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THE PRESENCE OF PFAS IN WASTES AND RELATED IMPLICATIONS ON THE CURRENT AND PROPOSED EUROPEAN REGULATORY FRAMEWORK: A SYSTEMATIC CRITICAL REVIEW

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ABSTRACT

Nowadays the use of PFAS is widespread in products in modern society and their exposure can occur directly through emissions such as dietary intake, water intake, air inhalation, and skin contact. Additionally, exposure can happen indirectly through the environmental release and degradation of their precursors. To fully understand the potential for life cycle emissions, it is necessary to consider the waste stage, as it is an integral part of a substance life cycle, whether PFAS exists in its pure form as part of a mixture or within an article. Overall, knowledge about the impact of PFAS on current and future waste management remains limited. Therefore, this study conducted a critical analysis of the presence of PFAS in relevant waste streams (plastic; metals; textile and leather; paper and cardboards). It also discussed how this presence could influence waste management, taking into account ongoing updates of the legal framework, with particular attention to proposed new provisions regarding their restriction in the REACH regulation. Within the discussed limits of the critical review, only a very small number of outliers were found to exceed the considered limit of 25 ppb for each material category. The percentage of exceedances ranged from nearly 1% (PFOS measurements in paper and cardboard waste) to 8% ("Other PFAS" in textiles and leather waste). Regarding the analytical methods and current limits identified, a pragmatic solution is suggested. This solution combines "not targeted" and "targeted" methodologies in a stepwise procedure, building upon the OECD definition of PFAS.

1. INTRODUCTION

The term "Per- and polyfluoroalkyl substances" (PFAS) identifies a class of synthetic compounds that have attracted much public attention since the late 1990s and early 2000s, when the hazards, the ubiquitous occurrence, and the persistence of two PFAS, perfluorooctanoic acid (PFOA) and perfluorooctane sulfonic acid (PFOS), started to be reported and recognized.

Nowadays the use of PFAS is widespread in products for the modern society. This includes for example applications in personal care products and cosmetics, ski waxes, firefighting foams, durable water and stain repellence in textiles, food contact materials, medical devices, pharmaceuticals, laboratory supplies, equipment, and instrumentation, perfluorosulfonic membranes, used in a wide range of chemical synthesis and separation operations and in analytical instrumentation (Cousin et al., 2019). Exposure to these substances may arise directly from emissions, such as dietary intake, water intake, air inhalation, and skin contact, or indirectly via the environmental release and degradation of their precursors.

As PFAS are chemicals with diverse molecular structures and physical, chemical and biological properties, the regulatory framework highly recommends that such diversity should be properly recognized.

The basic structure of a PFAS consists of a carbon chain with substituted fluorine atoms replacing hydro-

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gen atoms on the chain, and with different categories of PFAS chemicals possessing different substituents and functional groups within (e.g., ethers) or terminal to the chain (Williams et al., 2022). From this rough structural explanation different definitions have been made to encompass broad working scopes or satisfy narrower regulatory guidelines.

In one of the earliest attempts, Buck et al. (Buck et al., 2011) defined PFAS as aliphatic substances that "contain one or more carbon atoms on which all of the hydrogen substituents (present in the nonfluorinated analogues from which they are notionally derived) have been replaced by fluorine atoms, in such a manner that they contain the perfluoroalkyl moiety (– C_nF_{2n+1} –)".

According to the revised PFAS definition by OECD (2021), "PFAS are defined as fluorinated substances that contain at least one fully fluorinated methyl or methylene carbon atom (without any H/Cl/Br/I atom attached to it), i.e., with a few noted exceptions, any chemical with at least a perfluorinated methyl group (–CF_3) or a perfluorinated methylene group (–CF_2–) is a PFAS". The "noted exceptions" refer to a carbon atom with a H/Cl/Br/I atom attached to it (Wang et al., 2021). Hereafter, it is referred to as the OECD 2021 definition.

Finally, a new definition by Gaines et al. (Gaines et al., 2023) combined a chemical structure with the percentage of fluorine. Here PFAS are defined as substances based on four substructures (Figure 1) along with any structures where the molecular formula is at least 30% fluorine by atom count.

A 2015 study reported that more than 3,000 PFAS were on the global market for commercial use (Swedish Chemicals Agency, 2015). In 2018, the OECD found 4730 different CAS numbers for PFAS (OECD, 2018). The United States Environmental Protection Agency's (USEPA) toxicity database, DSSTox, lists 14735 unique PFAS chemical compounds (USEPA, 2022; Gaines et al., 2023).

The abovementioned synthetic and representative statistics demonstrate the large variation in the numbers of chemicals that could be considered as a PFAS, based on the different definitions, including the variations in chemical fluorination (Gaines et al., 2023; Williams et al., 2022).

Figure 2 shows the total number of substances of PFAS according to different definitions compared with the number of substances for which regulation limits are existing (see next paragraph).

The waste stage is part of the life cycle of a substance in a mixture or in an article. Therefore, its distribution via the supply chain including service-life of articles and waste stage as an emission source with its associated risks should be taken into account.

The understanding of waste management (recycling, landfill, incineration, or composting) and the fate of PFAS

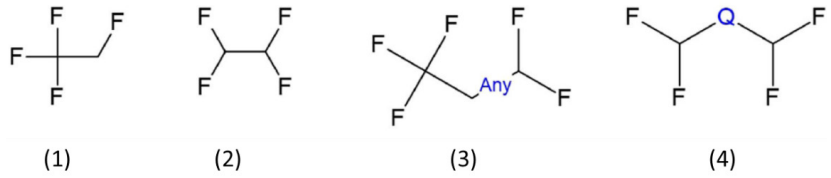

FIGURE 1: Substructures used in combination with percentage of fluorine. In Substructure 3 "any" indicate the type of bond between the two carbons. For Substructure (4) the heteroatom Q can be B, O, N, P, S, or Si.

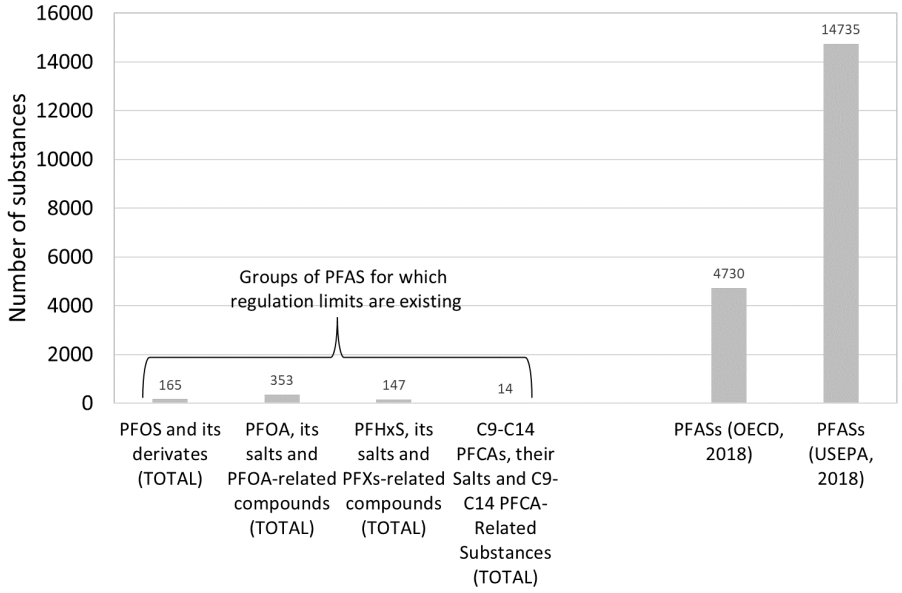

FIGURE 2: Number of substances for: PFOS and its derivates (Department for Environment, Food and Rural Affairs, 2015); PFOA, its salts and PFOA-related compounds (UNEP, 2022); PFHxS, its salts and PFHxS -related compounds (UNEP, 2019); C9-C14 PFCAs, their Salts and C9-C14 PFCA-Related Substances (ECHA, 2017); PFAS according to OECD classification (2018) and EPA classification (EPA, 2018).

in the waste stage is important if the aim is to understand the potential of complete life cycle emissions.

The EU waste classification system is primarily governed by the Waste Framework Directive (2008/98/EC), which sets out the general principles for managing waste in the EU. It establishes a harmonized waste classification system known as the European Waste Catalogue (EWC). The EWC provides codes and descriptions for different types of waste, allowing for standardized identification and classification of waste materials. PFAS, which are a group of man-made chemicals used in various industrial and consumer products, do not have specific codes or classifications within the EWC. However, depending on the specific characteristics and properties of the waste containing PFAS, it may fall under existing waste codes that pertain to related substances or materials. For example: if the waste containing PFAS exhibits hazardous properties according to the EU's Hazardous Waste Directive (Commission regulation (EU) No 1357/2014; 2017/997/EU), it would be classified as hazardous waste. Hazardous waste is further categorized based on specific hazardous properties, such as being toxic, flammable, or corrosive. PFAS-containing waste that displays any of these properties would be today classified accordingly.

Consequently, when it comes to the disposal or recovery of waste containing PFAS, several considerations come into play. These may include Hazardous Waste Classification that generally requires special handling, treatment, and disposal procedures.

Currently, the recycling of waste containing PFAS can be problematic due to the persistent characteristics and high bioaccumulation potential of these compounds. PFAS can be difficult to remove completely from materials and can persist into the recycling cycle, contaminating other materials or recycling processes.

Incineration is one method used for the treatment of certain wastes, including hazardous waste. Waste containing PFAS may require specific considerations when incinerated, as some PFAS compounds may release harmful by-products if inadequately burned. Proper incineration facilities equipped to handle PFAS-containing waste and prevent or minimise the release of PFAS into the environment are necessary.

Certain waste containing PFAS may be disposed of in regulated landfills designed to handle hazardous waste. Additionally, the long-term management and monitoring of landfill sites may be required due to the persistence and potential environmental risks associated with PFAS. Dedicated acceptance criteria for PFAS should be defined accordingly.

Research and development are ongoing to identify effective treatment technologies for PFAS-containing waste. Advanced treatment methods such as thermal desorption, chemical degradation, or adsorption techniques may be used to remove or reduce PFAS concentrations in the waste stream. In some cases, additional and final treatments can be required specifically for the residues generated where PFAS may be concentrated.

The End of Waste (EoW) procedure, is a regulatory process that establishes when certain waste materials cease to be classified as waste and can be considered as products or secondary raw materials. The EoW procedure aims to promote recycling and resource recovery by providing clarity on when waste materials have undergone sufficient treatment or processing to meet specific quality standards and no longer pose significant risks to human health and the environment. The specific criteria and procedures for determining the EoW status can vary between countries or regions. However, there are typically common elements involved, such as:

- quality standards: EoW criteria define the specific quality parameters that waste materials must meet to be considered as products or secondary raw materials. These parameters may include chemical composition, physical properties, contaminants levels, or other relevant characteristics;
- verification process: the verification process involves assessing and documenting that the waste material meets the established quality standards. This is typically done through testing, analysis, and documentation, either by the waste generator or a third-party certification body;
- regulatory approval: once the waste material has undergone the necessary treatment and meets the EoW criteria, it may require regulatory approval or acknowledgment to be officially recognized as a non-waste product or secondary raw material. This approval process may involve submitting applications, providing evidence of compliance with the criteria, and obtaining permits or certifications;
- market acceptance: Even if a waste material meets the EoW criteria, its acceptance in the market as a product or secondary raw material depends on factors such as demand, market conditions, and specific industry requirements.

The EoW procedure is important as it incentivizes the recycling and recovery of waste materials, reduces reliance on virgin resources and promotes a circular economy. Indeed, a new regulation on PFAS contents in substances, matrixes and articles could limit the circularity of materials.

There is another relevant aspect to consider: the presence of PFAS in the waste streams will remain for a long while; this is due to:

- nowadays it is neither practical nor reasonable to ban all uses of PFAS in one step, because some specific applications may serve a critical role for which alternatives currently do not exist (Cousin et al., 2019). This is the case for example of the use of PFAS for occupational protective clothing. In other words, the use of some PFAS is still considered "essential", meaning that they are "necessary for health, safety or is critical for the functioning of society" and that "there are no available technically and economically feasible alternatives" (United Nations, 1987);
- the long-life cycle of some products, such as laboratory supplies, equipment, and instrumentation, will determine the presence of PFAS in waste long after the time of

placing on the market of those products;

- in some cases, such as landfill mining, wastes potentially contaminated by PFAS, originally disposed of in a landfill as their final destination, can be newly considered for recovery;
- leachates from landfills can be contaminated by PFAS for a long period (Zhang et al., 2023; Lu et al., 2023).

The general objective of this study is to deepen the knowledge of the presence of PFAS in relevant waste streams for recycling issue, and to understand how this could influence current and future waste management and recycling practises, considering continuous updates of the relative legal framework, paying particular attention to the proposed new provisions on their restriction in the REACH regulation (BAuA et al, 2023a).

To do so, a systematic critical review of the scientific literature was performed to collect PFAS concentration ranges in waste items categorized within relevant four waste material categories (i.e., plastic; paper and cardboard; textile and leather; metals), as presented and discussed in both peer-reviewed articles already published and other significant documents. The collected results were compared with regulation limits to ease the understanding of their impacts on waste management practices.

2. THE EU REGULATORY FRAMEWORK

Here, the regulation analysis is concerned with the presence of PFAS in solid wastes and as a substance, whether on their own or in mixtures, or in articles. In fact, there is a strong connection between waste and manufacturing sectors because they both place those substances on the market on their own, in mixtures or in articles. At the end-of-life stage, products become waste and this can return on the market after a recycling process as EoW. Therefore, specific limits, provisions, restrictions, etc. on PFAS in one sector will have consequences in the other sector. Figure 3 graphically depicts the established concentration limits which have or may have impacts on waste management, while Table S1 in the Supplementary Material provides a comprehensive list of regulation limits or proposed limits on PFAS that currently impact or may potentially affect waste management.

In the section below, the state of art, derived from (I) the Stockholm Convention on Persistent Organic Pollutants (POPs), (II) the EU POPs Regulation, (III) Regulation on classification, labelling and packaging of substances and mixtures (CLP), (IV) REACH Regulation, (V) classification of waste, (VI) specific provisions for disposal or recovery

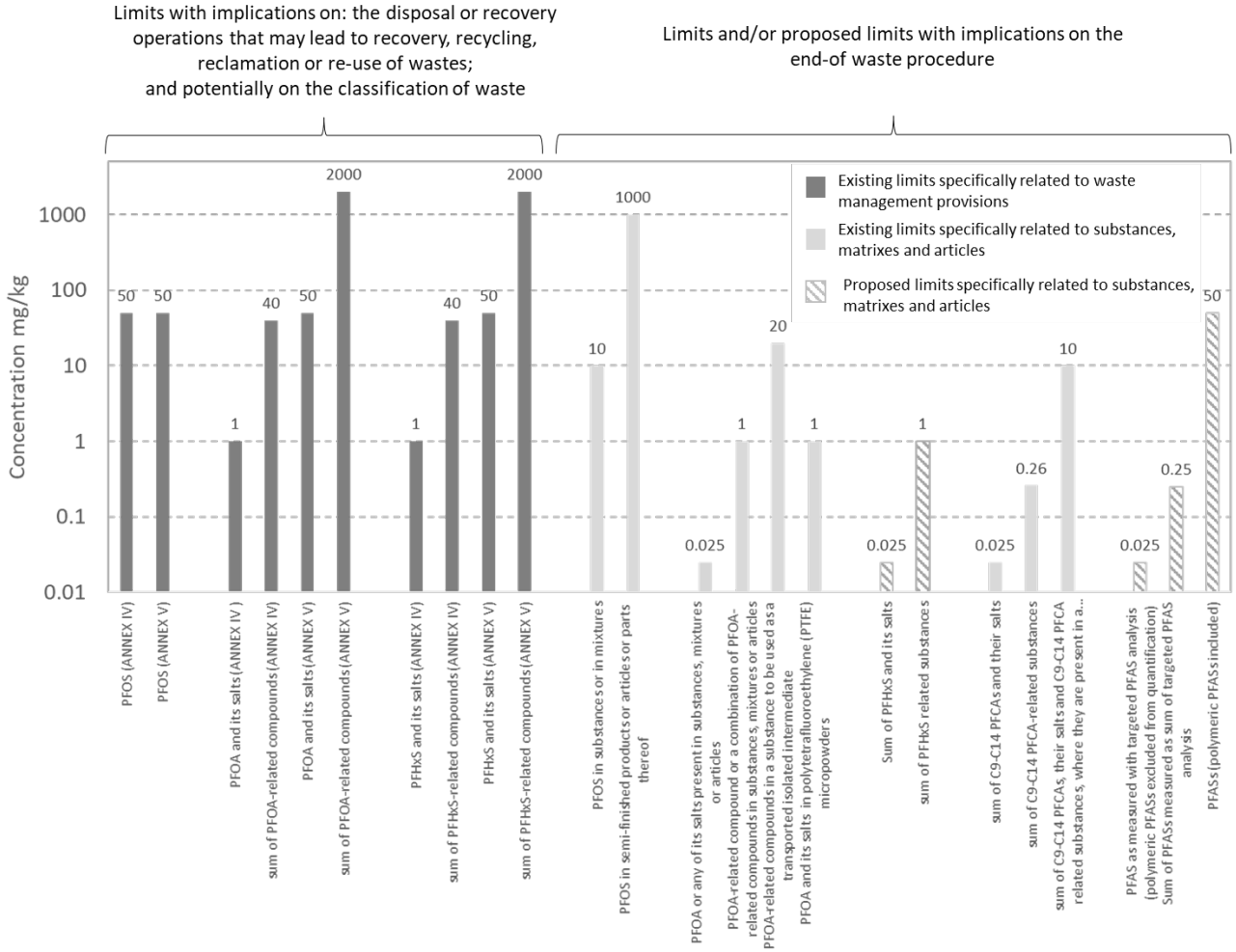

FIGURE 3: Graphical representation of limits on PFAS with implications on waste management.

operations of waste and (VII) the EoW procedure, is introduced and analysed.

Then, specific considerations regarding the application of the circular economy policy are introduced with respect to the recent restriction proposal under REACH from the national authorities of Denmark, Germany, the Netherlands, Norway, and Sweden (BAuA et al, 2023a). This is because this last proposal is the most conservative with consideration to all the PFAS.

2.1 The state of the art

Only few PFAS, PFOA, and PFOS, and PFHxS have been listed under the Stockholm Convention on Persistent Organic Pollutants (POPs) in Annex "A" or "B" (UN Environment Programme and Stockholm Convention, 2023). For chemicals listed in Annex "A" (PFOA and PFHxS), parties must take measures to eliminate their production and use, even if specific exemptions can be available. For chemicals listed in Annex "B" (PFOS), parties must take measures to restrict their production and use considering any applicable acceptable purposes and/or specific exemptions listed in the Annex.

Consequently, these chemicals are now restricted under the EU POPs Regulation (Regulation (EU) 2019/1021, as amended by Regulation (EU) 2022/2400).

Some other long-chain PFAS have a harmonised hazard classification under the Regulation on classification, labelling and packaging (CLP) of substances and mixtures.

Actually, REACH Regulation has included a PFAS group for restriction only for one case: C9-C14 PFCAs, their salts and C9-C14 PFCA- related substances. However, there are some proposals for other groups (see Table 1) for being assessed for a restriction under REACH.

For the purpose of this study, the recent restriction proposal under REACH from the national authorities of Denmark, Germany, the Netherlands, Norway and Sweden is relevant (BAuA et al, 2023a). Since the proposal considers all the PFAS according to OECD 2021 definition; it suggests a solution on a fundamental problem of the approach followed until now for PFAS in REACH: that the restriction or elimination is limited to individual substances or groups of closely related substances. Likewise, this proposal is in line with the recast of the Drinking Water Directive (Directive EU 2020/2184) which includes a grouping approach for all PFAS i.e., a limit of 0.5 µg/l for all PFAS.

The proposal further includes a combination of limits which can be detected in a solid matrix using different analytical approaches: targeted analysis regarding the limit of 25 ppb (0.025 mg/kg) for any potential PFAS (polymeric PFAS excluded from quantification); total oxidizable precursors (TOPs) regarding the limit of 250 ppb (0.25 mg/kg) for the sum of PFAS (polymeric PFAS excluded from quantification); non targeted method (total organic fluorine) regarding the limit of 50 ppm (50 mg/kg) for PFAS (polymeric PFAS included).

Concerning waste management only PFOA, and PFOS, and PFHxS have been identified in Annex IV and V of Regulation (EU) 2019/1021. The limits in Annex IV represent specific provisions in waste management such as the prohibitions of disposal or recovery operations that may lead to recovery, recycling, reclamation, or re-use of wastes with concentrations above these limits. The limits in Annex V represent the maximum concentration limits of substances listed in Annex IV allowed in specific waste streams to be otherwise dealt with in accordance with a method listed in Part 2 of Annex V (permanent storage in hazardous waste landfills or underground permanent storage (incl. salt mines) providing that competent authorities agreed).

For the classification of waste as hazardous, the EU Commission Decision 955 of 18 December 2014, amending Decision 2000/532/EC, states that wastes containing the first POPs indicated in the former POPs regulation (Regulation (EC) No 850/2004) exceeding the reported concentration limits listed in Annex IV shall be classified as hazardous. It is worth mentioning that in this list PFAS are not mentioned but amendments of the Directive 2008/98/EC or Decision 955 of 18 December 2014, modifying Decision 2000/532/EC, could introduce new concentration limits for PFAS affecting the classification of waste.

For the EoW procedure, a waste containing PFAS that achieves EoW status, the associated producer of this material, i.e., the person who places the material on the market for the first time after it ceases being waste, must ensure that the material meets any relevant requirements under REACH Regulation (EC) 1907/2006 and CLP Regulation (EC) 1272/2008. Therefore, provisions for restriction or elimination of PFAS in substances, mixtures, or articles must be applied also to materials from EoW procedure. This concept is clearly mentioned in the proposal from the national authorities of Denmark, Germany, the Netherlands, Norway, and Sweden is relevant (BAuA et al, 2023a). In their "Questions and Answers" the agencies affirmed that the "proposed restriction apply to articles regardless of whether they are made from virgin or recycled materials" (BAuA et al, 2023b).

Figure 4 shows a comprehensive overview of the most relevant regulations in the waste field where existing and proposed limits for PFAS could influence current and future waste management practises.

2.2 Implications on circular economy from the recent restriction proposal under REACH

This new proposal (BAuA et al, 2023a) could impact the circular economy. In fact, it must be considered that among the requirements to obtain the EoW status, the legislation in force provides that "the substance or object fulfils the technical requirements for the specific purposes and meets the existing legislation and standards applicable to products".

Therefore, if the proposal - as previously mentioned - will also apply to materials from a waste recycling process, since treatments for removing PFAS from waste are not currently available, they will not be destined for material recovery, but for disposal and, if possible, energy recovery.

This would create a conflict between two objectives; from one side the policies of the European Union require Member States to encourage the circular economy favouring as much as possible the recovery of waste (in particu-

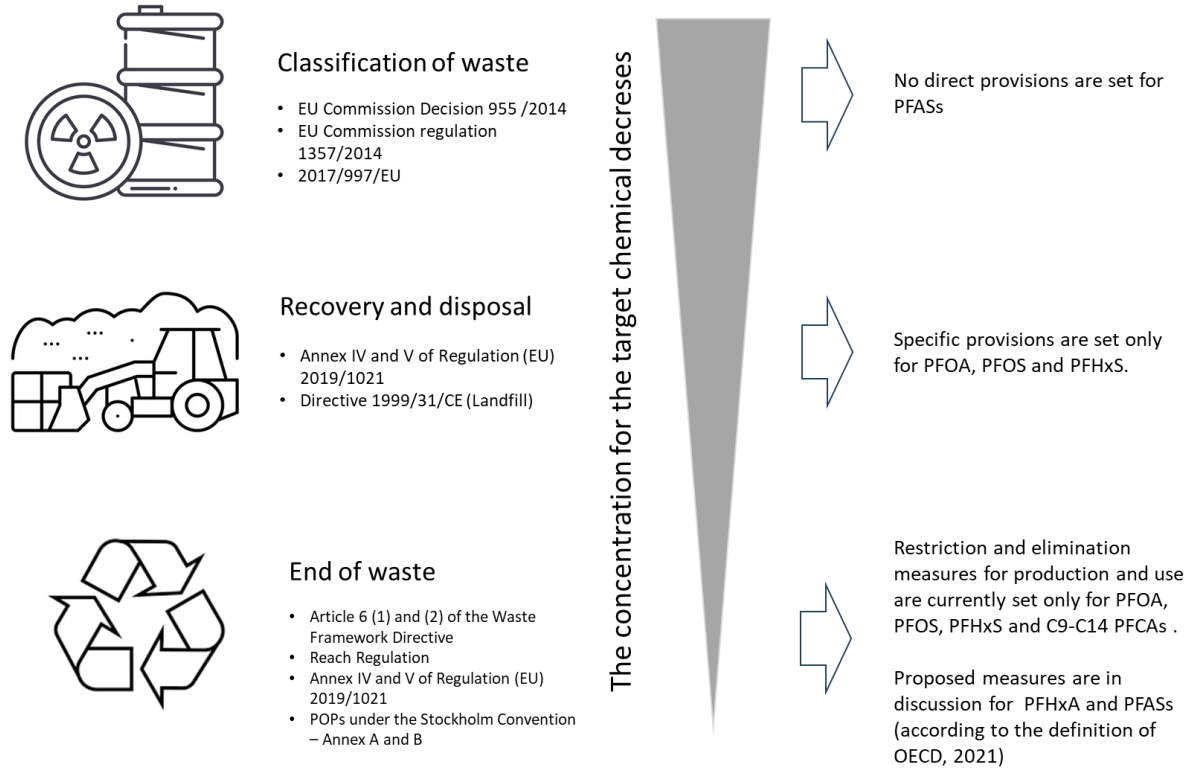

FIGURE 4: Graphical representation of the most relevant regulation applications in the waste field where PFAS can be interested.

lar of the textile and plastic sector), while on the other side they introduce new conservative limits of PFAS to prevent harm to human health and to the ecosystem which could limit the recovery of waste.

The legislator has the aim to resolve these conflicts considering also the conclusions of the EU Court of Justice (decision of 19 January 2023, case C-147/21) where two relevant principles are explicated: (I) the obligation of Member States to respect the principle of proportionality and (II) the burden of proving the existence of proportionality falls on those who invoke it, not on those who deny it. The aforementioned conclusions can be read as follows:

- *"53. it is for the national authorities, in each individual case, to demonstrate that the national legislation at issue satisfies the principle of proportionality, that is to say, that it is necessary to achieve the declared objective and that it could not be achieved by prohibitions or restrictions that are less extensive or have less effect on trade within the European Union. To that end, it is for those authorities to provide the necessary evidence to that effect. The reasons which may be invoked by a Member State by way of justification must thus be accompanied by an analysis of the appropriateness and proportionality of the restrictive measure adopted by that State, and by specific evidence substantiating its arguments (judgment of 23 December 2015, Scotch Whisky Association and Others, C-333/14, EU:C:2015:845, paragraphs 53 and 54).*
- *54. It follows that, where a national court examines national legislation in the light of the justification relating to the protection of the health and life of humans, under Article 36 TFEU, it is bound to examine objectively whether it may reasonably be concluded from the evidence submitted by the Member State concerned that the means chosen are appropriate for the attainment of the objectives pursued and whether it is possible to attain those objectives by measures that are less restrictive of the free movement of goods (see, to that effect, judgment of 23 December 2015, Scotch Whisky Association and Others, C-333/14, EU:C:2015:845, paragraph 59)."*

3. MATERIALS AND METHODS

The stepwise procedure for literature sources selection (Figure 5), was based on the so-called "Preferred Reporting Items for SRs and Meta-Analyses "(PRISMA) guidelines (Gurevitch et al., 2018; PRISMA, 2023). Documents identification was carried out in the electronic repositories of i) Elsevier Bibliographic Database (Scopus) and ii) Web of Science (WOS), by searching, in the Titles, Abstracts or Articles Keywords, the following keywords chains, one for each specific investigated waste stream:

- "PERFLUOROALKYL*" OR "POLYFLUOROALKYL*" OR "PFAS" AND "PLASTIC*" AND "WASTE*" AND NOT "WASTEWATER";
- "PERFLUOROALKYL*" OR "POLYFLUOROALKYL*" OR "PFAS" AND "PAPER* WASTE*" OR "CARDBOARD" OR "CELLULOSIC WASTE" AND "WASTE*" AND NOT "WASTEWATER";
- "PERFLUOROALKYL*" OR "POLYFLUOROALKYL*" OR "PFAS" AND "METAL* WASTE*" OR "FERROUS WASTE*" AND "WASTE* AND NOT WASTEWATER" OR "SCRAP*";

- "PERFLUOROALKYL*" OR "POLYFLUOROALKYL*" OR "PFAS" AND "TEXTILE*" OR "LEATHER" AND "WASTE*" AND NOT "WASTEWATER".

Documents were retrieved from the online databases according to the following criteria: electronic version of the articles is available; the language of the articles is English; and documents type was Article. Documents were excluded if consisting of Review Article, Proceeding Paper, Book Chapters, Letters etc. Furthermore, the documents cited in the literature reviews (identified but excluded from the scope of the current analysis according to the PRISMA guidelines) were further included as external sources.

Identified documents underwent a screening and those with abstracts not providing a description or a discussion of quantitative data about PFAS concentrations were filtered out.

A futher full-text reading of the screened documents was conducted toto include the papers considered eligible according to the scope of the analysis. In this phase, papers not showing quantitative data derived from analytical measurements, or those presenting PFAS concentration on substrates not relevant for this study (e.g., landfill leachates) were considered not eligible. Also, those items reporting data as mass per unit area (e.g, μg m^{-2}) were considered not eligible to ease direct comparison of PFAS concentrations and regulation limits.

Eligibility was extended to those documents, mostly derived from three identified but excluded review articles (Barhoumi et al., 2022; Bulson et al., 2023; Coffin et al., 2023), even if showing analytical data measured in products (i.e., still not yet "waste"), since they were considered as significant for the assessment of the presence of PFAS in the studied waste streams, as suggested by the review articles themselves. In other words, the so-included documents were assumed characterizing products representative of "waste-to-be", thus ultimately influencing their management practices (i.e., characterization and classification, treatment, disposal or potential recycling to achieve EoW status).

The final list of the documents considered eligible is reported in the Supplementary Materials.

Two reviewers (BG & GG) independently extracted data from each original publication. Data extraction performed on the eligible documents allowed the retrival of information regarding: the material streams of the analysed sample; the specific waste or product item of the analysed sample; the geographical origin of the sample; the year of sampling; the concentration values of the quantified parameters (i.e., single substance concentrations or sums of them); the analytical methods used for the analytical quantification; where available, the value of the Limit of Detection/Limit of Quantification (LOD/LOQ); information on possible specific treatments or analytical procedures performed on the sample that can be useful to include/exclude the single entries from the statistical analysis.

When concentration data were reported lower than the LOD or LOQ, they were extracted as equal to the reported LOD or LOQ value divided by two (EFSA, 2012; SNPA, 2021). Values of LOD or LOQ reported higher than 25 ppb (0.025 mg/kg) were excluded from the data extraction since they could not be compared with all the regulation limits (both established or proposed). Where LOQ or LOD values were not reported, an average value of LOD or LOQ values available in other documents were extracted, preferentially for each single parameter, or groups of parameters (e.g., FTOHs) where this latter approach could not be applied.

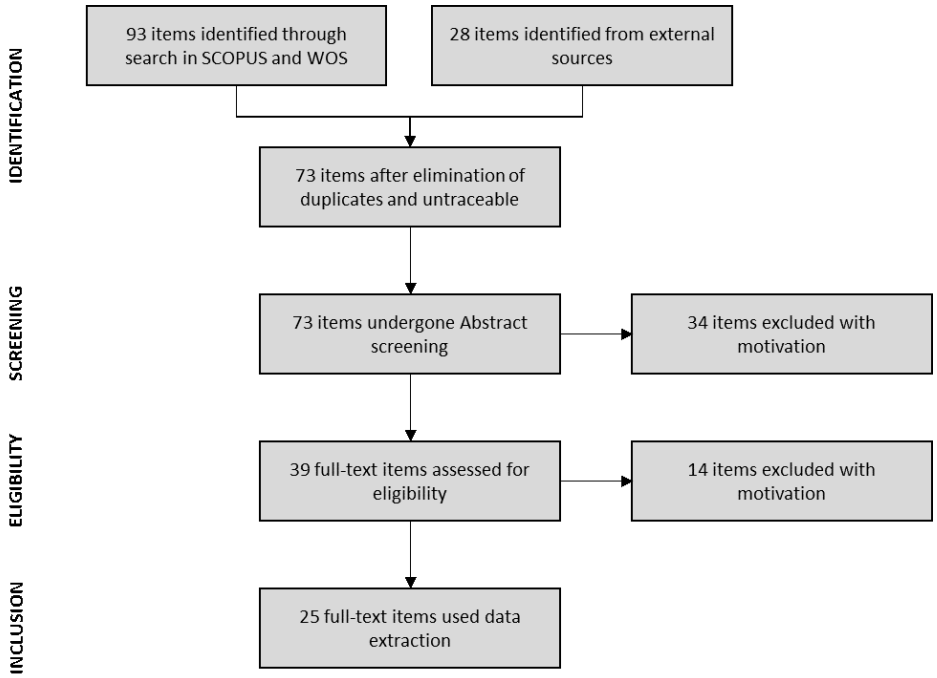

FIGURE 5: Flow diagram resuming the stepwise procedure followed for the performance of the systematic critical review.

Overall, a total of 5795 single concentration values from 25 papers were extracted. Among them, 4836 were used for descriptive statistical analysis and comparison with regulation limits; the exclusions included data which could not be organized consistently to the aim of the statistics such as sums of concentrations of different parameters (e.g., ∑PFAS, ∑PFCAs, ∑FTOHs, etc.).

Likewise, a specific comparison was performed considering material-specific PFAAs concentration data between two relevant analytical approaches. In this way it was possible to assess the influence of a specific sample preparation method (i.e., TOP assay) on the results of the quantification step.

These concentration values were organised in categories according to the detected substances and the waste stream of origin (i.e., paper and cardboard waste, textile and leather waste, plastic waste and metal waste) and to the measured parameters.

The analysis performed on categorized data included descriptive statistics (i.e., median, min-max range, Q1 and Q3 values, and 5%-95% percentiles) and graphical representation of data through box plots. The results of this latter allowed to compare the collected concentration data with the regulation limits and thus calculate the percentage of limit exceedances for each considered category.

For the purpose of this study, the comparison relied on the lowest limit value which could be applied to all single targeted PFAS i.e., 25 ppb (Figure 3).

3.1 Waste streams

Only the following waste categories were considered in the scope of this study because of the known potential presence of PFAS in these streams: plastic; metals; textile and leather; paper and cardboards.

The Table 1 shows the percentages of the above-mentioned waste streams on the total amount of waste excluding major mineral waste generated in EU for 2020.

Furthermore, the specific items (plastic straws, carpets, automotive shredder residues, plastic scraps from end-life vehicles, etc.), on which the targeted substances were measured, were recorded.

TABLE 1: Waste generation (Hazardous and non-hazardous) in 2020 for European Union - 27 countries. (reference: https://ec.europa.eu/eurostat/databrowser/view/env_wasgen/default/table?lang=en).

Waste stream	tonne	%
Textile [(1)]	1,950,000	0.25%
Plastic	19,030,000	2.45%
Paper and cardboard	43,490,000	5.60%
Metal [(2)]	82,180,000	10.59%
Other wastes [(3)]	629,640,000	81.11%
Total waste excluding major mineral wastes	776,290,000	100.00%

[(1)] including leather
[(2)] sum of metal wastes, ferrous, metal wastes, non-ferrous and metal wastes, mixed ferrous and non-ferrous
[(3)] sum of sludges and liquid waste from waste treatment, sorting residues, chemical and medical wastes, glass wastes, rubber wastes, wood wastes, equipment, animal and vegetal wastes, household and similar wastes excluding major mineral wastes

3.2 Detected substances

The PFAS measured in the selected papers have been organized according to the definition laid down by the OECD, (2021), as graphically represented in Figure 6. Different subgroups of PFAS have been associated with the class of chemicals as explicitly mentioned in existing or proposed regulations.

Finally, considering all the available data, concentration values were grouped into the following 5 categories for each considered material:

1. PFOS, i.e., values relative to single sample concentrations of PFOS;
2. PFOA, i.e., values relative to single sample concentrations of PFOA;
3. PFHxS, i.e., values relative to single sample concentrations of PFHxS;
4. C9-C14 PFCAs, i.e., values relative to single sample concentrations of PFNA, PFDA, PFUnDA, PFDoDA, PFTrDA and PFTeDA;
5. other, i.e., values corresponding to single sample concentrations of PFAS molecules not listed in the previous categories, but considered in the updated OECD definition, i.e., including non-regulated PFAAs together with the so-called "PFAAs precursors".

3.3 Analytical methods

From a testing point of view, PFAS analysis is carried out primarily through extraction and purification followed by liquid chromatography coupled with tandem mass spectrometry. As a "targeted" analytical procedure, its application is limited to the quantification of the concentration for a fixed set of parameters. Thus, it doesn't provide a measure of either the magnitude of the "pool" of PFAS that may exist, nor the potential for targeted PFAS formation due to natural transformation of precursor compounds. In this class, the following standard methods are identified:

- ASTM D 7968-17A: 2017 "Determination of Polyfluorinated Compounds in Soil by Liquid Chromatography Tandem Mass Spectrometry (LC/ MS/MS)";
- EPA 3550 C: 2007 "Ultrasonic Extraction" + EPA 8327: 2021 "per- and polyfluoroalkyl substances (PFAS) by liquid chromatography/tandem mass spectrometry (LC/MS/MS)".

In these methods, targeted analytes include a range of PFCAs and PFSAs as well as a smaller number of PFAS precursors and intermediate transformation products (e.g., partially oxidized PFAS precursors). Methods can be modified to broaden the set of quantified analytes to a limit defined by i) the commercial availability of appropriately certified reference materials and ii) the compatibility with existing types of analytical equipment. Currently, standards are available for almost 40 substances, as reported in Figure 7 (Ateia et al., 2023). However, the number of PFAS potentially analysed via targeted analysis will likely increase over time.

In this context, the Total Oxidizable Precursors (TOP) assay was developed to quantify the presence of targeta-

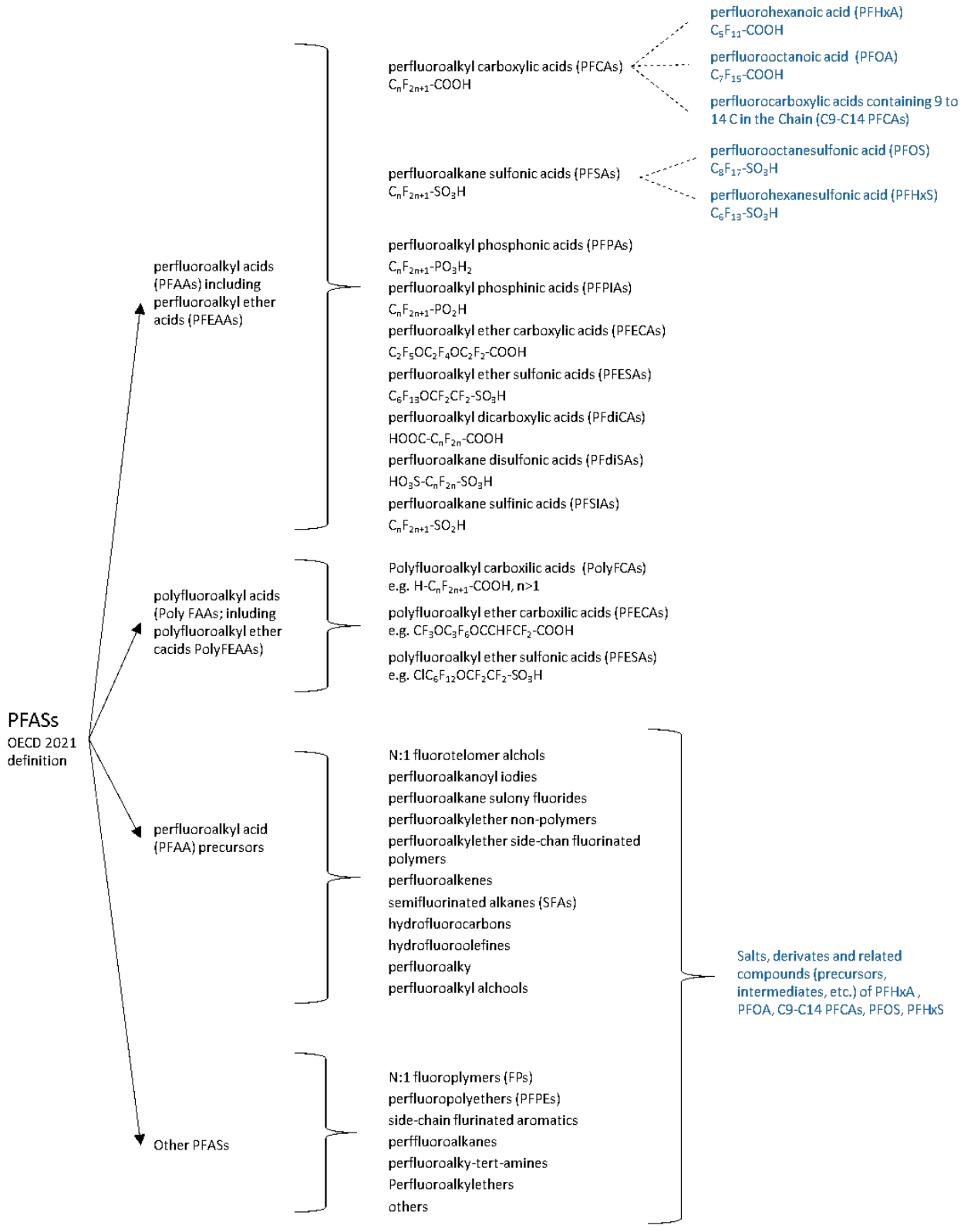

FIGURE 6: A comprehensive overview of PFAS groups associated with regulated classes of PFAS (based on OECD, 2021).

ble PFAS precursors. Here, the term "PFAS precursor" refers to PFAS with the potential for oxidative transformation resulting in terminal products such as PFAAs (Figure 6). The TOP assay allows the conversion of oxidizable PFAS precursors into PFAAs, which are then measured using a targeted PFAS analytical method. One of the most relevant benefits of this method is its compatibility with the same analytical instrumentation utilized in targeted analysis.

More recently, there has been a focus in the industry to develop and validate lower cost alternatives that also

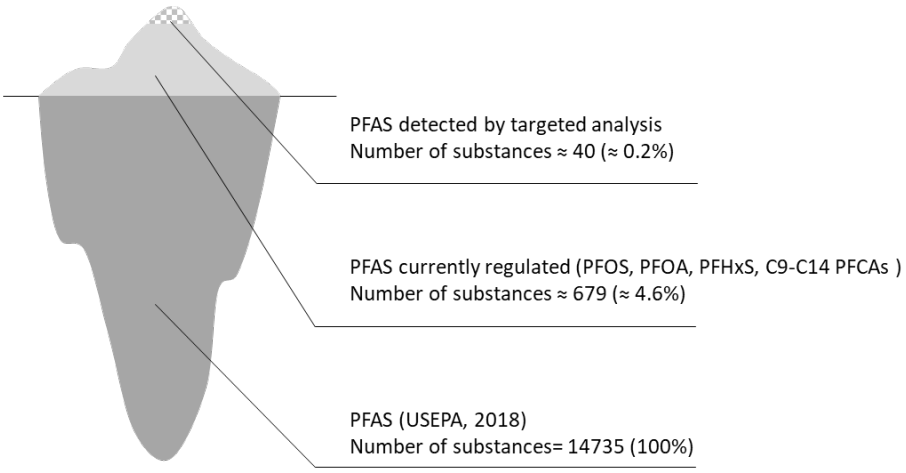

FIGURE 7: The "iceberg" knowledge of PFAS from a regulation perspective. Modified from Ateia et al., (2023).

provide a more comprehensive measure of total PFAS presence. This has resulted in several "non-targeted" methods able to quantify fluorine concentration, in terms of Total Fluorine (TF), Adsorbable Organic Fluorine (AOF), Extractable Organic Fluorine (EOF) and total organic fluorine (TOF). These methods are often used to screen out samples showing very low fluorine content from further targeted analysis. However, these methods do not identify the molecular configuration and may therefore deliver non-realistic estimations of PFAS concentration (Schwartz-Narbonne et al., 2023).

4. RESULTS AND DISCUSSION

The resulting box plots showing the distribution of concentration data extracted from the eligible papers are depicted in Figures 8, 9, 10 and 11. Further, the entire dataset is available in Supplementary Materials together with detailed descriptive statistics for each waste flow and parameter category.

4.1 Considerations on the waste streams investigated

Considering sample sizes of each considered material category, most eligible documents focused on the paper and cardboard waste (17 papers) and textile and leather waste (seven papers), with a total of 2680 and 1488 observations, respectively. The plastic waste category was investigated by a lower number of documents (six papers) resulting in 613 observations. Instead, metal waste was the category less investigated by the documents considered for the data extraction, with only two papers covering the category and a significantly smaller amount of extracted data (55). Looking at the analyzed waste items, measurements for paper and cardboard waste were mainly done on disposable food contact materials (e.g., bowls, paper table wares, beverage cups, paper bags, etc.), several dedicated to substitute single use plastics, and disposable fast food packaging materials (e.g., fast food wraps, pop-corn bags, boxes for French fries, etc.). Textile and leather waste data refers mostly to treated upholstery (both from hous-

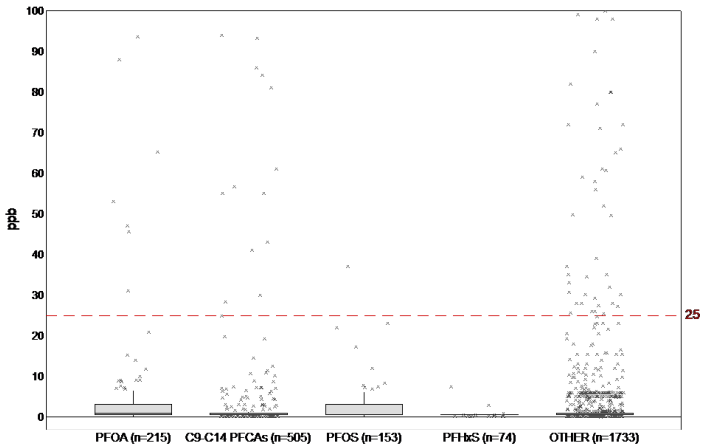

FIGURE 8: Box Plot resuming the concentration data gathered for the category "Paper and Cardboard waste". The line within the boxes shows the median value, the box denotes the range of 50% of data, whiskers range from the lower to the higher value within 1.5 interquartile ranges and asterisks stand for outliers. The y axis was trunked at 100 ppb to ease the comparison between the collected data and the proposed limit value of 25 ppb (red dashed line). The range of values for each investigated parameter is indicated in Supplementary Materials.

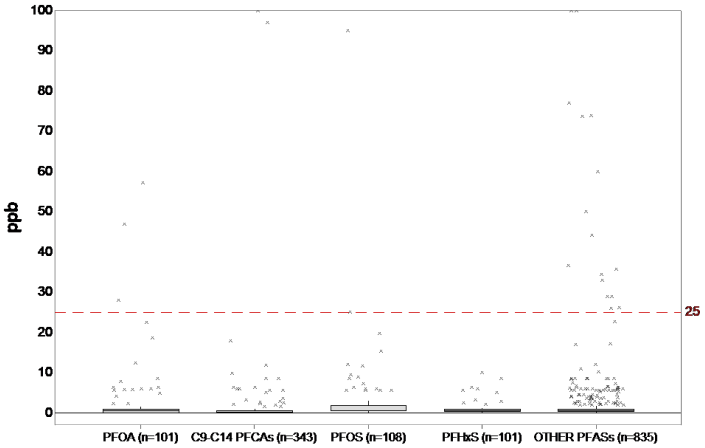

FIGURE 9: Box Plot resuming the concentration data gathered for the category "Textile and leather waste". The line within the boxes shows the median value, the box denotes the range of 50% of data, whiskers range from the lower to the higher value within 1.5 interquartile ranges and asterisks stand for outliers. The y axis was trunked at 100 ppb to ease the comparison between the collected data and the proposed limit value of 25 ppb (red dashed line). The range of values for each investigated parameter is indicated in Supplementary Materials.

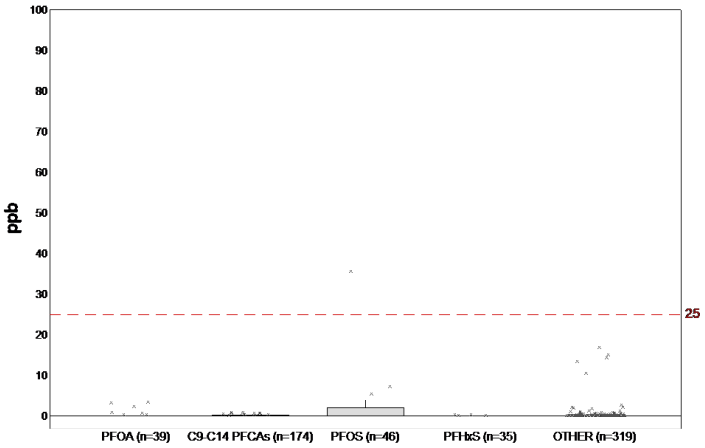

FIGURE 10: Box Plot resuming the concentration data gathered for the category "Plastic waste". The line within the boxes shows the median value, the box denotes the range of 50% of data, whiskers range from the lower to the higher value within 1.5 interquartile ranges and asterisks stand for outliers. The y axis was trunked at 100 ppb to ease the comparison between the collected data and the proposed limit value of 25 ppb (red dashed line). The range of values for each investigated parameter is indicated in Supplementary Materials.

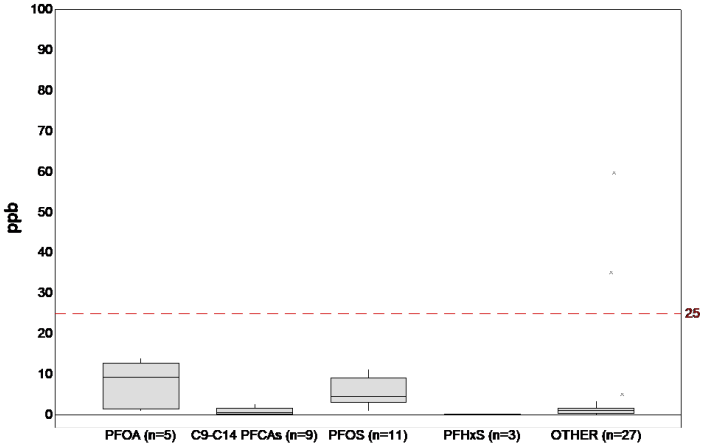

FIGURE 11: Box Plot resuming the concentration data gathered for the category "Metal waste and metal scraps". The line within the boxes shows the median value, the box denotes the range of 50% of data, whiskers range from the lower to the higher value within 1.5 interquartile ranges and asterisks stand for outliers. The y axis was trunked at 100 ppb to ease the comparison between the collected data and the proposed limit value of 25 ppb (red dashed line). The range of values for each investigated parameter is indicated in Supplementary Materials.

es -e.g., carpets- and End-Life-Vehicles (ELV)) and treated apparels. Plastic waste data were mainly collected from measurements done on single-use plastic packaging, ELV plastic components and parts of household appliances. Finally, metal waste dataset was mainly composed of concentration data derived from the analysis of Automotive Shredder Residues (ASR) derived from treatment of ELV. The detailed information about the specific waste types are included, for each entry, in the dataset available in the Supplementary Materials.

4.2 Comparison between PFAS concentrations and proposed limits

Looking at the box-plots, for each material, all the investigated parameters (or groups of them) show similar rightly skewed distributions, with the large majority of data characterized by very small concentration values and only few rare high to very high observations, with median values squeezed near the Q1 and lower whiskers approaching zero ppb, with the only exception of PFOA category for metal waste, which is however characterized by very small sample size.

Except for metal waste for which little data is available, high percentages of data for each group parameter were interestingly recorded below the LOQ or LOD, as quantified in Table 2. Within each material category, the lower fractions of concentration values recorded lower than LOQ or LOD were those related with PFOS and PFOA content, with an average of about 40%, 60% and 50% values < LOQ/LOD for paper waste, textile waste and plastic waste, respectively.Conversely, higher percentages of unquantified concentration values were recorded for the parameter PFHxS, ranging from 68% in textiles and leather waste to more than 90% of data in the paper and cardboard waste. Further, C9-C14 PFAS and the remaining parameters grouped under "Other PFAS", were also characterized by similar trends, showing a peak of 90% of C9-C14 PFAS data under the limit of detection in textile and leather waste and 76% of Other PFAS concentrations not detected for paper and cardboard waste.

As it is visible from the box-plots, just relatively few outliers resulted higher than the considered limit of 25 ppb, for each material category. The percentages of exceedances for each material and groups of parameters are resumed in Table 3. Overall, percentage of exceedances ranged from almost 1% (PFOS measurements in Paper and cardboard waste) to 8% ("Other PFAS" in Textiles and leather waste). With the only exception of plastic waste (characterized by exceedances occurring only for PFOS content), the higher number of exceedances was observed for the other waste categories for the "Other PFAS" groups, with no further clear trends among the other parameters investigated.

Indeed, the higher amount of data per category led to higher number of exceedances. In particular, the material category with the highest average percentage of data above the limit resulted the textile and leather waste, with an average 4,3% of non-compliances, followed paper and cardboard waste with an average total of 3,9% of exceedances. Whether these latter material categories were characterized by at least one exceedance for each parameter or groups of them (with the exception of PFHxS), plastic and metal waste were characterized by values measured above the limits for just one parameters category, namely PFOS for plastic waste (3,23% of exceedances) and "Other PFAS" (11,11%) for metal waste.

4.3 Consideration on the analytical methods

Among the identified papers, a wide range of performed methods could be noted for PFAS analysis in the chosen solid matrixes. Thus, no unique approaches could be observed for the extraction, purification and quantification phases and, unfortunately, not all studies indulged in a thorough description of the performed methods. The role of extraction and purification is crucial in assessing the varying degrees of accuracy of measured PFAS concentrations. Different analytical settings could determine under-estimated concentrations or false positives in the extracts subjected to further quantification step. Also, different waste types could necessitate different approaches for the best performance of PFAS quantification.

TABLE 2: Percentage of data recorded under the LOD/LOQ for each considered material category and investigated parameters.

Parameter	Paper And Cardboards	Textile And Leather	Plastic	Metals
PFOA	40.93%	63.37%	79.49%	0.00%
C9-C14 PFASs	71.68%	90.38%	88.51%	0.00%
PFOS	41.83%	60.19%	41.30%	0.00%
PFHxS	90.54%	68.32%	85.71%	0.00%
Other PFASs	76.28%	62.04%	78.06%	22.22%

TABLE 3: Percentage of data recorded over the concentration limits of 25 ppb.

Parameter	Paper And Cardboards	Textile And Leather	Plastic	Metals
PFOA	6.51%	2.97%	0.00%	0.00%
C9-C14 PFASs	5.35%	0.87%	0.00%	0.00%
PFOS	0.65%	7.41%	2.17%	0.00%
PFHxS	5.35%	0.87%	0.00%	0.00%
Other PFASs	6.98%	8.14%	0.00%	11.11%

Within the manuscripts investigated in this study, diverse methodological approaches concerning extraction media are presented. While methanol stands as the most frequently used, several other media such as ammonium, ethyl acetate, acetonitrile, tetrabutyl ammonium hydrogen sulfate along with methyl tert-butyl ether, acetone, and ethanol, among others, have been utilized. Various extraction times have been employed, ranging from under 60 seconds to overnight periods, achieved through methods like hand or orbital shaking, or vortex mixing. Additionally, a broad spectrum of extraction treatments has been applied, including sonication (focused ultrasound liquid extraction, executed at varying contact times and irradiation power), along with specific pressure (i.e., pressurized liquid extraction) and temperature regimes, ranging from 0°C to 80°C.

Furthermore, the effectiveness of the purification step plays a crucial role, as it eliminates compounds that could potentially interfere with accurate quantification, thereby avoiding false positives. Notably, the inclusion of the solid-phase extraction step is not consistent across all studies. When included, there are disparities in the materials constituting the cartridges, such as weak anion exchange resins, silica, fluorisil, alumina, and variations in the solvent mixtures used for their functioning.

Centrifugation of extracts is a standard practice, with variations in applied rpm values and process durations. Similarly, filtration methods encompass diverse filtering materials (e.g., pp, polyamide, regenerated cellulose) and mesh sizes, spanning from 0.2 µm to 0.45 µm.

While out of the scope of its scopes,this heterogeneity could, indeed, represent a limit of the present study, due to the possible inconsistencies in the comparison and/or underestimation of analytes concentration due to non-efficient extraction methods (e.g., considering quantification on test portions undergone or not to TOP assay in the extraction step). However, the considered papers were assumed as already validated works, where validation is ensured by their peer-reviewed nature and publication in indexed scientific journals. However, an additional comparison was performed to further investigate this issue, between those measurements performed on test portions undergone or not to TOP assays, for those waste categories showing suitable data.

The results of the comparisons between the extracted PFAAs concentrations data quantified on material-specific test samples undergone and not to TOP assay are summarized in Table 4 and depicted in Figure 12, 13 and 14 for textile and leather waste, plastic waste and metal waste, respectively. No data on post TOP targeted concentrations was available for the paper and cardboard waste category.

As the Figures 12,13 and 14 show, there is a significant trend characterizing overall higher concentration of the quantified analytes in samples undergone TOP assay. This pattern implies a notable increase in the quantified concentration values obtained through targeted analysis, as a result of TOP assays. This arises from the fact that targeted analysis can effectively measure the additional quantity of precursors as targeted PFAS, once they undergo oxidation during TOP, resulting in the formation of the analyzed PFAS terminal products. In the case of the metal waste, this could contribute to also a significant increase in the number of exceedances when concentration data are compared with the proposed Regulation limit (Figure 14).

4.4 Proposing a pragmatic solution for PFAS analysis in solid samples for regulatory purposes

Overall, the relevant knowledge on the impact of PFAS in solid waste management is still scarce. According to European regulations, waste is defined as hazardous if it satisfies at least one of the 15 hazard properties (HP) or contains concentrations of certain (POPs) over specific legal thresholds (European Commission, 2014; European Parliament and European Council, 2019). In this list of chemicals, PFAS are not included therefore the hazardous classification of waste is currently not directly impacted by the concentration of PFAS in the waste. However, the presence of PFASs could influence other hazardous properties, such as ecotoxicity (HP 14), when assessed by direct testing.

The current regulations governing the disposal and recovery of waste containing PFAS are limited to a few groups (less than 5% of the total of PFAS) and their implementation and impact in the waste sector are known, with one relevant exception. This is due to the “ambiguous” definitions of the correlated chemicals to the perfluoroalkyl acids (PFAAs) that should be covered by the regulations. In fact, the following general terms are reported: derivates of perfluorooctane sulfonic acid (PFOS); salts and related compounds of perfluorooctanoic acid (PFOA); salts and related compounds of perfluohexane sulfonic acids (PFHxS). These definitions involve hundreds of compounds for which no regulatory lists exist. Furthermore, only a few substances can currently be detected analytically inducing – de facto – a regulatory void.

TABLE 4: Descriptive statistics of PFAAs concentration data measured on test samples undergone TOP Assay (AFTER TOP ASSAY) and not (BEFORE TOP ASSAY) of textile and leather waste, plastic waste and metal waste.

Waste Type	Type Of Analysis	n	Median	75%	90%	95%
		-	ppb			
Textile	PRE TOP	1418	0.61	0.87	4.29	8.65
	POST TOP	424	1.95	8.25	10.78	37.91
Plastic	PRE TOP	591	0.08	0.11	0.34	0.83
	POST TOP	112	1.52	5.47	11.78	23.09
Metal	PRE TOP	55	1.21	4.23	10.53	20.34
	POST TOP	51	7.71	28.76	59.18	107.66

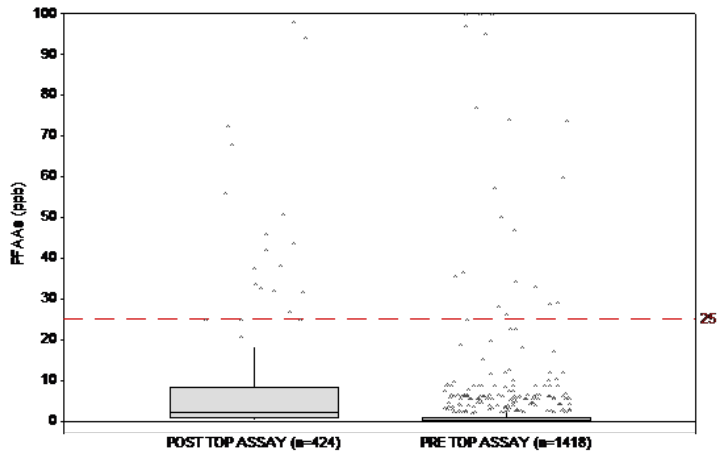

FIGURE 12: Box Plot resuming the differences between the PFAAs concentration data before and after TOP assay performance on textile and leather waste test samples. The y axis was trunked at 100 ppb to ease the comparison between the collected data and the proposed limit value of 25 ppb (red dashed line). The range of values for each investigated parameter is indicated in Supplementary Materials.

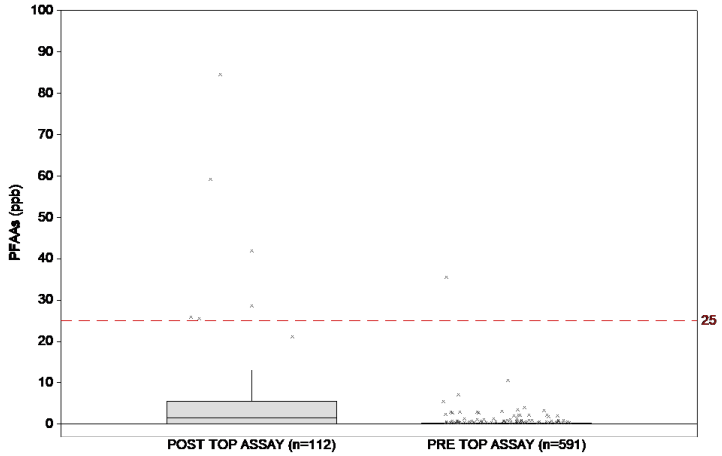

FIGURE 13: Box Plot resuming the concentration data gathered for the category "Plastic waste". The line within the boxes shows the median value, the box denotes the range of 50% of data, whiskers range from the lower to the higher value within 1.5 interquartile ranges and asterisks stand for outliers. The y axisBox Plot resuming the differences between the PFAAs concentration data before and after TOP assay performance on plastic waste test samples. The y axis was trunked at 100 ppb to ease the comparison between the collected data and the proposed limit value of 25 ppb (red dashed line). The range of values for each investigated parameter is indicated in Supplementary Materials.

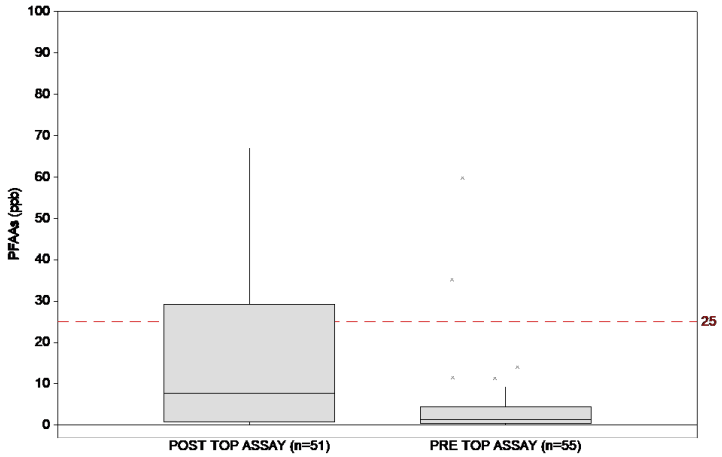

FIGURE 14: Box Plot resuming the differences between the PFAAs concentration data before and after TOP assay performance on metal waste test samples. The y axis was trunked at 100 ppb to ease the comparison between the collected data and the proposed limit value of 25 ppb (red dashed line). The range of values for each investigated parameter is indicated in Supplementary Materials.

Much more complex is the implication of the current, and more importantly, ongoing proposed regulations (see BAuA et al, 2023a) concerning limits specifically related to substances, matrixes, and article. The complexity is due to the connection between the characteristics of the new materials which should be guaranteed before being placed on the market and the criteria upon which the waste should respect to be gain the status of EoW. This issue can be analysed by considering three main points: (I) the time interval between the moment of implementation of new provisions on substances and the moment in which these limits are also required for the EoW procedure; (II) the analytical method to measure the PFAS; (III) the specific values of new regulatory limits.

The limits on the use of PFAS will determine a reduction of PFAS concentration in new articles and products only from the moment new regulations will enter into force. However, in the waste cycle older products containing higher concentrations of PFAS could end their life cycle and become waste. Besides, the use of PFAS in some products is still considered "essential" and if there isn't a good pre-selection before any recovery process, the presence of PFAS in waste cycles could endure for a long time. In this context, "tagging" products (chemically or optically through a label or code) to identify those with PFAS concentration higher than a limit (e.g., 25 mg kg^{-1}) could improve sorting before recovery.

These aspects must be carefully evaluated by eventually introducing express derogations for the EoW or at least defer the date for when these limits come into force. This would continue to allow the recovery of the waste already generated before the new limits become mandatory. Another solution to guarantee human safety could be the introduction of limits without any exemption for the waste sector (derogations, deferred time, etc.). This could have an impact on the recycling market.

Concerning the methodology to measure PFAS in solid samples, it should be considered that "PFAS" is a broad, general, non-specific term, which does not inform whether a compound is harmful or not, but only communicates that the compounds under this term share the same chemical structure. To this regard, whereas PFOA and PFOS have been well characterized in terms of their hazard, little to no toxicity information exists for the vast majority of PFAS. Evaluating thousands of PFAS using traditional toxicity approaches, in turn, would be impractical, costly and time-prohibitive, as well as requiring extensive use of animal testing. Therefore political decision makers must assess whether a transitional period is necessary to establish a comprehensive set of technical guidelines regarding the appropriate methodologies to analyse the parameters for the 'PFAS Total' and the 'Sum of PFAS'. Different aspects should be part of the analysis: such as human protection, the necessity of avoiding environmental disputes, and the aims of the circular economy.

A pragmatic solution could start from the proposal of the national authorities of Denmark, Germany, the Netherlands, Norway, and Sweden (BAuA et al, 2023), but considering the acknowledged limitations of the analytical procedures, the solution could include the following steps:

- since the definitions of PFAS will likely continue to be modified by developing regulations and enhanced research, it is necessary/important to identify an updated, and operative definition for PFAS that is accepted by the scientific community, as the proposal put forward by the OECD in 2021;
- define a first limit of concentration (screening level) for all the PFAS as a definite total to overcome the approach followed up until now in REACH, where the provisions of restriction and eliminations were limited to individual substances or groups of closely related substances. Unlike the mentioned proposal (BAuA et al, 2023), the analytical technique should be "non-targeted" as the total organic fluorine (TOF) analysis, since it represents a more accurate method compared to the total fluorine analysis (TF) suggested in BAuA et al. (2023a);
- if the screening level is not respected, a further analysis should be implemented considering "targeted techniques" on the base of a "positive" official list of specific chemicals for which the toxicity of the compounds is known or assessed by chemical similarity and/or toxicity modelling with state-of-the art tools. This list should name harmful substances and could be continually updated alongside the progress made from new scientific analysis. Furthermore, this list could represent a solution for the interpretation of general terms such as "salts", "related compounds" and "derivates" of perfluoroalkyl acids. It is worth mentioning that when a TOP assay is applied, a higher concentration of targeted PFAS should be expected as suggested by the systematic critical review.

5. CONCLUSIONS

Currently, the relevant knowledge on the impact of PFAS in solid waste management, from disposal to recycling, is still scarce. In this context, regulation updates on PFAS contents in substances, matrixes and articles could influence these latter, e.g. by limiting the circularity of recycled materials and affecting EoW procedures. In this context, this review tries to answer to the stringent need to discuss the possible consequences of proposed new limits for PFAS in products on relevant waste recycling sectors.

In order to achieve this, a systematic analysis of scientific literature was conducted to gather concentration ranges of PFAS in waste items across four key material categories (plastic, paper and cardboard, textile and leather, metals). This review encompassed peer-reviewed articles and other pertinent documents, aiming to compare the obtained results against the lowest proposed regulatory limit for any PFAS in products (i.e., 25 ppb). For each material category, very few outliers resulted higher than the considered limit of 25 ppb. The percentage of exceedances ranged from almost 1% (PFOS measurements in paper and cardboard waste) to 8% ("Other PFAS" in Textiles and leather waste). The analysis of various papers revealed diverse methods for PFAS analysis in solid matrices. Discrepancies in extraction, purification, and quantification methods could have impacted accuracy and hindered comparison.

Notably, results showed higher analyte concentrations in samples where TOP assay was performed prior to the quantification step.

Based on a thorough discussion on the specific nature of the term "PFAS" as defined in the current regulation and the current need for an harmonized analytical strategy to quantify the parameter 'PFAS Total' and the 'Sum of PFAS', a pragmatic solution for the waste sector was finally proposed. A stepwise procedure is suggested. First, there is the necessity to establish an updated and universally accepted definition for PFAS, reflecting changes in regulations and scientific advancements. Secondly, it supports setting an initial concentration limit (as a screening level) for "Total PFAS". This can involve a more accurate non-targeted analytical technique, such as total organic fluorine (TOF) analysis, rather than total fluorine analysis (TF). Finally, if the screening level isn't met, the proposal advocates employing "targeted techniques" based on a verified list of known harmful chemicals, derived from toxicity assessments.

6. FURTHER DEVELOPMENT

To better foresee the implication on management practices of PFAS containing waste, it is suggested to design a systematic experimental activity in which representative samples of wastes are collected and analysed by means of the same standardized methodologies under a uniquely defined scope. This could overcome the limits of the data made available by the systematic critical review which were obtained from the application of a set of different methodologies and often with different aims.

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