

Report summary waste

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1. INTRODUCTION

The Dutch National Institute for Public Health and the Environment (RIVM) has assigned Ramboll Deutschland GmbH to collect data on the fate of PFAS in the waste phase with a specific focus on the mapping of the PFAS freights emitted into the environment or ending up in recycled material.

This overall aim translated into the following specific aims:

- Determination of the PFAS loads for selected waste streams within the European Economic Area (EEA)[1];
- 1. Mapping of the distribution of PFAS loads for the selected waste streams and the associated waste treatment methods (e.g. incineration, landfill, recycling) while highlighting differences between waste treatment across the EEA;
- 2. Identification of waste treatment methods with high risks of PFAS emissions;
- 3. Identification of the role of waste treatment for PFAS emissions to the environment (air, water, soil).

[1] EU 27, Norway, Iceland and Liechtenstein

While the scope of REACH does not cover objects or substances which have become “waste”, its underlying methodology for the identification of risks of a substance for human health and the environment should take into consideration the waste phase. This report summarises the outcome of the conducted data collection and analysis concerning the fate of PFAS in a number of selected waste streams, as well as associated emissions. (See figure 1 bottom left of the scheme). Note: the information in this (summarized) report is based on information (tonnages) from other PFAS use reports.

This report summary provides the outcome of the conducted data collection and analysis exercise concerning the fate of PFAS in a number of focus waste streams, as well as associated emissions in the waste stage.

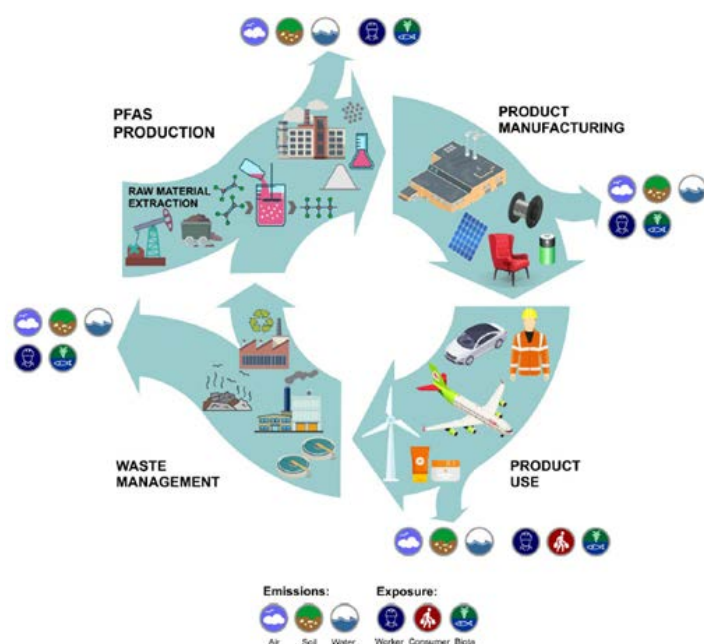


Figure 1: Emissions of an exposure to PFAS during their lifecycle (EEA-ETC report, Systemic view on fluorinated polymers, forthcoming 2020, as cited in European Commission, 2020)

2. Methodology

2.1 Identification of focus waste streams

In order to derive mass flows which cover a high PFAS freight likely related to significant emission risks during waste treatment and/or recycling, the most relevant waste streams for PFAS were selected based on a set of criteria. These included but were not limited to:

- Waste streams with high volumes in the EU/EEA;
- Waste streams with high average PFAS concentration or freight;
- Waste streams where high recycling rates occur/are assumed;
- Waste streams where high releases into the environment occur/are assumed (landfilling, land application, recycling).

This led to the following wastes being chosen for further analysis:

- Textiles
- Food contact material (paper and board)
- End-of-life-vehicles (ELV)
- Electrical and electronic equipment and
- Sewage sludge (not a work package in the restriction, but added because of possible PFAS load)

The information needed to assess the proposed criteria was derived from:

- Eurostat;
- Information provided from other work packages (WPs) under the restriction dossier preparation and
- A targeted literature research

Waste treatment categories were outlined according to European legislation

2.2 Emission factors and degradation products

For the emission factors and degradation products a literature review was performed. For this a variety of search operators based on the following terms were applied:

“Perfluoroalkyl”, “PFAS”, “WWTP”, “landfill”, “waste water”, “incineration”

Additionally, reports from environmental agencies, organisations and NGO’s were analysed. Many publications were identified in cooperation with RIVM. The focus was on European data, however, publications from north America, Asia and Australia were also included.

For an emission factor PFAS input as well as PFAS output is (i.e. to landfill). needed This picture often in not complete as in many cases the PFAS input (i.e. to landfill or incineration) is unclear. Therefore the total PFAS (mean and median) concentrations (i.e. from literature) were used to calculated emissions per inhabitant and total total yearly EEA PFAS freights.

2.3 Waste water treatment

For the WWTP the averages and sum concentrations for the influent, effluent and sludge were listed. Additionally, where possible, emission factors were calculated for each publication, however these were not used for further calculation as the final emission factors were derived from all publications. For the emission factors the following formula was used:

$$Emission\ factor = \frac{c(effluent)}{c(influent)} \cdot 100$$

With the acquired mean and median Σ PFAS values an emission factor was calculated. For this inflow and discharge as well as sewage sludge production data for the year 2016 from Eurostat was taken.

2.4 Landfilling

For landfilling the same principle as described for waste water treatment was applied. The data from the publications was listed and the mean and median concentrations for each PFAS group and the Σ PFAS was calculated. No difference was made between treated and untreated leachate as leachate in Europe is largely sent to a WWTP.

As there is no Eurostat data for the leachate generation in Europe a value of 0.2 – 1.0 m³/year generation of leachate per tonne of landfilled waste was taken from the literature (Brennan et al., 2016).

2.5 Waste incineration

For waste incineration, no sufficient quantitative data on PFAS and F-gases and flue gas could be acquired. As a consequence, no emission value for PFAS/F-gases from incineration via exhaust air could be derived. However, some publications with data on PFAS in bottom ash and one publication with data on fly ash could be found from which mean and median values for the PFAS groups and Σ PFAS were calculated. Eurostat provides data for combustion wastes however, this category also includes non-bottom ash wastes such as solid wastes from gas treatment and flue-gas dust. A publication from 2019 was found which lists the generation of bottom ash from European incinerators for 20 EU countries (Blasenbauer et al., 2020). For the fly ash, a generation factor of 3% of the total incinerated waste was used from Abis et al. (2020). With this data, the total amount of PFAS in incinerator ash was calculated. By dividing the total amount of PFAS by the population an emission value from incineration via bottom and fly ash with the unit mg/(year*person) was derived.

3. Waste streams

Based on the assessment described five waste streams were selected (see table 1 below). Information on the main waste treatment application, PFAS concentration and or PFAS freight within these waste streams is indicated.

Table 1: Relevant wastes and associated waste streams

Selected waste	PFAS quantities used in articles / PFAS consumption [t/a]	Relevant waste streams	Main waste treatment of waste stream	PFAS freight [t/a]	Average PFAS concentration [mg/kg]
Textiles ⁵	50,853	Household and similar waste	Disposal - landfill	43,605	5,095 ¹
		Textile waste ³	Recovery - recycling		135 ¹
		Medical waste	Disposal - incineration		228 ¹
Food contact material (paper and board)	827 – 4,962	Paper & cardboard waste	Disposal – landfill	2,894 t ²	0.07 ¹
		Household and similar waste	Disposal - landfill		
End-of-life-vehicles (ELV)	unknown	Shredder light fraction (ELV)	Disposal (not further specified)	2,129 ²	2 ¹
		Shredder heavy fraction (ELV)	Recovery - recycling		0.37 ¹
Waste Electrical and Electronic Equipment	unknown	Categories 2 to 4 according to the Directive 2012/19/EU	Recovery – recycling		unknown
		Batteries and accumulators	Recovery – recycling		
Sewage sludge	n/a	Sewage sludge from urban waste water treatment	Recovery - agricultural use	0.404 t ²	Median concentration 0.06124

¹ Concentration was calculated based on freights.

² Freight is based on a calculation.

³ As presented within Eurostat. Includes separately collected worn clothing, miscellaneous textiles waste and leather waste.

⁴ Section 0 elaborates why the median was chosen

⁵ Not all TULAC (textiles, upholstery, leather, apparel and carpets) could be considered within this analysis, as no information on treatment of technical textiles and “other” is available

n/a= not applicable

In the following, the derived mass flows and underlying assumptions are presented for the focus waste streams.

3.1 Textile waste

Eurostat records separately collected textiles, leather waste, and waste arising from processing textile fibres as “Textile waste”. However, only a small share of textiles is collected separately. Based on Watson et al. (2018) and their assessment of a separate collection of clothing and household textiles in the EU in 7 countries (DE, DK, FR, IT, NL, SE and UK, years assessed 2010-2016) an average share of separate collection of 36% can be estimated.

The overall amount of textile entering a specific waste treatment option cannot be determined precisely e.g. because of a lack of data on the share of textile waste in “Health care and biological wastes”). However, but due to the high share of textiles in the “Household and similar waste” (64% are not collected separately), where most of the waste volume is treated through energy recovery or is disposed of in landfills, it is assumed that largest proportion of textile waste is treated accordingly. In line with this, it was stated by Bioten (2021) that 87 % of the total fibre input in Europe is ultimately destined for landfill or incineration, with significant leakages into natural environments.

With regards to future trends, it was found that the average consumer today buys 60 percent more clothing than 15 years ago. Individual items however are kept only half as long (United Nations Environment Programme (UNEP), 2019). In the future, global clothing sales are expected to continuously grow and are predicted to reach 160 million tonnes in 2050. This is more than three times today’s amount and would ultimately result in an increasing share of textile waste (Ellen MacArthur Foundation, 2017).

The share of the average separate collection (36 %) was taken into account when calculating the concentration for “Textiles waste” and the concentrations for the “Household and similar waste” (64%) containing home textiles, consumer apparel and professional apparel. This led to a concentration of total PFAS of 5,095 mg/kg for “Textile Waste” and 136 mg/kg for “household and similar waste.

For “Health care and biological wastes” the freights calculated for medical textiles entering the waste stream were used. As no information on the share of textiles within this waste stream was available, no corrections were made for concentration and it is assumed that the entirety of medical textiles will enter the “Health care and biological wastes” leading to a concentration of 229 mg/kg.

In figure 2 a flowchart for Textile waste and Household waste (containing a.o. not separate collected Textile waste) is presented.

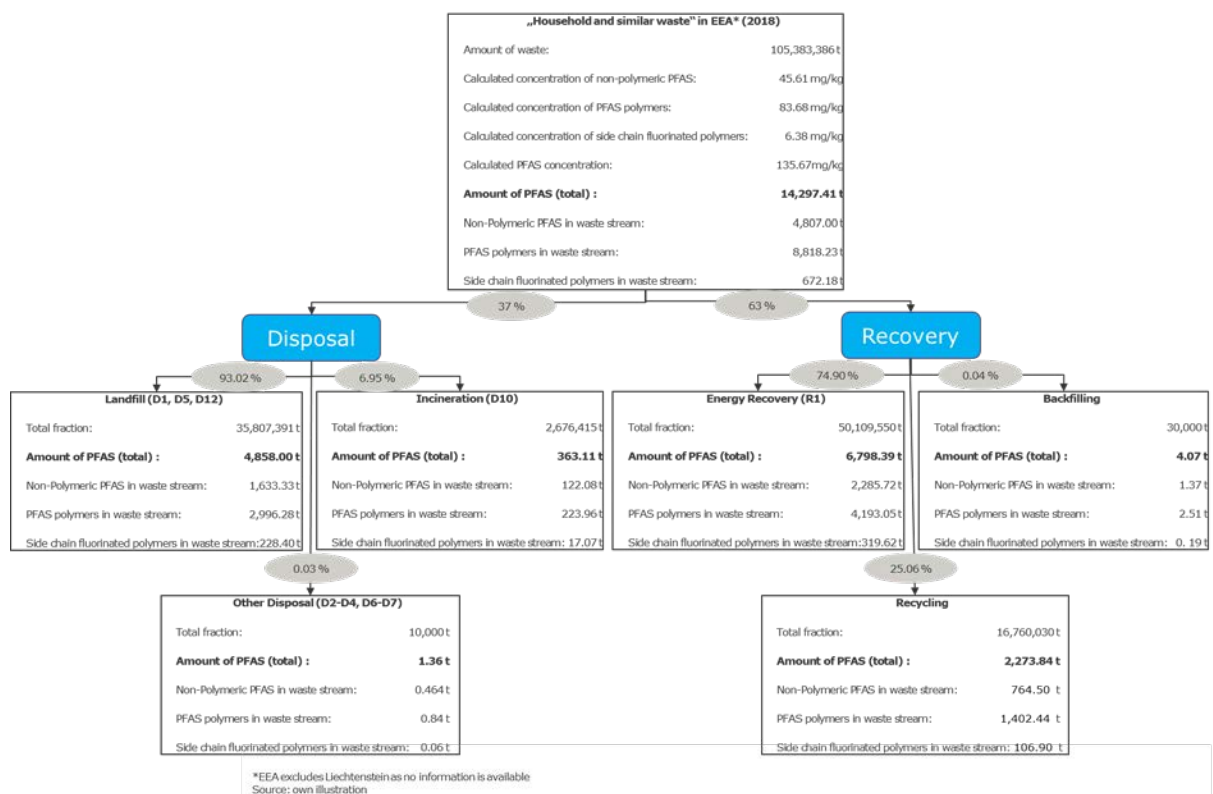
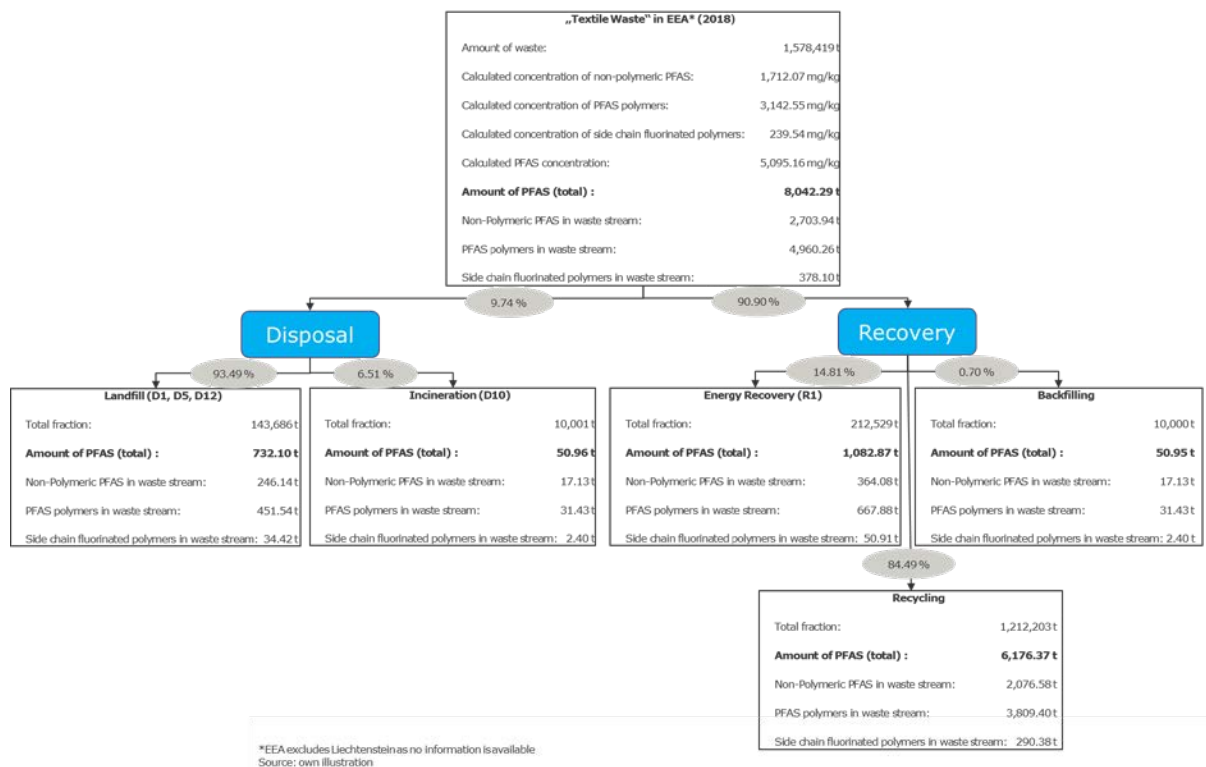


Figure 2: Flowchart Textile waste and household waste.

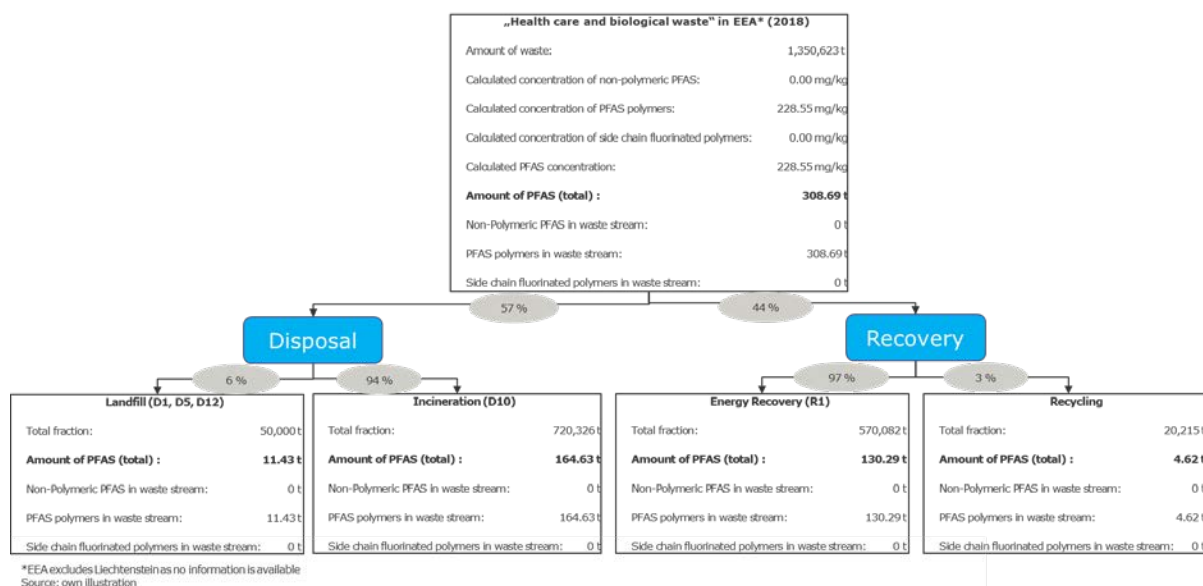


Figure 3: Flowchart Healthcare and biological waste (for Medical Textiles)

The mass flows indicate that 14,113 tonnes of PFAS (5,603 from landfills and other disposal and 8,510 from recycling and backfilling operations) are not destroyed and possibly enter into the environment within the EEA and/or remain in the material cycle.

Generally, it has to be noted, that apparel and other textiles which are reused can contribute to the global distribution of PFAS. Considering the average lifetime of textiles, it can be estimated that if a full ban of PFAS was to take place in 2024, PFAS concentration will still be present in waste streams until 2037 and beyond, dependent on the increase of recycling.

3.2 Waste from food contact material (FCM) - Packaging

The yearly EEA quantity of PFAS used in paper and board packaging was estimated with between 827 and 4,962 tonnes, based on intentionally added PFAS (Hollins, n.d.)

The average lifetime of food packaging can be assumed to be around one year based on information on plastic packaging (Conversio Market & Strategy GmbH, 2018) but could be higher for other applications such as cupcake forms.

Depending on the food collection system in place within the country and sometimes the municipality, the collection of these items can differ. Food-contact articles and thermal paper, wet-proof and/or greaseproof impregnated and/or glued paper and cardboard¹ shall be disposed of in the residual waste in Germany (German Environment Agency (UBA), 2020). Similar approaches are assumed for other European countries as paper for recycling must be kept separate from other waste as contaminated papers are not acceptable for recycling (EPRC, n.d.). The reality, however, can differ, and items can be and are partly disposed of via the separately collected paper waste.

Plastic waste was not further analysed for FCM and packaging applications as it was assumed that PFAS amounts are limited and quantitative data were lacking. Thus, only paper and cardboard waste

¹ such as posters, coffee-to-go cups, fast food wrappers, baking paper, muffin forms as well as solid, empty paper packaging such as pizza cardboard packaging, varnished, glazed or chromo papers, and boards produce with plastic varnishes or films as well as papers with adhesives applications which cannot be easily separated such as sticky notes, self-adhesive seals for envelopes

and household waste were evaluated for waste from FCM containing PFAS. Within Eurostat “Paper and cardboard waste” encompasses waste from paper and cardboard packaging (15 01 01) as well as paper and cardboard waste from mechanical treatment (19 12 01) and separately collected fractions (20 01 01) (European Commission, 2010). Considering the findings on the littering of plastic waste from Jepsen et al. (2020) it is not unlikely, that PFAS-containing paper and cardboard fractions are not accounted for within the waste streams as they enter into the environment directly via littering. This has been indicated within the mass flows.

Within Eurostat, the waste fraction “Household and similar waste” contains bulky waste (20 03 07) as well as street-cleaning residues (street-cleaning residues). Based on Eurostat data from 1990-2001 bulky waste presents a share of on average 8% of household and similar waste. This is based on data reported for 2000 and 2001 for several European countries (Communities, 2003). In absence of newer information, 8 % are deducted from the waste fraction “Household and similar waste” presented within Eurostat for 2018 to have a better representation of municipal (including residual) waste as typically no relevant paper and cardboard waste should enter the bulky waste fraction.

In the “Paper and Cardboard” waste stream all of the waste amount in EEA is treated by recovery through recycling, except a minor amount in Western Europe that is recorded under energy recovery (R1). Considering that FCM packaging end up in the paper and cardboard waste stream to a limited extent, it is likely, that a large share of FCM is either landfilled or incinerated if it is disposed of in the residual waste.

The PFAS concentrations within the relevant waste streams were calculated based on a range of PFAS freights in products (t/a) and the quantities of paper and board used for packaging in Europe in 2019 as stated in WP 4.C (Hollins, n.d.). This led to a PFAS concentration of 69.92 mg/kg.

With regards to the fraction of paper in household waste the Netherlands report around 20 % of the paper in the household residual waste based on samples taken (Rijkswaterstaat Environment, n.d.). Germany states that around 5.2 % of the residual household waste corresponds to waste paper, other European cities indicated shares of around 21 to 30 % of the paper in residual waste (BMU, 2020; Zero Waste Europe, 2020). Based on the calculated average, as no information on all EEA countries is available, an average share of paper and cardboard in the household/residual waste of 19 % can be assumed, which was taken into consideration when calculating the PFAS freight for household waste and was applied to all treatment fractions equally.

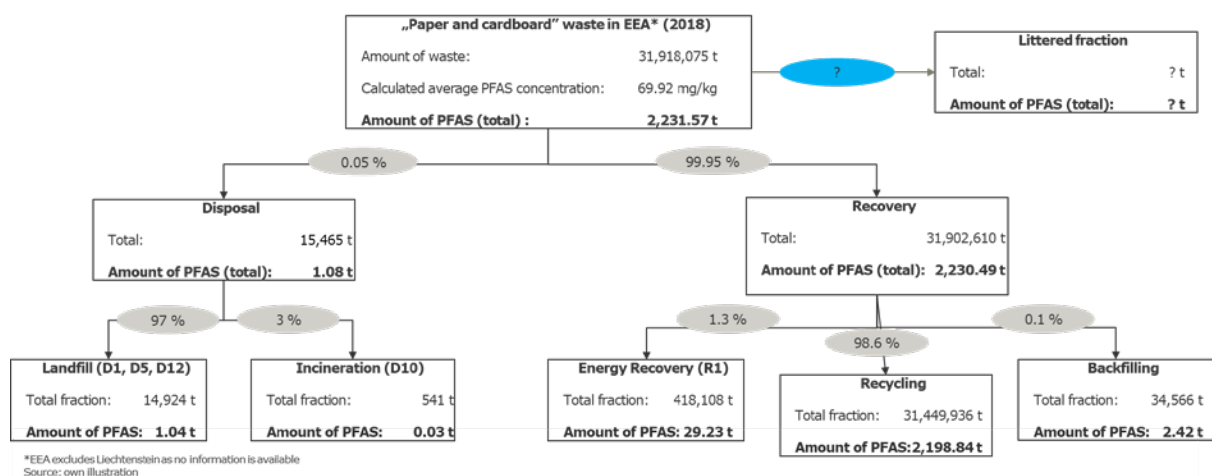


Figure 4: Flowchart Paper and cardboard waste

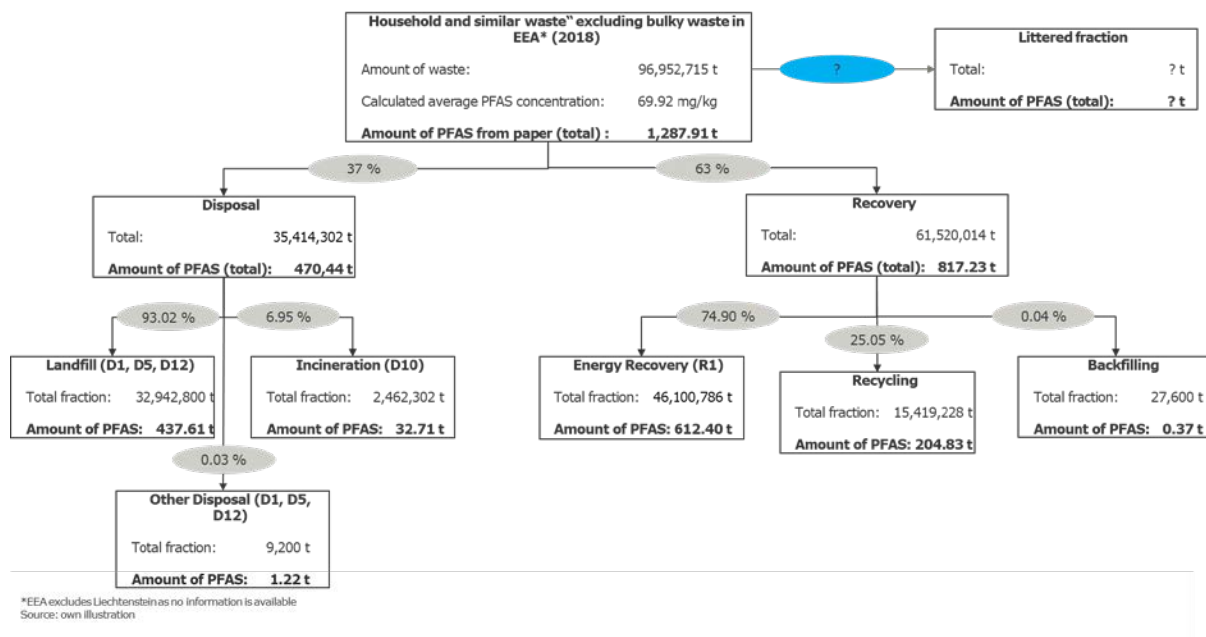


Figure 5: Flowchart Household and similare waste

The mass flows indicate that a possible total of 2,846 tonnes of PFAS (440 t landfilling and other disposal (D2-D4, D6-D7²) and 2,406 t recycling and backfilling) are not destroyed and can annually enter into the environment in the EEA and/or remain in the economic cycle. Considering the treatment paths, emissions can arise to air, water and soil.

3.3 WEEE

According to the World Economic Forum (2019), WEEE presents the fastest-growing waste stream in the world. It is estimated that the waste stream will almost triple the tonnage reported in 2018 without intervention by 2050. Considering the forecasts of an increase of electronic and electrical devices a drastic increase in the generation of this waste stream seems likely.

With regards to exports, a study conducted by the Basel Coordination Centre for Africa (BCCC) and the United Nations University (UNU) found a large number of incorrectly or completely undeclared e-waste and ELV exported to Nigeria during the research period (2015 to 2016). Appropriate disposal or recycling leading to the destruction of the PFAS content is not necessarily ensured in the importing countries. Thus, the disposal of WEEE in recipient countries may contribute to the global distribution of PFAS and thus to possible risks to human health and the environment. The concentration or freight of PFAS within WEEE entering the waste stage are not (yet) available.

It should be noted that the WEEE Directive currently does not contain any explicit provisions or requirements concerning PFAS. Furthermore, it should be noted that, despite the applicable regulatory framework of the WEEE Directive, illegal or unsound treatment may take place, creating a

² D2: Land treatment, D3: Deep injection, D4: Surface impoundment, D6: Release into a water body except seas/oceans, D7: Release to seas/oceans including sea-bed insertion according to directive 2008/98/EC

risk of emissions of PFAS contained in WEEE into the environment. Finally, while Regulation (EC) No 1013/2006 on shipments of waste (WSR) prohibits the export of WEEE to non-OECD countries for recovery, recycling and disposal, illegal export can occur (Odeyingbo et al., 2018). WEEE which is illegally transported to non-OECD countries could in turn undergo unsound treatment methods leading to emissions of PFAS and risks to the environment.

3.4 End of Life Vehicles (ELV)

The quantity of PFAS used in vehicles in the EU corresponds to 2,129 tons / year (see also Table). It was further noted by Plastics Europe within their report on fluoropolymer industry in Europe, that transport, as one of their key sectors, has the highest shares of fluoropolymer sales in 2015 with 18,500 tonnes (Fluoropolymers Committee PlasticsEurope AISBL, 2018). Within the U.S. transport is ranked as the second most important sector in terms of production value following electronics (Fluoropolymer Industry, 2018). Cars have an average lifetime of 17 up to 20 years (Potrykus et al., 2019).

As most PFAS-applications in vehicles are textiles and polymer applications, the relevant fraction in which PFAS from WEEE end up are non-ferrous materials from shredding also referred to as shredder heavy fraction (SHF) and the shredder light fraction (SLF). The SLF is a mixed fraction and includes, for example, textiles, foams, plastics and plastic films as well as broken glass, paint residues and wood (BDSV, 2012). Median values calculated based on several literature sources indicate that a share of about 74 % of plastics ends in the SLF. Median values calculated for the SHF indicate a share of 4 % of plastics (Martens, 2011; Ramboll Deutschland GmbH, 2020; Sander et al., 2020; Wilts et al., 2016).

In the future, it is expected that the quantities of vehicles placed on the market will increase (Kuhnert et al., 2018). However, a shift to electric cars or hybrid electric cars is expected. In Germany, around 14 % of all newly registered passenger cars in 2020 had an electrically powered engine (battery-electric, plug-in, fuel cell) (Kraftfahrt-Bundesamt, 2021). On the European market, the average share of new passenger plug-in electric cars lies at 11.4 % in 2020 (Kane, 2021). For relevant waste streams, this means increasing quantities of SLF and SHF.

The PFAS concentration within the waste streams was calculated based on the 350 g PFAS polymers per car as reported by Améduri (2020) and the number of ELVs in the EU in 2018 leading to a total PFAS freight of 2,129 t/a within ELVs. Considering the lack of data on historical production and partially historical usage data (see also (Z. Wang et al., 2014)) as well as data on losses during the use phase, it is assumed that the calculated freight, based on the number of ELV, enters into the waste stream. In the future, an increase in the use of PFAS polymers in vehicles up to 1-2 kg is predicted according to Améduri (2020).

Based on the plastic share within the SLF and SHF and the total quantities treated, a concentration of 20 mg/kg and 4 mg/kg for the respective fractions was calculated. In absence of further information, for the calculation of mass flows it was assumed that no PFAS ends within the other shredder fraction (other material arising from shredding and ferrous scraps). In reality concentration of SHF and SLF can be lower, as other fractions, besides plastics and textiles, end up in both fractions and PFAS might also be distributed to other fractions.

As no information on the concentration in batteries was reported, no concentration could be calculated.

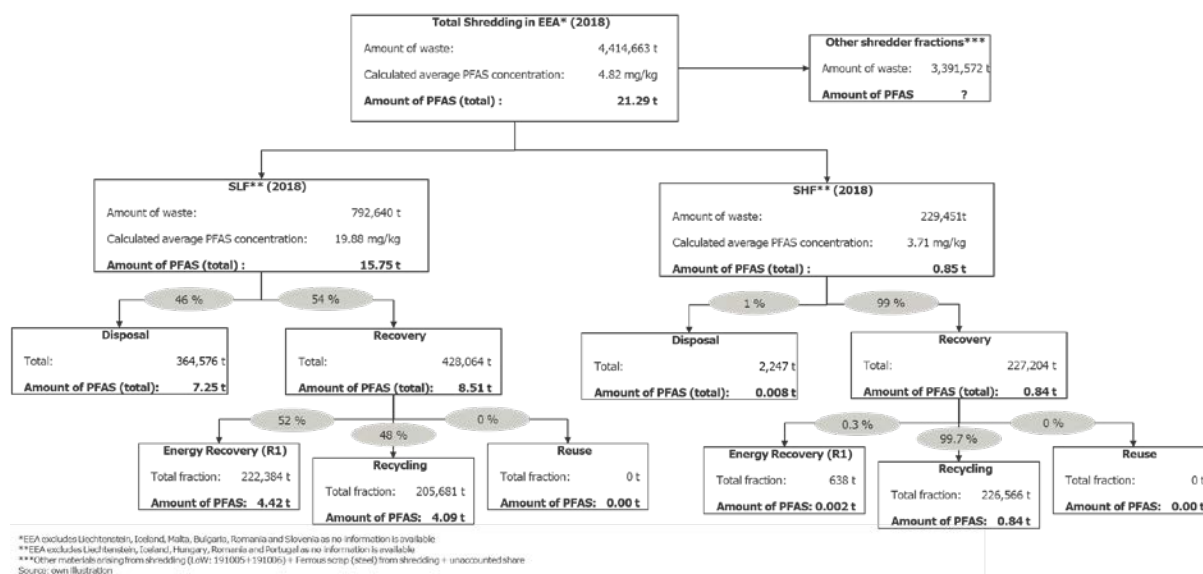


Figure 6: Flowchart ELV shredding

The mass flow indicates that a possible yearly total of 12 tonnes of PFAS (7 t from disposal and 5 t from recycling) are not destroyed. This number might increase in the future, as Améduri (2020) estimated an increase to 1,500 g PFAS/car in the future. With regards to disposal, data do not indicate if this refers to incineration without energy recovery or simply landfilling. In several Member States, it is still allowed to dispose of SLF in landfills other countries require a pre-treatment of the SLF before landfilling (Mehlhart et al., 2018). Generally, neither the recycling process nor landfilling destroys the PFAS content within the relevant fractions. Both paths can contribute to the distribution of PFAS in the environment.

3.5 Sewage sludge from urban waste water treatment

Sewage sludge is generated in WWTP by separating the undissolved particles from the water, which is done in lagoons or basins. As WWTP receives waters from urban and industrial sources they can contain PFAS originating from the production and use phase of PFAS products (e.g. cosmetics).

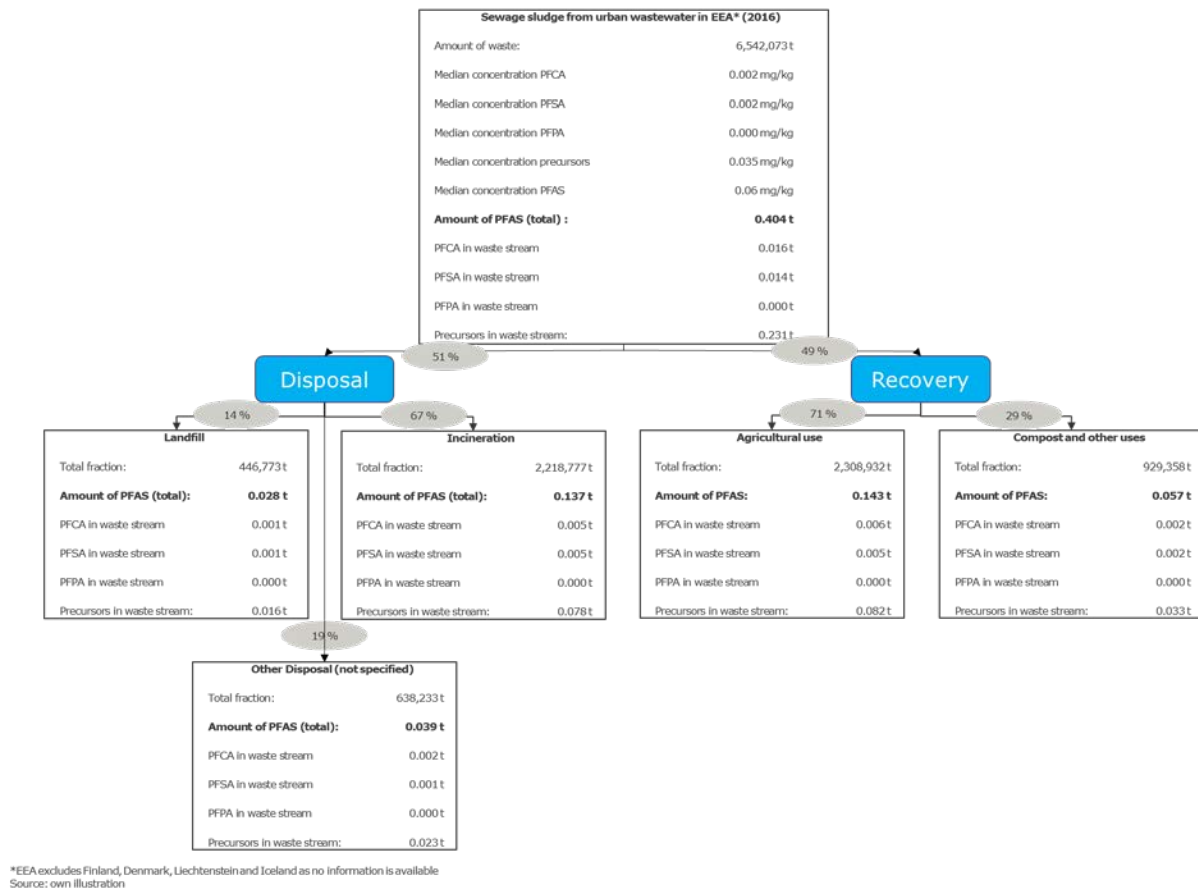
The most recent data (2016) was used to calculate the amount of sewage sludge for the EEA. Moreover, it has to be noted, that the data reported within Eurostat on production does not align with the tonnages treated as waste, which was also found by Pellegrini et al. (2016) who highlighted the structural lack of homogeneity and reliability on the Eurostat data on sewage sludge.

Generally, the total quantities fluctuate slightly but remain between 5,78 and 6,53 million tonnes per year since 2011. Within the EEA sewage sludge from urban waste water treatment is disposed of or recovered (land application) in roughly equal proportions.

Pellegrini et al. (2016) reported, that due to the Landfill Directive (1999/31/EC) amounts of sewage sludge disposed of in landfills will rapidly decrease in upcoming years as Member States reduce the amount of biodegradable waste sent to landfills by 2016.

In Eastern Europe, an increasingly larger percentage of households connected to treatment plants can be expected. Here, agricultural use of the sludge is still considered the preferred disposal method. The reuse of biosolids as soil improver/fertilizer in arable crops represented the most used disposal/recovery option in some European countries. This has led to restrictions in the use of

biosolids with Directive 86/278/EEC. However, an evaluation of the directive in 2014 has found shortcomings also with regards to contaminations such as PFAS. These are currently not regulated. Most countries in the EU have prohibited the use of untreated sludge on land, while some Member States (France, Ireland, and the UK) permit the use of untreated sludge (Collivignarelli et al., 2019). Currently, among the EU-27 countries France, Finland, Germany, Ireland, Italy and Spain have the highest share of biosolids recycled to land.



However, limitations in the calculation of PFAS concentrations and freights in waste have to be taken into consideration e.g. that all calculations are based on available data from the relevant work packages under the restriction dossier and the assumptions made.

Table 2: Overview of shares of PFAS not destroyed per waste stream per year

Selected waste	Relevant waste streams	Total freight of PFAS [t/a]	Incinerated freight of PFAS [t/a]	% PFAS possibly not destroyed	PFAS freight possibly not destroyed (Recycling, Reuse, Disposal, etc.) [t/a]	% PFAS possibly not destroyed from all considered (5) waste streams ³
Textile waste	Textile waste ²	8,042.29 ⁴	1,133.83	87	6,959.42	27.3
	Household and similar waste	14,297.41 ⁴	7,161.5	50	7,137.27	26.6
	Medical waste ¹	308.69 ⁴	294.92	5	16.05	0.06
Food contact material & other paper applications	Paper & cardboard waste ¹	2,231.57	29.26	99	2,202.3	8.4
	Household and similar waste (excluding bulky waste)	1,287.91 ⁴	645.11	50	644.03	2.5
Waste Electrical and Electronic Equipment	N/A	N/A	N/A	N/A	N/A	N/A
End-of-life-vehicles (ELV)	Shredder light fraction (ELV) ¹	15.75	4.424	72	11.34	0.04
	Shredder heavy fraction (ELV) ¹	0.85	0.0002	100	0.85	0.003
Sewage sludge	Sewage sludge from urban waste water treatment	0.404	0.137	67	0.269	0.001

¹ The waste stream shows the entirety of the stream as no information is available on the share of the relevant PFAS-fractions within it

² As presented within Eurostat. Includes separately collected worn clothing, miscellaneous textiles waste and leather waste

³ For which data is available

⁴ Discrepancies between total freight and sum of PFAS incinerated and not destroyed arise from discrepancies in the Eurostat data used

4. Emission factors

4.1 Waste water treatment plants

In general, the analysed literature concludes that WWTP are not effective in destroying/removing PFAS. Sometimes higher PFAS concentrations of the targeted PFAS were found in the effluent water than in the influent water.

Additionally, PFAS can also be found in the sludge resulting from the waste water treatment. Most authors concluded that long-chain PFAS tend to accumulate more in the solid phase than in the liquid phase due to higher intermolecular interactions between the long PFAS chain and the solids (Coggan et al., 2019; Glimstedt, 2016).

Some publications also measured PFAS in the air above the water and around the plants. However, these emissions are assumed to be up to a factor 10 lower compared to the PFAS emissions from the effluent (Ahrens et al., 2011). The following mean and median concentrations were calculated:

Table 3: Calculated mean and median concentrations for the PFAS groups in the influent and effluent water as well as sludge (WWTP). Data taken from 7 studies.

Substance	Mean influent [ng/L]	Median influent [ng/L]	Mean effluent [ng/L]	Median effluent [ng/L]	Mean sludge [ng/g]	Median sludge [ng/g]
ΣPFCA	618.21	150.15	388.11	101.52	50.58	2.42
ΣPFSA	59.19	12.16	162.40	21.28	16.40	2.08
ΣPFPA	4.41	0.00	0.90	0.00	0.00	0.00
ΣPrecursors	115.71	8.13	16.55	1.50	46.87	35.33
Total PFAS	795.47	395.67	567.38	182.80	113.85	61.81

Table 4: Reported influent, effluent and sludge data from Eurostat for 2016.

Influent [million m ³]	Effluent [million m ³]	Sludge production [tonnes in d.s.]
14,299	16,944	6,577,000

Table 5: Calculated total yearly PFAS freights for selected EU-Member States based on the available Eurostat data (WWTP). All values in [kg]

Mean PFAS freight influent	Median PFAS freight influent	Mean PFAS freight effluent	Median PFAS freight effluent	Mean PFAS freight sludge	Median PFAS freight sludge
11,374	5,657	9,614	3,097	748	406

From these freights, the removal efficiency and emission factors can be calculated. The removal efficiency and emission factors were calculated with the following formula:

$$Emission\ factor = \frac{freight(effluent) + freight(sludge)}{freight(influent)} \cdot 100$$

$$Removal\ efficiency = 100 - Emission\ factor$$

The calculated median removal efficiency of the European WWTP is 42 % meaning that roughly 58 % of the incoming PFAS would be emitted into the European surface waters or be found in the sludge, which in some cases is also applied on land

While these calculations show, that PFAS are removed to a certain degree their exact fate inside the WWTP is mostly unknown (Hamid & Li, 2016). One possible explanation is that the PFAS are removed/destroyed in the chemical treatment step of WTPs. These include precipitation, adsorption, ion exchange resins and oxidation via chlorine, ozone or UV-light (Samer, 2015).

If the calculated PFAS flows are divided by the population of the above-mentioned countries in 2016 a per capita value is obtained.

Table 6: Calculated per capita input and output values for EEA (WWTP). All values in [mg / (person*year)]

Mean influent per capita	Median influent per capita	Mean effluent per capita	Median effluent per capita	Mean sludge per capita	Median sludge per capita
76.5	38.0	64.6	20.8	2.1	1.1

From this data a median emission in the effluent of 20.8 mg/(person*year) and 1.1 mg/(person*year) for the sludge results.

It should be noted that the effluent from the WWTP directly flows into European surface waters and as such the contained PFAS are directly emitted into the aquatic environment. It is assumed that approximately 2/3 of the total freight of PFAS contained in sewage sludge is not destroyed as it is either applied on land, composted or disposed of in a landfill. Only roughly a 1/3 of the sewage sludge is incinerated.

The calculated per capita emission values can also be compared to literature values from various publications (Ahrens et al., 2011; Campo et al., 2014; Coggan et al., 2019). As only mean values are available, they will be compared to the calculated mean per capita emission value.

Table 7: Comparison of the calculated per capita emission values with emission values from literature (WWTP). In bracket the country from the publication. All values in [mg / (person*year)]

Mean effluent per capita	Median effluent per capita	Coggan et al 2019 [AUS] (mean)	Campo et al 2014 [ESP] (mean)	Ahrens et al 2011 [CA] to air (mean)
64.4	20.8	13	12 - 54	2.56

It can be seen that the calculated mean emission value is in the same order of magnitude as the literature values, although slightly higher. The emission values to air from Ahrens et al. (2011) is by a factor of 25 lower than the mean per capita emission value for the effluent. This indicates that PFAS releases from WWTP via effluent water and sludge are much more significant than via air.

4.2 Landfills

Landfilling in general does not actively destroy containing pollutants. Many modern landfills are equipped with a plastic liner capturing the leachate. However, this is not always the case for older landfills (Lang et al., 2017). In Europe the landfill directive (1999/31/EC) stipulates, that all newly constructed landfills need to be equipped with “a geological barrier and a bottom liner during the operational/active phase”, however it is unclear how many landfills in Europe are currently equipped with liners. It has been shown, that a WWTP receiving leachate from landfill can have up to three-time higher PFAS concentration in the influent than a WWTP that does not receive leachate (Masoner et al., 2020). This indicates the risk of PFAS emissions from landfills (Masoner et al., 2020).

In general, the individual influence of certain waste types cannot be determined. Waste with high PFAS concentrations (e.g. carpets) will contribute to a higher degree. Especially industrial waste has a high potential to contribute to the PFAS concentration in the leachate (ITRC, 2021; Solo-Gabriele et

al., 2020). According to Eurostat 96% of the landfilled waste was non-hazardous waste with only 4% being hazardous, which is then deposited on the respective landfill. The exact number of landfills and landfill types is unknown, however according to Eurelco there are an estimated 500,000 landfills in Europe with 90% preceding the EU-landfill directive 1999/31/EC. Chapter 18 of the ECHA ³ on the other hand provides default values for a landfill scenario stating that there are approximately 8,400 landfills in Europe from which ~400 are for hazardous, ~5,000 for non-hazardous and ~3,000 for inert waste. The data is however from 2006 and should be used with caution.

It has been shown that the climate and the age of waste deposited on the landfill do have an effect on the PFAS concentration in the leachate (Lang et al., 2017). It is assumed, that over time 100% of all containing PFAS or their degradation products will eventually end up in the leachate or in the ground. The project team assumes, that the containing contaminants are not destroyed by the storage on a landfill and will over time be washed out via rain or desorption processes. Additionally, intentional and unintentional fires on landfills may release PFAS into the environment.

As not many publications from Europe were found, several papers from the USA and Canada were included in the calculation. The PFAS concentrations in landfill leachate in North America are according to the analysed literature higher than those in Europe which leads to an overall higher PFAS load for this calculation (compare Fuertes et al., (2017) to Solo-Gabriele et al., (2020)).

Table 8: Calculated mean and median concentrations for the PFAS groups in landfill leachate. Calculation includes data from North America which tends to have higher PFAS concentration in leachate. Data taken from 9 studies. All values in [ng/L] (landfills).

Substance	Mean	Median
ΣPFCA	3,310.45	2,242.50
ΣPFSA	1,516.81	541.70
ΣPFPA	3.18	0.00
ΣPrecursors	2,630.21	778.05
Total PFAS	6,816.89	3,556.60

With the leachate generation factor from Brennan et al. (2016) (0.2 – 1 m³ leachate generated per tonne of waste landfilled per year) and the total amount of landfilled waste in 2018 (838,861,071 tonnes), the leachate generated in the EU from the waste landfilled per year can be calculated.

Table 9: Calculated total PFAS freights based on the available Eurostat data (landfills). All values in [kg]

PFAS freight mean min	PFAS freight mean max	PFAS freight median min	PFAS freight median max
1,144	5,718	597	2,983

³ https://echa.europa.eu/documents/10162/13632/r18_v2_final_en.pdf

4.3 Incineration

The incineration of PFAS containing waste is currently seen as the most effective treatment option for destroying PFAS. According to current literature temperatures of at least 900 °C are needed for the effective destruction of PFAS, however higher temperatures are recommended (Geertinger et al., 2019).

According to the industrial emission directive⁴ incinerators are required to operate at temperatures of at least 850 °C and a residence time of at least 2 seconds. For hazardous waste incinerators temperatures of at least 1,100 °C need to be reached.

In general, there is a lack of quantitative data on the F-gas emission from the incineration of PFAS containing waste. Most publications conclude that only very low amounts of PFAS can be found in the flue gas and that the PFAS are destroyed >99,95 % but do not provide data on F-gases (Geertinger et al., 2019; Ministry of Japan, 2013; Taylor et al., 2014). Going forward it is important to obtain/generate quantitative data on the formation of F-gases under municipal waste incineration and hazardous waste incineration conditions in order to accurately assess the possible emissions arising from the incineration of PFAS containing wastes. The formation of these greenhouse gases may also prove difficult to solve, as the shortest fluorinated gas tetrafluoromethane CF₄ requires temperatures of 1,400 °C for its complete destruction, further proving the need for high-temperature incineration of PFAS-contaminated waste (Tsang et al., 1998; US EPA, 2020).

In the following table the degradation products from the incineration of PFAS found in literature are listed (García et al., 2007; Geertinger et al., 2019; Huber et al., 2009).

Table 10: End products of the thermal degradation of reported PFAS compounds

Starting compound	End products
Fluoropolymers	CO ₂ , CF ₄ , C ₂ F ₆ , CHF ₃ , C ₃ F ₆ , CClF ₃ , C ₄ F ₈ , C ₂ Cl ₃ F ₃ , HF, Trifluoroacetic acid and other perfluorinated gases.
PFOS	CF ₄ , C ₂ F ₆ , CHF ₃ , C ₂ H ₂ F ₂ , HF
PTFE	CO ₂ , CO, CF ₄ , C ₂ F ₆ , C ₃ F ₆ , C ₂ F ₄ , and other fluorinated compounds

According to the PRTR⁵ there were 472 non-hazardous waste incinerators in Europe in 2017. The Confederation of European Waste-to-Energy Plants (CEWEP) reported 492 waste-to-energy (WtE) plants which treated a total of 96 million tonnes of waste thermally in 2018 (CEWEP, 2021). According to the waste incineration (WI) BREF⁶ there were 470 municipal solid waste incinerators in Europe in 2019 with a total capacity of 87.44 million tonnes per year. These plants however are only required to operate at 850°C and as such may not be suitable for the adequate destruction of PFAS-containing waste. As the literature indicates temperature of at least 1,100°C are needed to effectively destroy PFAS, hazardous waste incinerators may be better suited for their destruction. The WI BREF reported 121 hazardous waste incinerators in Europe in 2019 with a total capacity of 6.75 million tonnes of waste per year, however the exact incineration conditions are unknown. As such, the complete destruction of the containing PFAS may not be guaranteed as short-chain

⁴ DIRECTIVE 2010/75/EU OF THE EUROPEAN PARLIAMENT AND OF THE COUNCIL of 24 November 2010 on industrial emissions

⁵ <https://prtr.eea.europa.eu/#/industrialactivity> Sector 5.(b) reference year 2017

⁶ WI BREF 2019: https://eippcb.jrc.ec.europa.eu/sites/default/files/2020-01/JRC118637_WI_Bref_2019_published_0.pdf

fluorinated gases may be formed if temperatures are too low. The following table summarises this information.

Table 11: Summary of information on the number and capacity of European waste incinerators

Source	Number of non-hazardous incinerators	Number of hazardous waste incinerators	Capacity/incinerated waste [million tonnes/year]
PRTR	472		
CEWEP	492		96
WI-BREF	470		87.44
WI-BREF		121	6.75

For the complete destruction without side products temperatures above 1,400°C are needed which can be reached in cement kilns. Hereby temperatures can reach up to 1,800°C suitable for the destruction of PFAS.

Wang et al. (2015) indicate, that the addition of calcium hydroxide can catalyse the defluorination process. At temperatures of 900°C, this method showed high transformation rates, indicated by the formation of calcium fluoride.

Although PFAS can rarely be found in the flue gas and no quantitative data is available, some publications reported the occurrence of PFAS in the fly and bottom ash of representative waste incinerators, indicating that the PFAS are not destroyed 100 % at current times. Additionally, one publication also analysed PFAS in fly ash (Sandblom, 2014), however no fly ash data was available for Europe. Instead a fly ash generation factor based on the total incinerated waste was used. Abis et al. (2020) reports, that 3% of the incinerated material becomes fly ash and BiPRO (2005) states, that 2.25% of the incinerated material becomes fly ash. Ultimately the 3% from Abis et al. (2020) was chosen to reflect a worst case scenario. The data generated for fly ash should however be analysed with care, as the data is based on only one publication and data set.

From this data, an emission value can be calculated to compare the PFAS occurrence in bottom ash to the landfill and WWTP data. Three publications from Sweden and the Netherlands were identified resulting in the following mean and median values (Rijkswaterstaat, 2020; Sandblom, 2014; Wohlin, 2020):

Table 12: Calculated mean and median concentrations for the PFAS groups in incineration bottom ash and fly ash. Data taken from three studies. n.A.: not analysed. All values in [pg/g] (incineration).

Substance	Mean (bottom ash)	Median (bottom ash)	Mean (fly ash)
ΣPFCA	811.55	43.10	8,981
ΣPFSA	312.18	0.00	1,805
ΣPFPA	n.A.	n.A.	n.A.
ΣPrecursors	578.26	0.00	n.A.
Total PFAS	1,663.44	83.60	10,786

Table 13: Amount of bottom ash from Blasenbauer et al. (2020) and incinerated waste in 2018 in Europe as well as the amount of fly ash generated from that waste. A generation factor of 3% for the fly ash was used from Abis et al. (2020).

Amount of bottom ash [t]	Amount of incinerated waste 2018 [t]	Amount of generated fly ash [t]
15,323,000	144,076,711	4,322,301

With this data the amount of PFAS in bottom and fly ash can be calculated.

Table 14: Calculated total PFAS amounts in incinerator bottom and fly ash. All values in [kg]

Mean PFAS amount (bottom ash)	Median PFAS amount (bottom ash)	Mean PFAS amount (fly ash)
25.5	1.3	46.6

While most bottom and fly ash is landfilled in Europe it can also be recycled for example in pavement or highway foundation (Blasenbauer et al., 2020). Fly ash is often used in cement, concrete and gypsum as well as restoration and filling material in open cast mines, quarries and pits.

4.4 Land application/composting

Organic waste can be further reused via composting or through the application on land. In the case of sewage sludge, 50 % is reused through these two methods, however these methods are not suitable for the destruction of PFAS. As such it is assumed, that through the use as compost and other land applications of organic waste, PFAS are not significantly destroyed in reasonable time frames, in contrast they can be considered as directly released to the environment.

4.5 Recycling

Europe is striving to achieve a circular economy and to increase its circular material use rate. Products from recycled materials are only possible if substances of concern, such as PFAS, are avoided in products as much as possible. This is usually not the case in the typical recycling processes of the considered waste streams. Hence, PFAS are often maintained in the economic cycle and may pose an obstacle to produce safe products from recycled materials.

4.6 Waste transfer stations

Waste transfer stations are generally used to sort, crush and bulk waste in order to enable an efficient waste packing for further transport. During this process liquids are usually decanted into a separate container (WI-BREF, 2018). During the sorting and crushing process liquids may be spilled from the waste and can end up in the leachate from the waste transfer station. It has been shown in literature, that the amount of PFAS in the leachate from waste transfer stations can reach levels of up to 46 µg/L (Wang et al., 2020). This study was performed in China and may not be representative of the European waste transfer stations, however it indicates the risk arising from these transfer stations (Rijkswaterstaat, 2020).

According to the European Waste Management Association (FEAD) there are 2,400 recycling and sorting centers in Europe (FEAD, 2021). It is unclear how many of these are waste transfer stations. The actual number of waste transfer stations in Europe is unknown.

Comparison

The calculated freights and emission values can be compared and the contribution of each waste treatment option to the total PFAS freight determined.

Table 15: Comparison of the calculated median freights for the WWTP, landfills and incinerator bottom ash for the EEA on a yearly basis. All values in [kg]

Median WWTP effluent + sludge	Median landfill min	Median landfill max	Median incineration ash	Total min	Total max
9,884	597	2,983	49	10,530	12,916

Table 16: Comparison of the calculated yearly per capita emission values for the EEA on a yearly basis. All values in [mg / (person / year)]

Median WWTP effluent + sludge	Median landfill min	Median landfill max	Median incineration ash	Total min	Total max
21.9	1.2	6.60	0.108	23	29

Table 17: Comparison of the contribution of the regarded waste treatment options to the total PFAS freight. All values based on a maximum amount of leachate generated. All values in [%] and rounded

Total contribution effluent	Total contribution landfills	Total contribution incineration
77	23	0.38

4.7 Conclusion

The WWTP has the highest contribution to the PFAS emission ranging from 77 – 94 % followed by the landfills with 6 - 23 %. While the WWTP effluent represents a direct PFAS emission into the environment the landfill leachate is in some cases treated in a WWTP in Europe (Brennan et al., 2016). As such, part of the landfill leachate is also included in the WWTP emissions.

The contribution of the incinerator bottom ash is below 0.5 % indicating that this is the best treatment options for the destruction of PFAS. However, the incineration data only takes the ash into account disregarding the emissions from flue gas. As such the contribution from the waste incineration may be higher.

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