destroy PFAS. Perfluoroalkyl acids can also enter leachate through the decomposition of side-chain fluorinated polymers. These polymers have a carbon main chain and PFAS as a side chain, which are typically linked to the main chain via ester or amide bonds. These bonds are easily cleaved, allowing the side chain PFAS to slowly separate and leach out over time (OECD, 2022). Many modern landfills are equipped with collection systems to capture the leachate generated. This is then either treated on site or sent directly to a wastewater treatment plant, but as mentioned earlier, these are not capable of destroying PFAS

Waste treatment	Quantity of the substance (kg/a)	Possible risks related to the PFAS content
		(W. Zhang et al., 2022), nor is on-site treatment of landfill leachate. (Travar et al., 2020). In the U.S., it has been shown that WWTPs receiving leachate from landfills have up to three times higher PFAS concentrations in influent than WWTPs not receiving leachate. PFAS concentrations in the leachate were 10 times higher than in the influent of the WWTPs (Masoner et al., 2020). In this context, short-chain PFAS tend to accumulate in leachate, while long-chain PFAS are more likely to bind to solids (Knutsen et al., 2019). Therefore, both untreated and treated leachate pose a risk. For example, wastewater contaminated with PFAS can enter surface waters and eventually enter the food chain. Likewise, leachate can leach into the soils below the landfill and contaminate them with the pollutants it contains. Landfills and wastewater treatment plants can also emit PFAS to air, particularly volatile precursors (e.g., FTOH, FOSE, FOSA) that are commonly used in the textile industry (Ahrens et al., 2011; B. Wang et al., 2020). Once airborne, PFAS can travel long distances and deposit on the ground, which is why some PFAS, PFOS, PFOA, and PFHxS have been identified as persistent organic pollutants (POPs). Near their release, they can also be inhaled by on-site personnel. In Canada, FTOH, FOSA, and FOSE could be detected in and around landfills and wastewater treatment plants. Concentrations at landfills could reach as high as 17 ng/m³ (8:2 FTOH) for individual compounds and as low as 12 ng/m³ (6:2 FTOH) at wastewater treatment plants (Ahrens et al., 2011). However, both values are well below the TRGS 900 limit of 0.01 mg/m³ for PFOS (since no limit for FTOH is available, the limit of PFOS is used as a comparative value). The highest PFOS (171 pg/m³) and PFOA (47.3 pg/m³) concentrations were measured in a wastewater treatment plant, but these values are also far below the TRGS 900 limit value. To minimize risks (e.g., direct release of PFAS from the landfill to the environment), regular maintenance of leachate collection systems is recommended. Precipitation and adsorption are effective methods to remove PFAS from the liquid phase and subsequently destroy them thermally (D. T. Held & Reinhard, 2020). In general, PFAS-contaminated soils should not be landfilled because the PFAS are not destroyed (UBA, 2020b).thermal treatment or appropriate landfilling with appropriate safeguards is the recommended form of disposal. Likewise, underground disposal in suitable repositories can be considered, especially for highly contaminated soils.

3.6 Considerations on waste limits for PFAS

The methodology for deriving the waste limit values is described in chapter 2.9 is described. This methodology was developed for the derivation of POP waste limits, but can also be applied here in a modified form. However, some difficulties and challenges arise.

Selection of the PFAS to be investigated

A key challenge here is the question of which PFAS should be considered in the derivation. Since POP substances are often single substances or congeners, the required analysis is simplified, but PFAS are a complex group of substances with over 5,000 individual substances. Therefore, substances have to be selected against which the limit value can be measured.

For example, after PFOS was included in the Stockholm Convention, shorter-chain PFAS such as PFHxS were used as alternatives, which were not banned. Due to the similar properties of this substance to PFOS, PFHxS was also listed as a POP in 2021. Such substitution of a hazardous substance by a similarly hazardous substance is called "regrettable substitution" and should be avoided if possible.

The danger of such a substitution exists in particular with PFAS, since structural analogues are very easy to produce. So-called telomer substances are often used, such as 6:2 FTS, which has the same carbon skeleton as PFOS. However, the two carbon atoms close to the acid are not substituted with fluorine, but still have their hydrogen atoms. Such substances are precursors (or polyfluorinated compounds), since these two carbons can be degraded in nature, resulting in the analogous perfluoro(carbon)acid (in this case PFHxA).

Therefore, precursors should also be included in a waste management limit. If only perfluorocarboxylic and sulfonic acids are considered in the limit, similarly hazardous precursors will not be considered, thus underestimating the PFAS load and associated risk. However, since the number of possible precursor compounds is very large, it is unrealistic to measure all precursor substances individually.

Therefore, a method is needed by which the precursor substances can be included in the limit value observation without measuring them as individual substances. Currently, the most frequently used method for such a determination is the TOP assay. Here, the sample to be analyzed is oxidized by means of a peroxide, whereby it is assumed that all precursor substances are oxidized to the analogous perfluoro(carbon)acids (compare chapter 3.1.5). In reality, this is not always the case. For example, shorter-chain perfluorocarboxylic acids or precursor compounds that are not completely oxidized can also be formed (Al Amin et al., 2021). By a single substance determination of perfluorocarboxylic and sulfonic acids before and after the top assay, the loading of precursors can be estimated. It is assumed that perfluorosulfonic acids are not attacked in the TOP assay and are stable (Göckener et al., 2021). A conclusion on the exact precursor substances used is not possible.

However, no standard exists yet for the TOP assay. Göckener et al. (2022) was able to show that different laboratories determined different concentrations after the TOP assay, although all laboratories had examined the same sample. To ensure that the results of the required analyses are comparable, it is recommended to establish a standard for the TOP assay.

For the waste limit, easily detectable substances could be used as a basis. For example, DIN 38407-42 and DIN 38414-14 analyze the following PFAS:

- PFBA
- PFPeA

- PFHxA
- PFHpA
- PFOA
- PFNA
- PFDA
- PFBS
- PFHxS
- PFHpS
- PFOS
- 6:2 FTSA (H4PFOS)
- PFOSA

It is proposed to expand the selection of perfluorosulfonic acids to match the carboxylic acids so that the C4-C10 acids of both substance groups are considered. This results in a number of 14 PFAS to be measured:

- C4-C10 perfluorocarboxylic acids
- C4-C10 perfluorosulfonic acids

These 14 substances should be measured before and after a TOP assay to determine the waste limit relevant concentrations of the waste under consideration. Since 6:2 FTSA and PFOSA are degraded to perfluorocarboxylic acids in the TOP assay, they should not be included in the analyses.

Derivation of the limit values

Another challenge in adapting the POP system is the derivation of the limit value. Often only one substance is considered, resulting in only one value for each criterion. However, since 14 individual PFAS are proposed to be measured here, 14 individual limit values would also have to be developed, which would be summed up at the end. However, not enough data could be identified for each substance. In particular, for environmental risks, there is a lack of meaningful data to identify an upper bound criterion.

For the further procedure, a possible limit value is therefore derived for PFOS and PFOA, since most data could be identified for these. It is assumed for the derivation of the aggregate limit value that the other carboxylic and sulfonic acids behave similarly and thus the same limit value as for PFOS and PFOA is applied. For example, if a limit of 1 mg/kg is derived for PFOS, this would result in a limit for the sum of the 7 perfluorosulfonic acids of 7 mg/kg (analogous approach to REACH restrictions, see e.g. cumulative limit for the long-chain perfluorocarboxylic acids (entry 68 in Annex XVII of REACH)). The same applies to PFOA and the perfluorocarboxylic acids.

For further derivations in the future, more data are needed on the individual perfluoroalkyl acids, especially on the environmental effects of these substances, so that a separate limit value can be derived for each individual substance.

TOP Assay

The project team proposes to include the TOP assay in the consideration. Therefore, data for such measurements are needed. However, since this is a relatively new procedure, only very few data on TOP assay measurements could be identified so far (see chapter 3.1.5).

For example Norwegian Environment Agency (2021) detected up to 52-fold higher concentrations for PFBA in carpet samples after a TOP assay. In general, concentrations of perfluorocarboxylic acids were 15-fold higher after the TOP assay than before the TOP assay, suggesting a high load of precursors.

For the soil readings from Rastatt, TOP assay measurements were also carried out from 2019. Here, a maximum 16-fold higher concentration of the sum of perfluorocarboxylic and sulfonic acids (C4-C14 PFCA and C4-C10 PFSA) could be detected. The average increases were around a factor of 4, but only a sum value was given for the measurements after the TOP assay, which means that it cannot be concluded which perfluoro acids were formed.

For an accurate derivation of a limit value, sufficient measurements must be available for the waste stream under consideration before and after the TOP assay and for each of the individual substances under consideration. Thus, an average concentration increase after the TOP assay could be determined for each individual substance, with which the average load of the waste stream with precursor substances could be estimated. This would provide valuable data when deriving the background load.

In the absence of TOP assay data for this project, the precursors were "computationally oxidized." It was assumed that each precursor compound is completely oxidized to the analogous perfluorocarboxylic acid. By-products or incomplete oxidation as well as perfluorocarbon chain breakup were not considered. The following Table 53 shows the precursor compounds and analogous perfluorocarboxylic acids found:

Table 53: Precursors and associated perfluorocarboxylic acids identified in this project according to the TOP assay.

Forerunner	Perfluorocarboxylic acid(s) according to the TOP assay
PFOSA	PFOA
HPFHpA	It is assumed that this substance is not oxidized
H2PFDA	PFNA
H4PFUnDA	PFNA
4:2 FTS	PFBA
6:2 FTS	PFHxA
8:2 FTS	PFOA
FOSAA	PFOA
6:2 diPAP	PFHxA
8:2 diPAP	PFOA
diSAmPAP	PFOA
SAmPAP	PFOA

Forerunner	Perfluorocarboxylic acid(s) according to the TOP assay
H-DONA/ ADONA	Perfluoro-3-methoxypropanoic acid (PFMOPrA; CAS 377-73-1). (C. Zhang et al., 2019)
N-MeFOSAA	PFOA
N-EtFOSAA	PFOA
6:2 PAP	PFHxA
8:2 PAP	PFOA
PFECHS	It is assumed that this substance is not oxidized
7H-PFHpA	It is assumed that this substance is not oxidized
EtFOSAA	PFOA
N-MeFOSA	PFOA
N-EtFOSA	PFOA
9Cl-PF3ONS	Is not oxidized (C. Zhang et al., 2019)
11Cl-PF3OUdS	It is assumed that this substance is not oxidized
HFPO-DA	Is not oxidized (C. Zhang et al., 2019)
10:2 diPAP	PFDA
6:2/8:2 diPAP	PFHxA, PFOA
8:2/10:2 di-PAP	PFOA, PFDA

Using the molar mass of the precursors and the perfluorocarboxylic acids, the (theoretical) concentrations were calculated according to the TOP assay.

Due to the complexity of deriving the waste limit values and the partially inadequate data situation, the limit value is derived using the soil data as an example, since by far the most data are available for this waste stream. However, data for the other three waste streams are also presented, where identified.

The average concentrations for PFAS in soils after and before the computed TOP assay, can be found in Table 54. For further consideration, the concentrations after the calculated TOP assay were used to include the precursor substances.

Table 54: Average PFAS concentrations in soils before and after the calculated TOP assay. All values in [µg/kg].

Substance	Mean value	Median	Mean value after TOP assay	Median after TOP assay
∑PFCA	11.90	0.14	13.59	0.14
∑PFSA	3.50	0.00	3.50	0.00
∑PFPA	0.00	0.00	0.00	0.00
∑Precursors	2.14	0.00	0.00	0.00

Substance	Mean value	Median	Mean value after TOP assay	Median after TOP assay
∑PFAS	**17.53**	**0.21**	**17.55**	**0.21**

It can be seen that the mean value for the perfluorocarboxylic acids is higher after the top assay than before and that the precursors have a concentration of 0 µg/kg because they were oxidized. The concentrations for the perfluorosulfonic acids are unchanged. It should be mentioned that these concentrations probably underestimate the actual load, since on average concentrations higher by a factor of 4 were detected after the TOP assay (see above).

In addition, per- and polyfluorinated polymers, especially side-chain fluorinated polymers, are often not considered in the TOP assay. Extraction with methanol is usually not able to dissolve these polymers, which means that they are not oxidized in the TOP assay. As a result, they are often not analyzed as well, which can lead to underestimation of the actual load of PFAS even with TOP assay. To overcome this problem, the direct TOP assay (dTOP) has recently been applied in which the extraction with methanol is not performed, but the sample is directly added with the oxidant. For example. Göckener et al. (2021) have successfully applied this method to the analysis of soil materials.

3.6.1 Evaluation of the lower and upper boundary criteria for the waste stream of soils.

3.6.1.1 Lower limiting criteria

3.6.1.1.1 Analysis method

The limits of quantification of the PFAS analytical methods were largely extracted from the publications analyzed. However, since these did not contain costs, a cost request was made to an analytically specialized research institute.

Table 55: Limits of quantification and costs for the analysis of PFAS in the 4 selected waste streams.

Product	Limit of determination	Costs	Sources
Textiles	0.5 µg/kg		(Kotthoff et al., 2015).
	0.1-10 µg/kg (perfluoro acids) 40-200 µg/kg (precursors)		(Janousek, Lebertz, et al., 2019).
	0.5 µg/kg		(Müller & Schlummer, 2011)
	1 µg/kg	150-300 €	(Research Institute, 2022)f
Sewage sludge/soil (as solid)	10 µg/kg dry matter		(DIN 38414-14, 2011)
	1 µg/kg		(Stahl et al., 2018)
	1 µg/kg		(Toshovski et al., 2020).
	2-3 µg/kg		(Saxony-Anhalt, 2008)

Product	Limit of determination	Costs	Sources
	0.5 µg/kg (Target analysis) 2.5 µg/kg (dTOP)		(Göckener et al., 2021).
	1 µg/kg	150-300 €	(Research Institute, 2022)
Paper	0.3-1.7 mg/kg (perfluoro acids), 0.8-260 mg/kg (precursors)		(Strakova et al., 2021)
	0.5 µg/kg		(Müller & Schlummer, 2011)
	0.5 µg/kg		(Kotthoff et al., 2015).
	1 µg/kg	150-300 €	(Research Institute, 2022)

It can be seen that the typical limit of quantification for PFAS in soils varies between 0.5-10 µg/kg, with most limits of quantification found to be 1 µg/kg. Furthermore, the limits of quantification in individual publications vary somewhat depending on the compound considered, but not on a significant scale (mostly a difference ≤1 µg/kg). Therefore, a limit of quantification of 1 µg/kg is used for the perfluoroalkyl acids for the further determination of the waste limit.

The typical costs for such a determination vary between 150-300€ and are largely dependent on the number of PFAS analyzed. Since this cost range is below 500€, these analyses are considered economically justifiable by the project team (see also chapter 2.9.1.1 and (Potrykus et al., 2015).).

Table 56: Limit values for PFOS and PFOA based on typical detection limits for PFAS in soils.

	PFOS	PFOA
Limit value [µg/kg]	1	1

3.6.1.1.2 Background contamination

Some PFAS are ubiquitous substances that can be transported over long distances, which means that three PFAS (PFOS; PFOA and PFHxA) are already listed as POPs. Therefore, many soils are contaminated with these PFAS even though they are not near a point source.

Brusseau et al. (2020) summarizes literature data on PFAS concentrations in soils from various publications (background and potentially contaminated), which are presented in the following Table 57:

Table 57: Summary of PFAS concentrations in soils from. Brusseau et al (2020). All values in [µg/kg]

Thumb	Minimum PFAS concentration	Maximum PFAS concentration	Number of PFAS measured	Maximum PFOA concentration	Maximum PFOS concentration	Country
2010/2013	0.3	3.9	12	3.4	1.7	Korea

Thumb	Minimum PFAS concentration	Maximum PFAS concentration	Number of PFAS measured	Maximum PFOA concentration	Maximum PFOS concentration	Country
2010	141	237	15	47.5	10.4	China
2010/2019	0.7	22	9	2	20	China
2011	<0.3	9.4	9	0.5	2.4	China
2011	0.1	8.5	12	2.8	0.9	China
2011	<0.1	1.7	12	0.5	0.7	China
2012	1.3	11	12	0.9	9.4	China
2012	<0.5	150	13	32	10	USA. China. Japan. Norway. Greece. Mexico
2012	<0.1	5.8	18	1.5	5.4	Tierra del Fuego & Antarctica
2013	<0.1	1.8	22	0.3	0.4	China
2013	<0.1	4.1	16	0.2	0.2	China
2014	<0.05	1.6	2	0.7	0.9	Korea
2014	<0.1	1.8	16	0.3	0.1	Nepal
2015	6	135	2	28	126	USA
2014/2015	0.7	28.8	11	9	0.3	China
2015	0.04	3.6	13	2.3	1.9	China
2016	0.3	5	17	25	2	China
2016	0.05	15	32	3.4	3.1	Multiple
2016	<0.5	71	12	33	59	USA
2016	0.1	4	21	4.2	2.7	China
2017	<0.05	3.6	2	1.8	2.7	Korea
2017	1.9	126	12	123.6	2.7	China
2018	3	64	12	5	4.2	China
2018	1.3	30	7	3	12	USA
2018	<0.02	20	28	0.6	1.7	Sweden
2018	<0.2	5.1	6	0.5	3.1	USA
2018	0.4	174	17	3.3	162	Norway
2018	1.7	7.9	26	0.9	3	Uganda
2018	<0.001	0.01	2	0.0	0.0	China

Thumb	Minimum PFAS concentration	Maximum PFAS concentration	Number of PFAS measured	Maximum PFOA concentration	Maximum PFOS concentration	Country
2019	0.5	35	17	4.9	9.7	USA
2019	0.6	5.1	17	2.7	0.1	China
2019	0.8	53	15	3.7	37	Belgium
2019	0.1	13.9	17	2.1	0.7	Korea
2019	NR	64.7	21	16.6	2.8	China
2019	2.5	8.8	19	0.3	1	Korea
2019	<0.05	7.1	14	0.0	7.1	Norway
2019	4.2	49	12	23	1.2	China
Mean value				**8.2**	**2.8**	
Median				**1.7**	**1.9**	
Maximum				**123.6**	**162.0**	

Since the values come from different regions (urban, agricultural, gardens, schoolyards, industrial, etc.) they clearly show that these substances can be detected almost everywhere, but some of the samples here do not necessarily represent background contamination, since they may have been measured near point sources. Maximum PFOA and PFOS concentrations of 123.6 µg/kg and 162 µg/kg could be identified.

Data on PFAS contamination could also be extracted from soil findings in North Rhine-Westphalia (LANUV, 2022b). Non-polluted areas (i.e. background soils) were specifically analyzed for PFAS. Maximum PFOS and PFOA concentrations of 12.44 µg/kg and 10.36 µg/kg, respectively, were found. The significance of these data is considered to be better than that reported by Brusseau et al. (2020) because, on the one hand, the background load was specifically measured and, on the other hand, the data originate from Germany.

Finally, the data analyzed in this project were also evaluated. Since the majority of the data came from the district of Rastatt and specifically contaminated areas were measured, high concentrations could be detected. In addition, the concentration of PFOA was again increased by the computational TOP assay. Maximum concentrations of PFOS and PFOA of 1,040 µg/kg and 1,666.5 µg/kg respectively could be determined.

In general, the project team assigns the highest representativeness for background pollution in Germany to the data from North Rhine-Westphalia, which is why these values are considered for the further procedure.

As described in chapter 2.9.1.2 the highest identified background load is multiplied by 10 to obtain lower limit values for the background load. This results in the following limit values (Table 58) for PFOS and PFOA:

Table 58: Limit values for PFOS and PFOA based on the background contamination assumed here in Germany

	PFOS	PFOA
Limit value [µg/kg]	124.4	103.6

3.6.1.1.3 Disposal and recovery capacities

For the destruction of PFAS, thermal treatment is the most suitable method, so thermal waste treatment is considered in particular. The following Table 59 shows the maximum and used thermal waste treatment capacities in Germany.

Table 59: Maximum and used waste incineration capacities in Germany

Source	Total capacity [t/a]	Amount of waste incinerated [t].	Capacity of waste incineration plants (year, number) [t/a].	Capacity of refuse-derived fuel power plants (year, number) [t/a].	Hazardous waste incineration plant capacity (year, number) [t/a].
(Flamme et al., 2018).	28,579,612 (26,945,532 without hazardous waste)	27,620,628 (26,269,135 without hazardous waste)	20,634,782 (2016, 66)	6,310,750 (2016, 32)	1,634,080 (2016, 31)
(Destatis, 2022)		28,002,600 (26,668,000 without hazardous waste)	- (2020, 84)	- (2020, 35)	- (2020, 29)
(NABU, 2022)	26,500,000	26,300,000	20,600,000 (2022, 66)	5,800,000 (2022, 32)	-
(UBA, 2016)	25,800,000		19,600,000 (2016, 68)	4,700,000 (2016, 30)	1,500,000 (2016, 30)
(Pohl et al., 2022)	26,982,000 (2020)	26,270,000	20,661,000 (2020, 66)	6,321,000 (2020, 34)	

In general, there are different types of waste incineration plants in Germany. Household waste incinerators burn the waste at a minimum of 850 °C. There are about 66 of these in Germany. Furthermore, waste with a high calorific value can be incinerated in refuse-derived fuel power plants. There are about 34 such power plants in Germany. Finally, there are hazardous waste incineration plants, which burn waste at particularly high temperatures above 1,100 °C. There are about 30 such plants in Germany. There are around 30 such plants in Germany.

A total of approx. 27,000,000 tons of waste can be incinerated annually in Germany. Of this capacity, approx. 26,200,000 are utilized, which means that an additional approx. 800,000 tons can be incinerated (see Table 59 and the values from (Flamme et al., 2018 NABU, 2022 Pohl et al.)).

In addition, soil material can be treated in other ways, such as thermal desorption or soil washing. In general, a distinction is made between thermal, chemical-physical and biological processes. Frauenstein & Mahrle (2020) give the following capacities for stationary soil treatment in Germany:

Table 60: Capacities for stationary soil treatment in Germany from Frauenstein & Mahrle (2020).

Procedure	Thermal process	Chemical-physical processes	Biological methods
Approved plant capacity (t/a)	312,000	2,270,000	3,139,000
Plant utilization (%)	66.83	45.17	47.00
Mass throughput (t/a) based on specific plant capacity utilization (Basis 2017-2019)	208,510	1,025,360	1,475,330
Available capacities (t/a)	103,490	1,244,640	1,663,670

Overall, this results in an overcapacity in Germany of approximately 3 million tons per year for soil treatment (as of 2020). More recent data could not be identified.

However, not all processes are suitable to remove/destroy PFAS from soil. For example, biological processes are only able to degrade non-fluorinated parts of a molecule, while perfluorinated compounds cannot be destroyed/degraded (Bolan et al., 2021). Thermal processes are generally suitable to destroy PFAS especially at temperatures >850 °C (see chapter 3.5), however, studies indicate that temperatures as low as 550 °C are sufficient to desorb 71-99% of the PFAS investigated (Bolan et al., 2021). In terms of chemical-physical processes, there are many different methods that are used. Some of the methods are unsuitable to destroy PFAS (e.g., chlorination, chloramination, chemical oxidation, and UV treatment), but PFAS could be successfully destroyed in the laboratory by, for example, electrocatalytic oxidation and persulfate oxidation (Bolan et al., 2021).

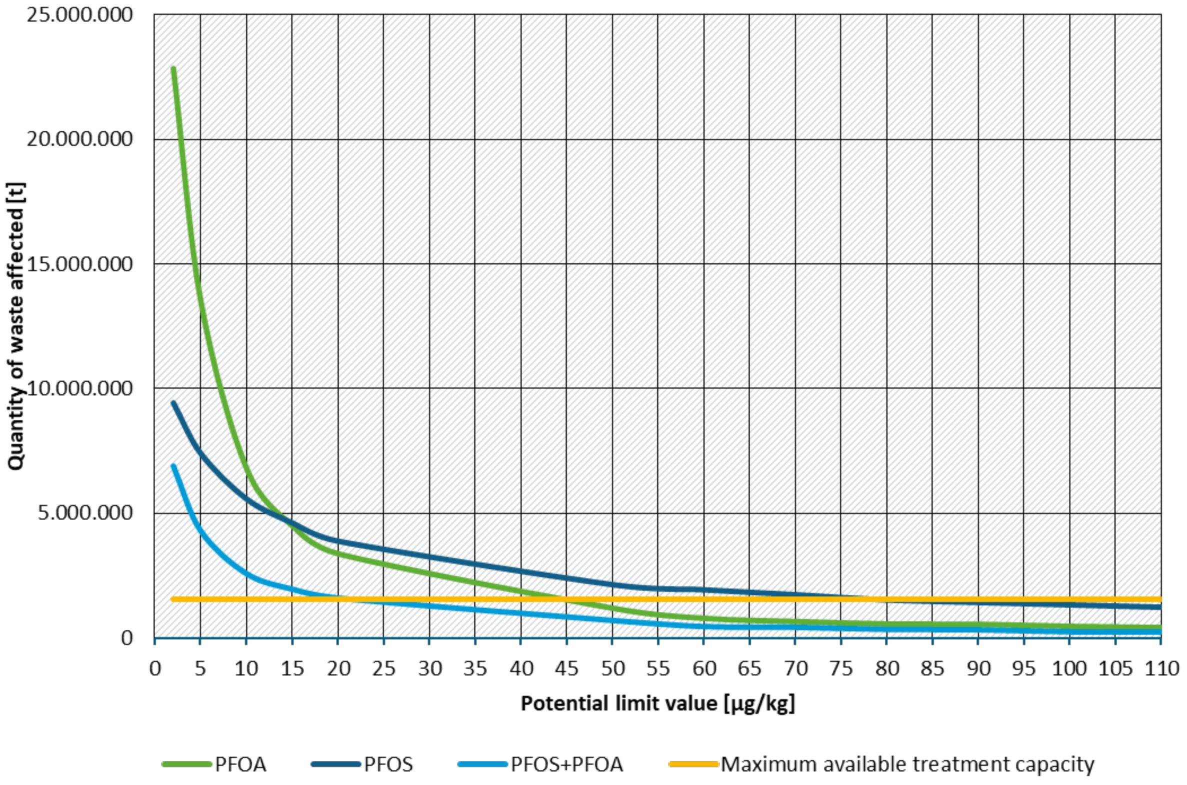
In the publication by Frauenstein & Mahrle (2020) it is mentioned that only one thermal plant, 6 soil washing plants and two biological plants are approved to treat PFAS and accept these soils. The authors do not mention capacities of these plants and assume that in the future, due to the amount of PFAS contaminated soils, there will be a capacity bottleneck at the thermal soil treatment plants. In general, the exact location of the treatment capacities of PFAS-containing soils in Germany is unclear. Better data is needed for an exact calculation.

For the further procedure an optimistic approach is chosen and it is assumed that 50% of the thermal and chemical-physical plants in Germany are able to destroy PFAS effectively. Although only 9 plants are currently approved for the treatment of PFAS, it is assumed that more plants will be approved in the future as more PFAS-contaminated soils are treated.

This results in an excess capacity for soil treatment in Germany of ~674,000 tons per year. Together with the 800,000 tons from the waste incineration plants, an additional ~1,474,000 tons of soils can be treated in Germany that the PFAS contained therein are destroyed or removed.

A total of about 129 million tons of soil material was generated as waste in Germany in 2018, with only about 30,000 tons being thermally treated (cf. Table 43). From the obtained and researched data on PFAS in soils, theoretical limit values can now be set to estimate the potentially contaminated amount of soil material. For this purpose, a potential limit value is set and it is calculated how many samples lie above this limit value. If the percentage is set in relation to the number of total samples (e.g. 35% of the samples are above this limit value), it can be calculated with the waste data from Table 43 how much soil material would thus have to be disposed of properly. This is calculated separately for PFOS and PFOA and shown in Figure 16. Also shown is the line when the concentrations for PFOS and PFOA in a sample are both above the limit. The maximum treatment capacity of approximately 1,474,000 tons is shown as a yellow line:

Figure 16: Potential limits for PFOS and PFOA and the amount of waste for soils affected by them.



Source: Own representation

Within Figure 16 it can be seen that the limit at which the treatment capacity is exceeded is lowest for the sum of PFOS and PFOA, since both substances must be above the potential limit. This point is reached at ~22 µg/kg. For PFOA, the treatment capacity is already exceeded at ~46 µg/kg and for PFOS at ~82 µg/kg. The limits should therefore be above these two values respectively.

Table 61: Limit values for PFOS and PFOA based on the quantities of waste to be disposed of and the maximum treatment capacity.

	PFOS	PFOA
Limit value [µg/kg]	82	46

3.6.1.1.4 Economic impact

This criterion analyzes the economic costs incurred when a waste may no longer be treated as before because PFAS concentrations are above the limit.

For this purpose, the current costs for recycling the soils are first calculated based on the researched costs in Table 17 and the treatment routes from Table 43.

Table 62: Soil disposal methods and associated costs (rounded).

Waste total	Elimination			Recovery		
	Landfill	Incineration	Other	Energy recovery	Recycling	Backfilling
128,858,796	19,193,317	20,697	52,310	10,763	20,830,599	88,751,111
Cost (rounded) [€]						
7,269,707,000	2,878,998,000	3,518,000	2,092,000	1,830,000	833,224,000	3,550,044,000

The category "other" includes e.g. processes such as biological treatment of soils, injection of pumpable wastes into boreholes, surface application of wastes as well as discharge into water bodies such as lakes and seas (it cannot be identified from the data which process is allowed and also applied). Since the Eurostat data do not allow further conclusions on the exact treatment method, it is assumed that the soils are either biologically treated or surface applied. Therefore, the costs for recycling were assumed (40€/ton, see chapter 2.9.1.4).

This results in total costs of ~7.3 billion euros for the current disposal of soils in Germany.

In general, it is assumed that highly contaminated soils (e.g., from the vicinity of point sources) will be decontaminated or thermally treated. For the further procedure, a worst-case scenario is therefore assumed, in which the soils that have PFAS concentrations above the limit value are thermally treated instead of being recycled or backfilled. This results in a cost difference of 130€ per ton of waste to be disposed. Since no costs for soil remediation could be determined, a cost difference of 130€ per ton is also applied for this.

For the calculation, potential limit values for PFOS and PFOA are applied analogously to the procedure for the disposal methods and the associated waste quantities to be disposed of are calculated (see chapter 3.6.1.1.3). The waste quantities are multiplied by the assumed 130€/ton to calculate the theoretical costs.

This results in the following costs for PFOS and PFOA:

Table 63: Incurring disposal costs for soil material for potential limit values for PFOS and PFOA (rounded)

PFOS			PFOA		
Limit value [µg/kg]	Quantity of waste affected [t]	Additional costs (rounded) [€]	Limit value [µg/kg]	Quantity of waste affected [t]	Additional costs (rounded) [€]
100	1,333,807	173,395,000	50	1,203,680	156,478,000
110	1,236,212	160,708,000	60	797,031	103,614,000
120	1,057,286	137,447,000	70	666,904	86,697,000
150	748,233	97,270,000	100	471,712	61,323,000
200	487,978	63,437,000	120	390,383	50,750,000
230	406,649	52,864,000	150	325,319	42,291,000
240	357,851	46,521,000	200	276,521	35,948,000

Based on these data, it can be seen that if already 1% of the soil material (~1,200,000 tons) is above the limit, costs of more than 150 million euros are incurred. This is already the case at 50 µg/kg for PFOA and 110 µg/kg for PFOS. In general, a cost increase of 1% is considered to be still acceptable (see chapter 2.9.1.4). This corresponds to a value of 73 million euros and is the case at ~185 µg/kg for PFOS and at ~77 µg/kg for PFOA. The potential limits should therefore be above these values.

Table 64: Limit values for PFOS and PFOA based on economic impact.

	PFOS	PFOA
Limit value [µg/kg]	185	77

3.6.1.2 Upper limiting criteria

3.6.1.2.1 Limits

Various limit values already apply to various PFAS, which are shown in the following list:

Table 65: Current applicable limit values for PFAS in Germany.

Substance	Law	Limit value
PFOS	POP-VO	50 mg/kg (waste) 10 mg/kg (UTC)
PFOA + related compounds	POP-VO	0.025 mg/kg (UTC) for PFOA and its salts 1 mg/kg (waste) for PFOA and its salts 0.025 mg/kg (UTC) for related compounds 40 mg/kg (waste) for related compounds
PFHxS + related compounds	POP-VO	1 mg/kg (waste) for PFHxS and its salts 40 mg/kg (waste) for related compounds
C9-14 PFCA + related compounds	REACH-VO	0.025 mg/kg (UTC) for the sum of C9-C14 PFCAs. 0.26 mg/kg for the sum of related compounds
SUM PFOS + PFOA	Fertilizer Ordinance	0.1 mg/kg dm
PFAS total	EU Drinking Water Directive*	0.5 µg/l
20 PFAS	EU Drinking Water Directive (mentioned in Annex III (3))*.	0.1 µg/l

* The EU Drinking Water Directive is not legally binding in Germany due to its nature as a directive. It is transposed into German law by the Drinking Water Ordinance, but currently (as of January 2023) no PFAS are listed there.

The limit values in the POP Ordinance are particularly relevant for the soil material. For PFOS, a limit value of 50 mg/kg in waste and 10 mg/kg as a trace contaminant in products applies.

For PFOA and PFHxS, a waste limit of 1 mg/kg applies for the substance itself and its salts, and a limit of 40 mg/kg for the related compounds (see chapter 1.2.3.2). For PFOA as a trace contaminant, a limit value of 0.025 mg/kg (or 1 mg/kg for the related compounds) also applies.

The waste limit values should therefore be below the waste limit values of the POP Regulation.

Furthermore, there are limit values for long-chain perfluorocarboxylic acids (C9-C14), but these are not relevant for PFOS and PFOA.

Other current applicable limit values can also be found in the Table 65.

Thus, the following limit values for PFOS, PFHxS and PFOA result based on the currently applicable limit values (Table 66).

Table 66: Limit values for PFOS and PFOA based on the currently applicable waste limit values in Annex IV of the POPs Ordinance.

	PFOS	PFOA	PFHxS
Limit value [mg/kg]	10	1	1

3.6.1.2.2 Potential environmental and health impacts

Finally, the possible environmental and health effects are also considered. To avoid a risk, the concentrations in the environment should be below ecological limits.

In order to be able to estimate the potential effects, PNEC values were researched, which are shown in the following Table 67 are presented. The two publications by Gałęzowska et al. (2020) and Meng et al. (2021) and supplemented with further PNEC values found in other publications.

Table 67: PNEC value found in the literature.

Substance	Salt water (µg/L)	Freshwater (µg/L)	Soil (µg/kg DM)	Sediment (µg/kg DM)
PFBA	11 (Q. Wang et al., 2019a)[a]			241.3 (Gałęzowska et al., 2020)
PFPeA	3.2 (Hoke et al., 2012)[b]	32 (Hoke et al., 2012)		227.2 (Gałęzowska et al., 2020)
PFHxA	9.7 (Hoke et al., 2012)[b]	97 (Hoke et al., 2012)		2,825.3 (Gałęzowska et al., 2020)
PFHpA				1,002.6 (Gałęzowska et al., 2020)
PFOA	125 (Colombo et al., 2008).[c] 3 (Q. Wang et al., 2019b)[a]	20 (pelagic organisms) (Ankley et al., 2021) 1.428 (Phung et al., 2021) 100 (Hoke et al., 2012)	86 (Zhao et al., 2013).[d]	86 (Zhao et al., 2013)d 814.3 (Gałęzowska et al., 2020)

Substance	Salt water (µg/L)	Freshwater (µg/L)	Soil (µg/kg DM)	Sediment (µg/kg DM)
PFNA	10 (Hoke et al., 2012)[b]	1,000 (Phung et al., 2021) 100 (Hoke et al., 2012)		255618.8 (Gałęzowska et al., 2020)
PFDA	1.1 (Hoke et al., 2012)[b]	0.045 (Phung et al., 2021) 11 (Hoke et al., 2012)		514.8 (Gałęzowska et al., 2020)
PFUdA		0.008 (Phung et al., 2021)		410.7 (Gałęzowska et al., 2020)
PFDoA		0.001 (Phung et al., 2021)		320.2 (Gałęzowska et al., 2020)
PFTrDA				10,644 (Gałęzowska et al., 2020)
PFTeDA				38,325 (Gałęzowska et al., 2020)
PFHxDA				716061 (Gałęzowska et al., 2020).
PFODA				26,156,990 (Gałęzowska et al., 2020)
PFBS	37 (Q. Wang et al., 2019a)[d]			1,723 (Gałęzowska et al., 2020)
PFHxS		250 (Phung et al., 2021)		38,8703 (Gałęzowska et al., 2020)
PFOS	2.5 (Zhao et al., 2013)[d] 0.21 (Q. Wang et al., 2019a)[a]	0.61 (Phung et al., 2021)	373 (Footitt et al., 2004)[f] 4.9 (Zhao et al., 2013)[d]	37.3 (Footitt et al., 2004)f 0.49 (Zhao et al., 2013)d
PFDS				1,203 (Gałęzowska et al., 2020)

Species:

a: *Brachionus calyciflorus*

b: *Daphnia magna*, *Pseudokirchneriella subcapitata*, *Oncorhynchus mykiss,* and *Pimephales promelas.*

c: *Pseudokirchneriella subcapitata*, *Daphnia magna*, and *Oncorhynchus mykiss*.

d: *Mysidopsis bahia.*

e: *Scenedesmus obliquus*

f: *Eisenia foetida*

Since excavated soil is considered as a waste stream, the PNEC values for PFOS and PFOA in soil can be used directly. The precautionary principle should be applied and the lowest value considered in each case. Ideally, PNEC values for humans, or for the most sensitive organism, should be used for limit derivation. However, these could not be identified, so the lowest existing PNEC values are used.

For PFOS and PFOA, this is 4.9 µg/kg DM and 86 µg/kg DM, respectively (cf. Table 67), but little data on PNEC values in soils could be found. To better derive a waste limit for PFAS total in the future, more PNEC values for PFAS in soils are needed. This is also applicable to the waste stream of sewage sludges, as some of these are also directly applied to arable soils and can thus directly pose an environmental risk.

For the other waste streams (textiles, paper), the lowest PNEC values found in each case can be multiplied by a factor of 10,000. The background for this is the assumption that suitable recycling leads to the concentration in the waste appearing in the environment at a maximum of one 10-thousandth (Potrykus et al., 2015). This means that not every PFAS molecule present in textile waste will be released into the environment. However, this is not the case for soils and sewage sludges, which are applied directly to soils and thus all PFAS are found in the soil. Thus, this factor does not apply here.

Thus, the following upper limits for PFOS and PFOA result based on the PNEC values found for soils:

Table 68: Limit values for PFOS and PFOA based on the PNEC values found in soils

	PFOS	PFOA
Limit value [µg/kg DM]	4.9	86

3.6.2 Conclusion for the waste limits

For the waste stream of waste soils, the lower and upper limiting criteria were determined for PFOS and PFOA, respectively, which are presented in the following Table 69 summarized below:

Table 69: Results for the lower and upper limiting criteria for PFOS and PFOA in soils. All values in [µg/kg].

Evaluation criterion	PFOS	PFOA
Lower limiting criteria		
Analysis method	1	1
Background contamination	124,4	103,6
Disposal and recovery capacities	82	46
Economic impact	185	77
Upper limiting criteria		
Limits	10,000	1,000
Potential environmental and health impacts	4.9	86

Based on this representation, it can be seen that the limit value should be above 185 µg/kg for PFOS and 103.6 µg/kg for PFOA. The greatest limitation of the lower assessment criteria for PFOS is based on the economic consequences, which are based on a worst-case scenario. For PFOA, background contamination is the limiting factor. These potential limits are well below the existing waste limits in the POPs Regulation, but above the researched PNEC values for PFOS and PFOA in soils. This means that if the economic consequences as well as the background contamination are taken into account, the allowed PFOS and PFOA concentrations would be above the environmental limits. Thus, if such soils were reused, there would be a risk of negative environmental impacts.

Therefore, no meaningful limit value for these substances can be derived directly on the basis of the method used.

If the limit of 4.9 µg/kg and 86 µg/kg for PFOS and PFOA are applied, a waste volume of 7,644,993 tons and 553,042 tons, respectively, would be above the limits. This corresponds to 5.93% and 0.43% for PFOS and PFOA, respectively. This would result in additional disposal costs of ~1 billion euros and ~72 million euros, respectively, which within this project for PFOS, could no longer be considered economically justifiable, as they represent more than 1% of the previous disposal costs (see chapter 2.9.1.4). For comparison, it is assumed that soil remediation at Nuremberg Airport and Düsseldorf Airport would cost €10 million and €100 million, respectively (Nordic Council, 2019). Given this data, higher removal costs than assumed in this report, could be assumed to be economically justifiable. In this report here, 1% of the current treatment costs were considered economically justifiable (see chapter 2.9.1.4 and 3.6.1.1.4), but a higher value could be applied in the future.

In addition, in the case of PFOS, it would no longer be possible to remove all soils with PFOS concentrations above 4.9 µg/kg, as the available capacities would be exceeded. The limit value for PFOS and PFOA would be below the background level, which would probably affect too many soils.

The low environmental limits are related to the type of waste, in this case soils. Normally, it can be assumed that not all PFAS in a waste are released into the environment. Generally, this method of deriving limits assumes that only one 10,000th of the concentration of the waste will enter the environment if the waste is properly treated, which allows the environmental limits to be multiplied by a factor of 10,000. This is not the case for soils (and sewage sludge), since they are directly reused in the environment and the PFAS thus find a direct entry. Thus, the limits for soil wastes should be below the concentration at which adverse effects are expected. However, for the other wastes considered in this project (paper and textiles), this method could still be used since the safety factor can be applied. The limitations described in chapter 3.6 however, also apply to these two waste streams.

However, the data situation for PFAS PNEC values in soils is poor and current data could not be identified. Therefore, it is recommended to identify new PNEC values for PFAS in soils in order to better derive the waste limit values.

If the environmental limits are excluded, a limit value for PFOS should be above 185 µg/kg but below 10,000 µg/kg and for PFOA above 103.6 µg/kg but below 1,000 µg/kg.

Generally, in the last step, the determined concentrations for PFOS and PFOA would be multiplied by 7 to obtain the respective concentrations described in chapter 3.6 to include the perfluorosulfonic and carboxylic acids selected in chapter 3. Since no soil-relevant environmental thresholds could be identified for these substances, PFOS and PFOA were used as guide substences to simulate these substences. In the optimal case, a limit value should be derived individually for each PFAS compound to most effectively mitigate the risk and consequences posed by that compound.

For comparison, PFAS limits for soils were introduced in the Netherlands in 2018, with a limit of 0.1 µg/kg DM for each of PFOS and PFOA (Cousins et al., 2022). However, these levels were so low that 70% of construction activities involving excavated soil had to be stopped, whereupon the Dutch government set the levels higher (0.91 µg/kg DM for PFOS and 0.81 µg/kg DM for PFOA) (RIVM, 2019). Environmental risk data were included in the calculation for these limits, but no economic or waste consequences were included. The limits are therefore probably significantly lower than the limits calculated in this report.

3.7 Recommendations for limit values

3.7.1 Limits

In this project, an attempt was made to derive a limit value for certain PFAS for soils as an example, but no conclusive result could be achieved. The reason for this is that the values of the upper limiting criteria are below those of the lower limiting criteria (see chapter 3.6 and in particular 3.6.2). This is due to the fact that the researched PNEC values are in a very low concentration range (<100µg/kg for PFOS and PFOA). These values are usually multiplied by a safety factor of 10,000, since it can be assumed that with appropriate waste treatment only 1/10,000 of the hazardous substances will be released into the environment (Potrykus et al., 2015). However, this is not the case with soils, as they are directly reintroduced into the environment, which means that the factor does not apply here.

A similar situation prevails with sewage sludges, as these can be used in agriculture, which means that the PFAS can be released directly into the environment. Here, the safety factor is also not applied. However, a calculation for a limit value was not carried out within the framework of this project due to time constraints and should still be carried out in the future.

However, the papers and textiles are not affected by this limitation, since these wastes are not directly used in the environment. In general, the method described in this project (see chapter 2.9) can be applied to these two waste streams and it is assumed that a reasonable limit can be derived. However, this was also not carried out in this project due to time constraints and should therefore still be attempted in the future.

Since the methodology used here was developed for POP substances, some additional problems arise when it is applied to the substance group of PFAS. First, it should be mentioned that most POPs are single substances, which simplifies the data search. Some POP substances are comparatively manageable substance groups, such as polychlorinated biphenyls and short-chain chlorinated paraffins, but the number of individual substances is limited and within the substance group the properties are very similar, resulting in very similar physical, chemical and toxicological properties. This is not the case with PFAS, however, as this is a substance group with more than 4,500 individual substances. Some PFAS are similar and can be divided into various groups (*e.g.*, perfluorocarboxylic acids and perfluorosulfonic acids), but the diversity of PFAS is too great to consider them as one substance group in the context of this methodology. In particular, there are enormous differences in the chemical structure of PFAS among precursor compounds (see, for example, diPAPs and fluorotelomer substances). In this project, it was therefore decided to focus on 14 PFAS, namely the C4-C10 compounds of perfluorocarboxylic and -sulfonic acids, but no data could be found for many of these compounds (especially for the PFAS with an odd number of carbon atoms). It was therefore decided to use PFOS and PFOA (for each of the perfluorosulfonic and perfluorocarboxylic acids) as lead substances to derive a possible limit value. From a toxicological point of view, however, an individual limit value should be derived for each individual substance, since each substance behaves differently in the organism. This is particularly visible in the PNEC values, which can differ greatly for the respective PFAS (see Table 67). Ideally, the most sensitive organisms or humans (mostly DNEL values) should be considered for the PNEC values.

Due to the great diversity of PFAS, there is the additional problem that not every substance can be detected analytically individually, which means that the total PFAS load is often unknown. Especially the precursor compounds are very diverse. In order to be able to consider these compounds as well, it is therefore suggested to use the TOP assay (for TOP assay see chapter 3.1.5). In this assay, the precursor compounds are largely oxidized to the analogous perfluorocarboxylic acids. If these are measured before and after the TOP assay, the total PFAS load can be estimated. However, in this project, concrete TOP assay measurements were lacking. Therefore, the precursor compounds were oxidized by calculation. This results in a concentration increase of the perfluorocarboxylic acids by a factor of about 1, while in real samples a concentration increase of a factor 4-52 is common (compare chapter 3.6). It is therefore recommended to carry out the TOP assay for the measured samples for further limit values in order to obtain a realistic picture of the total PFAS load.

3.8 Recommendations for disposal routes

3.8.1 Textiles

Affected by waste

PFAS are used in textiles primarily for coating, to make the textiles water and dirt repellent. Therefore, mainly (outdoor) jackets and professional protective clothing (e.g. police, fire department, hospital, etc.) are affected. The PFAS can be applied to the textiles either directly during production of the products, or later by the consumer using impregnation sprays. Carpets are also often treated with PFAS, but other textiles may also be treated with PFAS such as bus and train seats and awnings. In general, PFAS can be washed out when textiles are washed, thereby contaminating other textiles (e.g. by washing in a washing machine).

An average PFAS concentration of ~390 µg/kg in textiles in Germany was calculated (compare chapter 3.3.1).

In Germany, old clothes, including outdoor jackets, are collected separately in the old clothes containers and mostly recycled (~90%). The remaining 10% is thermally/energetically recycled or otherwise disposed of. In addition, there are textiles that can be found in the residual and bulky waste, such as old carpets, chairs, cleaning rags and curtains. It is assumed that these will be used for energy recovery, as they are usually of poor quality due to their disposal in residual and bulky waste. Finally, the waste stream falling under CN code W076 is also considered. This contains industrial textile waste from fiber production, as well as textile packaging and leather waste, of which approx. 2/3 is recycled and approx. 1/3 is recovered for energy.

A differentiated presentation of the individual PFAS-relevant waste streams was not possible due to the lack of specific data on this (e.g. there was no data only for the waste stream of outdoor jackets, which meant that all used textiles in the used clothing containers were considered, although not all of them are treated with PFAS).

Treatment of the waste concerned, risks and recommendations

A total of ~2 million tons of textile waste was generated in the three waste streams mentioned above in Germany in 2018, corresponding to a total PFAS load of ~760 kg. Of this, ~70% was recycled, ~28% thermally treated and ~2% otherwise disposed.

Material recovery

During the material recycling of textiles, a carry-over of PFAS can generally occur. For example, through direct reuse of the textiles (e.g. outdoor jacket) or "downcycling" (e.g. cleaning rags are made from an old fabric), the PFAS remain in the material cycle and thus still pose a risk. If the textile fibers are recycled, the PFAS can be washed out and eventually end up in wastewater treatment plants. Likewise, volatile PFAS in particular can become airborne and thus pose a risk to personnel. However, measured PFAS concentrations in the air of carpet stores are in the ng/m^3 range and are thus far below typical PFAS limits in the mg/m^3 range. PFAS are not destroyed during material recycling.

As elevated concentrations of PFAS in air could occur in a recycling plant of appropriate textile waste, consideration could be given to taking personal protective measures to minimize the risk of PFAS and avoid their inhalation and possible skin contact. This could include, for example, wearing respirators and gloves. However, to determine the need for such protective measures, appropriate air measurements should first be taken.

It is recommended to identify products with a high probability of being significantly contaminated with PFAS, such as outdoor jackets, work clothing as well as outdoor materials such as awnings, at an early stage, to sort them out and send them for thermal recycling in order to avoid carry-over of PFAS.

Thermal treatment

In Europe, waste incineration plants must operate at a minimum temperature of 850°C and a residence time of at least 2 seconds. Since this is a minimum criterion, the actual temperatures are usually somewhat higher. Various scientific studies have shown that these conditions are sufficient to destroy PFAS to a large extent (see chapter 3.5). Insignificant amounts of PFAS could be detected in the ash. Measurements of fluorine gases in the exhaust air could not be identified. It is therefore recommended to measure such gases in the exhaust gas of waste incineration plants in Europe in order to be able to determine the corresponding destruction efficiency of the plants.

In general, thermal treatment is the recommended disposal method for PFAS-contaminated waste, as the PFAS are largely destroyed in this treatment method. It is therefore recommended that PFAS-contaminated fractions of textile waste should also be submitted to thermal waste treatment wherever possible.

Landfilling/other treatment

In Germany, textiles may not be landfilled because their loss on ignition is too high. A small proportion of textiles are disposed of elsewhere, but the exact treatment method is unknown. Therefore, no risks and recommendations can be derived.

3.8.2 Sewage sludge

Affected by waste

PFAS enter wastewater treatment plants through wastewater. Both industrial wastewater and municipal wastewater can be contaminated with PFAS. Various studies have investigated the PFAS contamination of sewage sludges in Germany, and PFAS were found in virtually all the sewage treatment plants sampled (see chapter 3.1.4.2 and 3.3.2). Similar results could be obtained in the samples investigated in this project (see chapter 3.2.2). Studies have shown that industrial sewage sludges often have higher PFAS concentrations than sewage sludges from municipal wastewater treatment plants, especially if they can be assigned to industries that use PFAS (e.g. paper or textile industry) (see also chapter 3.2.2 and 3.3.2).

It can therefore be assumed that the majority of industrial and municipal sewage sludge in Germany is contaminated with PFAS.

An average PFAS concentration of ~67 µg/kg in sewage sludge in Germany was calculated (cf. chapter 3.3.2). It should be mentioned that only municipal and mixed sewage treatment plants were considered in the calculation. Purely industrial sewage sludges were not considered. In order to obtain a complete picture of the PFAS load of sewage sludges in Germany, industrial sewage sludges should also be sampled and analyzed in the future.

Treatment of the waste concerned, risks and recommendations

A total of ~1.74 million tons of municipal sewage sludge was generated in Germany in 2020, corresponding to a total calculated PFAS load of ~117 kg. Of this, approx. 22% was sent for material recycling, approx. 77% was disposed of thermally and approx. 1% was disposed of directly in other ways.

Material recovery

In the case of material recycling of sewage sludge, the sludge is used as a soil-related fertilizer in agriculture or in landscaping measures. A part of it is also used for other material recycling (see chapter 3.4.2.1). PFAS are not destroyed during recycling. Therefore, PFAS are usually released directly into the environment during recycling, where they can spread further. They can be washed into the groundwater by rain.

In addition, volatile PFAS were detected in the air at the wastewater treatment plants, which generally poses a risk to humans. However, measured PFAS concentrations in the air of wastewater treatment plants are in the ng/m^3 range and are thus far below typical PFAS occupational exposure limits in the mg/m^3 range.

Limit values for PFAS in sewage sludge already exist in the Fertilizer Ordinance (100 µg/kg for the sum of PFOS and PFOA), which reduces the input of these substances into the environment. The number of substances to be measured for a suitable limit value and the level of the limit value should be discussed.

In general, it is recommended to determine the PFAS contamination of sewage sludges and then to subject the contaminated sludges to thermal treatment in order to destroy the PFAS as far as possible.

Thermal treatment

In Europe, sewage sludge incineration plants must operate at a minimum temperature of 850°C and a residence time of at least 2 seconds. Since this is a minimum criterion, the actual temperatures are usually somewhat higher. Various scientific studies have shown that these conditions are sufficient to destroy PFAS to a large extent (see chapter 3.5). Insignificant amounts of PFAS could be detected in the ash. Measurements of fluorine gases in the exhaust air could not be identified. It is therefore recommended to measure such gases in the exhaust gas of waste incineration plants (including sewage sludge incineration plants) in Europe in order to be able to determine the corresponding destruction efficiency of the plants.

In general, thermal treatment is the recommended disposal method for PFAS-contaminated waste, since the PFAS are largely destroyed by this treatment method. Therefore, it is recommended that PFAS-contaminated sewage sludge be submitted to thermal waste treatment.

Landfilling/other treatment

The disposal of sewage sludge in landfills is prohibited. Other direct disposal of sewage sludge includes, for example, delivery to drying plants and other unknown disposal routes. Since the exact treatment routes are unknown, no specific risks and recommendations can be derived. It is assumed that PFAS are not destroyed in drying plants, whereby the risks and recommendations of material recycling apply.

3.8.3 Paper

Affected by waste

Similar to textiles, PFAS are used in the paper industry to impart water- and grease-repellent properties to products. These properties are particularly desirable in food contact papers, which means that these often contain PFAS (see chapter 3.1.3.1 and 3.3.3). Within the scope of this project, however, PFAS could also be detected in the regular recovered paper fraction. (see chapter 3.2.2), which can be attributed either to contamination of these papers or to the targeted use of PFAS. In general, it is assumed that not all papers entering the recovered paper are treated with PFAS (e.g. books, printer paper, newspapers, etc.).

For recovered paper, an average PFAS concentration of ~374 µg/kg was calculated in Germany (compare chapter 3.3.3).

In general, most food contact papers should be disposed of either in packaging or household waste and not in wastepaper (depending on the type of product, compare chapter 3.4.3.1), but it cannot be ruled out that they also end up in the separately collected wastepaper. A specific proportion of food contact paper in the separately collected wastepaper could not be determined.

In general, no conclusive data can be found regarding the wastepaper waste stream. The data of two reports of the paper industry (2020, 2022) are used as a basis and a recycling rate of 80% is assumed. The remaining papers are thermally recycled or landfilled.

Treatment of the waste concerned, risks and recommendations

A total of ~14.5 million tons of wastepaper was generated in Germany in 2020, corresponding to a total PFAS load of ~4,000 kg. Of this, approx. 80% was sent for material recycling, approx. 19.7% was disposed of thermally and approx. 0.3% was landfilled.

Material recovery

The recycling of wastepaper does not destroy the PFAS present. Papers are mixed with water and certain chemicals during recycling to obtain paper fibers. These can then be processed into new paper products. The PFAS either adhere to the fiber and are thus present in the new product, or they enter the water phase, from where they are discharged into local wastewater treatment plants.

Limit values for wastewater from the production of paper, board or cardboard already exist for various pollutants, but no limit values for PFAS are listed in Annex 28 of the Wastewater Ordinance. These could be established in the future.

During recycling, volatile PFAS may be released into the air, which generally poses a risk to humans. For example, various PFAS were detected in a kitchen, but with concentrations in the low ng/m^3 range. These are thus well below typical PFAS limits in the mg/m^3 range. It is possible that PFAS concentrations in paper recycling plants and plants using recycled paper could be higher, but this should be verified by measurements. PFAS are not destroyed during recycling.

In the past, wastepaper was applied to soils together with compost, resulting in area-wide PFAS contamination of soils in some areas (*e.g.*, presumably in Rastatt, see chapter 3.5.3). Composting of residues from the paper industry is now no longer carried out and should generally be avoided.

Furthermore, it is recommended that highly contaminated paper waste or paper waste with a high risk of containing PFAS be separated from the other paper waste and sent for thermal treatment. In this process, the PFAS are destroyed for the most part. Analogous to textiles, measures to raise public awareness could also be helpful here, so that paper containing PFAS is disposed of by consumers either as packaging or as residual waste and is thus frequently thermally recycled.

Thermal treatment

In Europe, waste incineration plants must operate at a minimum temperature of 850°C and a residence time of at least 2 seconds. Since this is a minimum criterion, the actual temperatures are usually somewhat higher. Various scientific studies have shown that these conditions are sufficient to destroy PFAS to a large extent (see chapter 3.5). Insignificant amounts of PFAS could be detected in the ash. Measurements of fluorine gases in the exhaust air could not be identified. It is therefore recommended to measure such gases in the exhaust gas of waste incineration plants (including sewage sludge incineration plants) in Europe in order to be able to determine the corresponding destruction efficiency of the plants.

In general, thermal treatment is the recommended disposal method for PFAS-contaminated waste, since the PFAS are largely destroyed in this treatment method. It is therefore recommended that PFAS-contaminated fractions of paper waste should also be submitted to thermal waste treatment wherever possible.

Landfilling/other treatment

A small proportion of paper residues is still landfilled due to the system. PFAS can often be detected in leachate from landfills, which may be due to leaching by rain, or to degradation of, for example, side-chain fluorinated compounds. The leachate may enter the landfill soil, but is usually discharged to the local wastewater treatment plant where it is treated.

Volatile PFAS were also detected in the air around a landfill. However, the concentrations were in the low ng/m^3 range and are thus far below typical PFAS occupational exposure limits in the mg/m^3 range.

To minimize risks (e.g., direct release of PFAS from the landfill to the environment), regular maintenance of leachate collection systems is recommended. Precipitation and adsorption are effective methods to remove PFAS from the liquid phase and subsequently destroy them thermally (see chapter 3.5.3).

In general, PFAS-contaminated (paper) waste should not be landfilled because the PFAS are not destroyed and there is a risk that they will be released into the environment via air or leachate.

3.8.4 Soils

Affected by waste

PFAS are not intentionally applied to soils, but are a contaminant. For example, they can be applied to soils through the application of PFAS-contaminated sewage sludge, but also through atmospheric deposition. In some areas in Germany there are larger area contaminations of soils with PFAS e.g. in Rastatt by the application of PFAS contaminated paper sludge, as well as in the area of Gendorf by the production and use of PFOA and fluoropolymers and associated emissions among others into the air and subsequent deposition on the soils of the surrounding area and finally in the vicinity of airports by the use of fire extinguishing foams containing PFAS.

An average PFAS concentration of ~18 µg/kg in Germany was calculated (compare chapter 3.3.4).

Waste soils can be found under EWC code W126 (see chapter 3.4.4.1). The waste stream of waste soils is by far the largest waste stream considered in this report in terms of volume, with about 129 million tons. In this context, 85% of the soils are recycled and about 15 are disposed of, mostly by landfilling (see chapter 3.4.4.1).

Treatment of the waste concerned, risks and recommendations

A total of ~129 million tons of waste soil was generated in Germany in 2018, corresponding to a total PFAS load of ~2,300 kg. Of this, ~85% was sent for material recycling, ~0.2% was thermally treated, and ~15% was landfilled.

Material recovery

The recycling of waste soils does not destroy the PFAS present. Soils are mostly backfilled, which means that they are used for recultivation of excavations or for construction purposes in landscaping. PFAS limits for recycling of soils already exist (see chapter 3.5.4), but these do not apply to waste treatment.

In general, PFAS can be leached out of the soils and eventually reach the groundwater. However, there are differences in the binding strength of soils, as some bind PFAS more strongly, but concrete studies are still lacking on this (see chapter 3.5.4). It is therefore recommended to analyze this.

During excavation, volatile PFAS in particular may become airborne and be inhaled. It might therefore be considered to wear suitable protective equipment (*e.g.*, protective mask and gloves) during such operations. However, in order to determine the need for such protective measures, appropriate air measurements should be carried out.

It is generally recommended to remediate PFAS-contaminated soils before reuse in order to remove or destroy the PFAS contained. Various chemical and thermal processes are capable of effectively destroying PFAS, but these are often not yet established on a large scale, which is why there is still a need for research in this area. Alternatively, PFAS can also be immobilized, washed out or removed from the soil by means of pump-and-treat processes (see chapter 3.5.4).

Thermal treatment

In Europe, waste incineration plants must operate at a minimum temperature of 850°C and a residence time of at least 2 seconds. Since this is a minimum criterion, the actual temperatures are usually somewhat higher. Various scientific studies have shown that these conditions are sufficient to destroy PFAS to a large extent (see chapter 3.5). Insignificant amounts of PFAS could be detected in the ash. Measurements of fluorine gases in the exhaust air could not be identified. It is therefore recommended to measure such gases in the exhaust gas of waste incineration plants in Europe in order to be able to determine the corresponding destruction efficiency of the plants.

In general, thermal treatment is the recommended disposal method for PFAS-contaminated waste, since the PFAS are largely destroyed by this treatment method. For this reason, it is recommended that even heavily PFAS-contaminated fractions of soil waste should be submitted to thermal waste treatment wherever possible.

Landfilling/other treatment

Approximately 15% of soil material generated as waste is deposited in landfills. PFAS can often be detected in leachate from landfills, which may be due to leaching by rain, or to degradation of, for example, side-chain fluorinated compounds. The leachate may enter the soil of the landfill, but is usually discharged to the local wastewater treatment plant, where it is treated.

Volatile PFAS were also detected in the air around a landfill. However, the concentrations were in the low ng/m^3 range and are thus far below typical PFAS limits in the mg/m^3 range.

To minimize the risks (*e.g.*, direct discharge of PFAS from the landfill to the environment, inhalation of PFAS at the landfill), regular maintenance of the leachate collection systems is recommended. Precipitation and adsorption on, for example, activated carbon filters are effective methods to remove PFAS from the liquid phase and subsequently destroy them thermally (see chapter 3.5.3).

In general, PFAS-contaminated (soil) waste should not be landfilled untreated because the PFAS are not destroyed. For soils, underground disposal can also be considered as a waste treatment, but the PFAs are not destroyed in this process either.

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A Appendix

Table 70: Exemption for PFAS according to POPs Regulation (consolidated version of 2.2.2021)

PFAS	Exception for use as an intermediate or other specification
Perfluorooctane sulfonic acid and its derivatives (PFOS)	The use of products already in use in the Union before 25 August 2010 and containing PFOS shall be permitted. The third and fourth subparagraphs of Article 4(2) shall apply to such products.
	Provided that the amount of PFOS emissions to the environment is minimized, the production and placing on the market shall be permitted until September 7, 2025 for use as a spray suppressant for non-decorative hard chrome plating (chromium VI) in closed loop systems. Provided that Member States where PFOS is used report to the Commission by September 7, 2024 on the progress made in eliminating PFOS and justify why this use is still necessary, the Commission will consider whether the exemption for this use of PFOS should be extended for a maximum of five years from September 7, 2025.
Perfluorooctanoic acid (PFOA), its salts and PFOA-related compounds	By way of derogation, the production, placing on the market and use of PFOA, its salts and PFOA-related compounds are permitted for the following purposes: (a) photolithographic or etching processes in semiconductor manufacturing, until July 4, 2025; (b) photographic coatings on films, until July 4, 2025; (c) oil- and water-repellent textiles to protect workers from hazardous liquids that pose risks to their health and safety, by July 4, 2023; (d) invasive and implantable medical devices, by July 4, 2025; (e) production of polytetrafluoroethylene (PTFE) and polyvinylidene fluoride (PVDF) for the manufacture of. (i) high performance, corrosion resistant gas filter membranes, water filter membranes, and medical textile membranes, (ii) industrial waste heat exchangers, (iii) industrial sealants that can prevent leakage of volatile organic compounds and PM2.5 particulate matter, until July 4, 2023.
	By way of derogation, the use of PFOA, its salts and PFOA-related compounds in firefighting foam for the suppression of vapors from liquid fuels and fires of liquid fuels (Class B fires) already filled in systems, both mobile and fixed, is allowed until July 4, 2025. July 2025, subject to the following conditions: a) firefighting foam that contains or may contain PFOA, their salts, and/or PFOA-related compounds shall not be used for training purposes; b) firefighting foam that contains or may contain PFOA, their salts, and/or PFOA-related compounds shall not be used for testing unless all releases are captured; c) beginning January 1, 2023, uses of firefighting foam that contain or may

	contain PFOA, their salts, and/or PFOA-related compounds shall be prohibited. January 2023, uses of firefighting foam that contains or may contain PFOA, their salts, and/or PFOA-related compounds shall be permitted only at sites where all releases can be captured; (d) inventories of firefighting foam that contains or may contain PFOA, their salts, and/or PFOA-related compounds shall be managed in accordance with Article 5
	By way of derogation, the use of perfluorooctyl iodide containing perfluorooctyl bromide for the manufacture of pharmaceuticals shall be permitted subject to a review and evaluation by the commission to be conducted by December 31, 2026, every four years thereafter, and by December 31, 2036.
	The use of products containing PFOA, its salts and/or PFOA-related compounds already in use in the Union before 4 July 2020 shall be permitted. The third and fourth subparagraphs of Article 4(2) shall apply to such products.
	By way of derogation, the manufacture, placing on the market and use of PFOA, its salts and PFOA-related compounds shall be permitted until 3 December 2020 for the following purposes: a) medical devices other than implantable medical devices within the scope of Regulation (EU) 2017/745 ; b) latex printing ink; c) plasma nanocoatings.

Source: Own representation based on POP-VO

Table 71: PFAS regulated in ANNEX I of Regulation (EU) No. 10/2011 (consolidated version of 23.9.2020).

PFAS	Limitations and/or specifications
Chlorotrifluoroethylene	
Tetrafluoroethylene	
Hexafluoropropylene	
Perfluoromethyl perfluorovinyl ether	For use only in non-stick coatings; -fluoropolymers and perfluoropolymers intended for multi-use applications where the contact ratio is 1 dm2 surface in contact with at least 150 kg of food.
Perfluoropropyl perfluorovinyl ether	
Perfluorooctanoic acid, ammonium salt	Only for use with reusable items that are sintered at high temperatures
Perfluoroacetic acid alpha-substituted by the copolymer of perfluoro-1,2-propylene glycol and perfluoro-1,1-ethylene	Only for use up to 0.5% in the polymerization of fluoropolymers processed at 340 °C or above and intended for reusable articles

glycol, with chlorohexafluoropropyloxy end groups.	
Perfluoro[2-(poly(n-propoxy))propionic acid]	For use only in the polymerization of fluoropolymers processed at 265 °C or above and intended for reusable articles.
Perfluoro[2-(n-propoxy)propionic acid]	For use only in the polymerization of fluoropolymers processed at 265 °C or above and intended for reusable articles.
3H-Perfluoro-3-[(3-methoxypropoxy)propionic acid], ammonium salt	Only for use in the polymerization of fluoropolymers when: processed at temperatures above 280 °C for at least 10 min, processed at temperatures above 190 °C up to 30% by weight in blends with polyoxymethylene polymers and intended for reusable articles.
2H-perfluoro-[(5,8,11,14-tetramethyl)-tetraethylene glycolethylpropyl ether]	Only for use as an auxiliary in plastics production in the polymerization of fluoropolymers for (a) reusable and disposable materials and articles that are sintered or processed (not sintered) for not less than 10 minutes at not less than 360 °C or correspondingly shorter at higher temperatures; (b) reusable materials and articles processed (not sintered) for not less than 10 minutes at temperatures between 300 °C and 360 °C
Perfluoro[(2-ethyloxy- ethoxy)acetic acid], ammonium salt	Only for use in the polymerization of fluoropolymers processed at temperatures above 300 °C for at least 10 min.
(Perfluorobutyl)ethylene	For use only as a comonomer up to 0.1 wt% in the polymerization of fluoropolymers sintered at high temperatures.
Perfluor {acetic acid, 2-[(5-methoxy-1,3-dioxolan-4-yl)oxy]}, ammonium salt	Only for use as an auxiliary in the manufacture of plastics in the production of fluoropolymers at high temperatures (at least 370 °C).
2,3,3,4,4,5,5-Heptafluor-1-penten	Only for use together with tetrafluoroethylene and/or ethylene comonomers to produce fluorocopolymers applied as polymer processing aids at a level not exceeding 0.2% by weight of the food contact material, and when the low molecular weight fraction below 1 500 Da in the fluorocopolymer is not more than 30 mg/kg.

Source: Own representation based on (EU) No. 10/2011