

Review

## **An Overview of Treatments for Ultraviolet Quenching Substances (UVQS) and Per- and Polyfluoroalkyl Substances (PFAS) Removal from Landfill Leachate**

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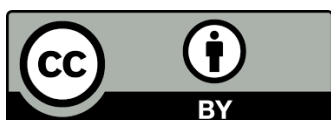
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### **Abstract**

Organics found in landfill leachate (humic acids, fulvic acids, and hydrophilic fraction) have a solid ability to absorb ultraviolet (UV) radiation, which negatively affects UV disinfection; leachate ultraviolet quenching substances (UVQS) can, therefore, have a significant impact on the cotreatment of landfill leachate and sewage in wastewater treatment plants. On the other hand, per- and polyfluoroalkyl substances (PFAS), which pose potential risks to the environment and human health, commonly exist in landfill leachate due to their wide application in various industrial and consumer products. Therefore, removing UVQS and PFAS from landfill leachate is crucial. In this work, the advances in removing UVQS and PFAS from landfill leachate in the last decade are reviewed to find a standard treatment for both contaminants to lower the costs and space required for the leachate treatment process. The benefits and drawbacks of biological, physical, chemical, and electrochemical treatments were examined. Physical, chemical, and electrochemical treatments showed advantages over biological treatments but higher energy and/or material costs. The global analysis indicated that similar technologies, such as adsorption or osmosis, can be used as effective methods to remove UVQS and PFAS from landfill leachate and suggested that both types of pollutants



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could be eliminated simultaneously with a single treatment based on one of these two technologies.

### **Keywords**

Ultraviolet quenching substances; per- and poly-fluoroalkyl substances; landfill leachate; removal technologies

## **1. Introduction**

The evolution of society has currently reached a life model based on a constant increase in consumption whose immediate consequence is the generation of a series of solid wastes that can harm the environment [1]. Although the generation of waste by man has always existed, for a long time, the waste of animals and plants contributed to the support of the life of the ecosystems. However, the constant increase in their generation rates has caused, in many cases, a breakdown of the balance between the biosphere and human activities [2]. Waste generation rates are increasing drastically. Factors such as population growth, urbanization, economic growth, and consumer shopping habits determine this increase [3]. In 2020, the globe produced 2.24 billion tonnes of municipal solid waste (MSW); in 2050, global waste generation is expected to grow to 3.88 billion tonnes per year, a 73% increase compared to 2020 levels [3].

The increasing generation of MSW has become a significant burden to society because of the serious environmental and economic problems during waste disposal. Compared with other technologies, such as composting or incineration, landfilling is a relatively convenient, inexpensive, and widely employed method for MSW management [4].

MSW disposal in landfills involves some risk since solid waste will be exposed to degradation, further releasing hazardous constituents to the environment. Although modern landfills are highly engineered facilities designed to suppress or diminish the adverse impact of waste, producing leachates remains a severe problem for MSW landfills because these leachates constitute a significant danger to soil, surface water, and groundwater [5].

Leachate is produced from waste degradation and rain penetration through the landfill [6]. It results from two main processes occurring in a landfill: 1) water percolation in the dumped waste mass and 2) mass transfer of chemicals from waste to percolating water [7].

Pollutants in municipal landfill leachate are toxic organic and inorganic compounds, including microorganisms [8, 9]. The most common specific categories of pollutants found in leachates include dissolved organic matter, trace ions, xenobiotic organics such as polychlorinated biphenyls or pesticides, and ammonia [10, 11]. However, leachates can also release heavy metals, polychlorinated organic compounds, and emerging contaminants into the ecosystem [12-14]. Therefore, leachate collection and treatment are essential to avoid the environmental pollution from landfills [15, 16].

Although some landfills use leachate pre-treatment before discharge, and others provide leachate treatment prior to permitted discharge to the environment, most leachate is sent directly to wastewater treatment plants for treatment. Although leachate cotreatment with municipal wastewater can remove significant amounts of pollutants (e.g., organic matter, nitrogen,

phosphorus) [17], this treatment is being hampered by emerging contaminants and strict regulations [18].

Emerging contaminants such as ultraviolet quenching substances (UVQS), per- and poly-fluoroalkyl substances (PFAS), microplastics (MP), antibiotics (A), endocrine-disrupting chemicals (EDC), or antibiotic resistance genes (ARG) [13, 19-22], have made it challenging to use traditional wastewater treatment methods (primary and secondary wastewater treatments). These pollutants are becoming more concerning due to their co-occurrence and impact on overall treatment effectiveness [14]. This problem implies the need for action by wastewater treatment plants and landfills for enhanced emerging contaminant removal or treatment [23].

Among emerging contaminants, at present, UVQS and PFAS are the most concerning leachate pollutants because the presence of UVQS in the final effluent after biological treatment that hampers ultraviolet (UV) removal of pathogens in subsequent disinfection processes [24] and the dominance of PFAS (dangerous for human health and the environment) in leachate compared to wastewater and freshwater [14].

Although there are many studies on eliminating UVQS or PFAS from landfill leachate, none have evaluated the possible existence of a standard treatment for both contaminants. Consequently, it is crucial to explore the UVQS and PFASs removal from landfill leachate through advanced remedial measures to find a standard therapy for both pollutants, reducing leachate treatment costs and space. Therefore, this research aims to document the existence or absence of common treatments for UVQS and PFAS removal from landfill leachate. For this purpose, this paper reviews the latest potential technologies for UVQS and PFAS elimination from landfill leachate. It discusses the advantages and disadvantages of these methods for treating landfill leachate. In this context, the study has four specific objectives: to characterize the landfill leachate in terms of UVQS and PFAS, to review the recent technologies of UVQS removal from landfill leachate, to review the new technologies PFAS removal from landfill leachate, and to use the information obtained in previous objectives to determine a possible treatment for simultaneous removal of UVQS and PFAS from landfill leachate.

## **2. Leachate Characteristics**

Landfill leachate is a complex wastewater with considerable variations in composition and volumetric flow [25].

The physical-chemical characteristics of leachate depend on a series of factors such as [4]:

- Nature, composition, and total volume of stored waste
- Landfill age, design, and operational practice
- Hydrogeology, climatic conditions, and seasonal weather changes.

Among these factors, the landfill age becomes the critical factor in determining the leachate composition once the leachate is collected in the leachate collection pond [26, 27]. Based on the landfill age, leachates are classified as young, intermediate, and old [28]. Young leachates are those that come from landfills less than 5 years old, where the aerobic and acidic stages prevail, and biodegradability is essential; the pH is ~6.5; carboxylic acids constitute more than 80% of the organic compounds; the biodegradable (as biochemical oxygen demand, BOD) and total organic matter (as chemical oxygen demand, COD) are above 2000 and 10000 mg/L, respectively. The BOD/COD ratio is above 0.3 [28]. Old leachates are those from facilities that are more than 10 years old, generated

during the maturation phase; they are mainly composed of a refractory mixture of UVQS (mainly humic acids and fulvic acids) with pH higher than 7.5, COD below 4000 mg/L, BOD below 150 mg/L and BOD/COD ratio below 0.1 [28]. Intermediate leachates are generated in landfills of 5 to 10 years old, and so in the methanogenic phase, and show intermediate characteristics between those of the young and old leachates [28].

Age-related progressive alteration of humic compounds in leachates is mainly responsible for their heterogeneity and difficulty in characterization. In young leachate, fulvic acids are produced first, their concentration higher than that of humic acids. Still, with humification, humic acid concentration rises over time and finally falls as landfills stabilize and natural dilution or degradation occurs [19]. Humic compounds are frequently viewed as the byproducts of the oxidative and biodegradative pathways due to the prolonged exposure of the precursors to water, oxygen, and sunshine; because of this, humic compounds are regarded as "old molecules" resistant to further biological processing but vulnerable to other specific treatments such as advanced oxidation [19]. The concentration of UVQS (consisting of humic acids, fulvic acids, and hydrophilic fraction) in landfill leachate can range from tens to tens of thousands of mg/L, and it is significantly higher than UVQS concentration in freshwater or municipal wastewater (Table 1). PFAS in leachate derived from environmental and consumer products deposited in landfills (such as electronics, cosmetics, cleaning products, dental floss, water-repellent fabrics, carpets, food packaging materials, non-stick cookware, upholstery...) [29-31] can cause adverse biological effects in animals and humans [32], and are reportedly extremely difficult to degrade [13]. The amounts of PFAS in landfill leachates are exceptionally high when compared to the concentration of PFAS found in other liquid matrices such as freshwater or municipal wastewater (Table 1). The reported total PFAS contents in raw leachates ranged from a few to tens of thousands of ng/L (Table 1). The wide range of concentrations of PFAS in leachate is linked to factors such as the kind of waste, landfill management, and treatment practices [29, 33].

**Table 1** Concentration of the different categories of UVQS and PFAS in landfill leachate, municipal wastewater, and freshwater.

Compound	Landfill leachate	Municipal wastewater	Fresh water	References
UVQS (mg/L)				
Humic Acids (HA)	16-8800	11-96	0-20	[19, 34-38]
Fulvic Acids (FA)	56-14400	14-133	0-34	[19, 34, 35, 37, 38]
Hydrophilic fraction (HPI)	199-14020		0-14.5	[19, 35, 39]
PFAS (ng/L)				
Perfluoroalkyl carboxylates (PFCA)	35-24,413	10-980	5-970	[14, 40-46]
Perfluoroalkane sulfonates (PFSA)	30-13,269	5-965	4-960	[14, 40-46]
Perfluorooctane sulfonamide (PFOSA)	1-46	2-25	0-44	[14, 40-46]

### 3. Ultraviolet Quenching Substances (UVQS) Removal

Some refractory organic compounds in landfill leachate with UV quenching characteristics (UVQS) decrease the UV transmittance of wastewater and, hence, can interfere with ultraviolet disinfection when landfill leachate is co-treated with municipal sewage [47]. Ultraviolet quenching

substances (UVQS) are divided into three categories: humic acids (HA), fulvic acids (FA), and hydrophilic fraction (HPI), each of which has specific characteristics and behaviors during the treatments [47]. HA, FA, and HPI concentrations in landfill leachate appear in Table 1.

In the last decade, different technologies have been used to deal with UVQS: biological, physical, chemical, and electrochemical treatments. However, biological treatment is less effective than the other alternatives [48].

### **3.1 Biological Treatments**

The efficiency of biological treatments for landfill leachate depends mainly on the nature and composition of the leachate [49] and can be carried out in either aerobic or anaerobic reactors.

Aerobic biological treatment mainly comprises fixed film and activated sludge bioreactors [19]. After aerobic biological treatment, the decline in UV absorbance and UVQS can range from 2.5 to 55% and 26-60%, respectively [19]. Following biological therapies, the average reduction in HA, FA, and HPI concentrations was 50.2, 53.9%, and 50.1%, respectively [19]; the elimination of UV absorbance for the corresponding fractions was 43.5, 23.5%, and 19.5% [19]. Biological treatment is probably less effective at lowering UV absorbance than total organic carbon due to its ineffectiveness for recalcitrant organic compounds (mainly humic substances) [19].

Organics and UV absorbance can only be removed to a limited extent by anaerobic biological treatment of leachate. For instance, a two-stage anaerobic membrane bioreactor removed 39% of the HA, 37% of the FA, and 55% of the HPI fraction from a landfill leachate, while the reduction of UV absorbance was 46% [50].

Because independent biological treatments are ineffective for removing UVQS, combinations of biological and physicochemical therapies have been researched. For example, leachate blended with sewage and treated by nanofiltration after biological treatment increased the UV transmittance from 35% to 62% when leachate represents 5% of the overall volumetric flow [51]. When powdered activated carbon was added to an activated sludge process, HA, FA, and HPI removal increased from 27% to 53.1%, 40% to 44.2%, and 59.8% to 59.8%, respectively [51].

### **3.2 Physical Treatments**

Most physical treatments use sorption and membrane separation-based methods. Physical treatments typically remove between 23 and 97% of the UV absorbance and between 34 and 98% of the UVQS [19]. These treatments have the advantage that the removed UVQS are not destroyed but separated and can be recovered.

Among the sorption-based methods, HA with a molecular weight range of 0.5-2 kDa and FA with a molecular weight range of 1-3 kDa were preferentially absorbed by carbon nanotubes [52]. Iron oxide coated may also effectively remove UVQS; for instance, using an amount of 40 g/L for 6 hours, it was possible to remove 89.1% UV absorbance and 65.5% UVQS from leachate that had been treated by a microbial fuel cell [53]. Powdered activated carbon reduced UV absorbance by 92%, showing preferential adsorption by the hydrophobic fraction (HA and FA) [54]. Under optimal conditions (90 min of stirring time, 80 g/L laterite, and pH 3.5), laterite showed the higher removal of humic acids: 97% [55]. The regeneration of the sorbents limits the application of sorption-based therapy once they are spent [19].

Among the membrane separation-based methods, nanofiltration and forward osmosis processes removed around 90% [56] and 98% UVQS [57], respectively, from landfill leachate during water recovery. It was possible to remove 60% HA, 11% FA, and 40% HPI in an ultrafiltration-based membrane bioreactor (MBR) [58]. Humic compounds from landfill leachate had reported removal effectiveness of 43% by coupled lime precipitation (8 g/L lime) and microfiltration, while nanofiltration improved the removal to 86% [59]. However, the pore size of membranes has a significant impact on performance. For example, a study on ultrafiltration of biologically treated leachates showed an increase of UV absorbance removal close to 43% when pore size decreased from 700 nm to 7 nm; in this pore size range, HA and FA were mainly retained; for pore size of 4 nm, additional UV absorbance removal was obtained due to retention of HPI [60, 61]; in consequence, the smaller the pore size, the greater the removal of UVQS. The main handicap of using membranes to remove UVQS is the increase of membrane fouling in the presence of these compounds [62, 63].

### **3.3 Chemical Treatments**

Leachate UVQS can be treated chemically using coagulation-flocculation or advanced chemical oxidation (Fenton, ozone, percarbonate). Chemical treatments have been shown to reduce 7-92% UV absorbance and remove 5-85% UVQS [19].

Coagulation-flocculation with ferric chloride and aluminum sulfate, the two coagulants most typically employed, removed 54%-74% organic matter from landfill leachate; notably, aluminum sulphate only marginally reduced the UV absorbance of treated leachate, but ferric chloride coagulation increased it dramatically by up to 10 times; the UV absorbance increase is probably caused by the complexation of soluble ferric and leachate organic matter [64].

Fenton's oxidation is a very efficient chemical treatment that can remove an average of 84.5% UV absorbance and 67.4% UVQS from leachate [65-67]. However, other chemical treatments, such as ozone treatments, can only remove 5.6-30% of UVQS, although these treatments can reduce UV absorbance by 65% [19, 66, 68]. This is due mainly to O<sub>3</sub> preferential interaction with hydrophobic chemicals, which change to hydrophilic smaller molecular weight fraction after ozone treatment [66]. For instance, following ozone treatment, a 100% decrease in HA, a 16.2% decrease in FA, and a 78% increase in the HPI fraction were obtained [68]. Leachate had also been subjected to sodium percarbonate oxidation, but this treatment is less effective in eliminating UV absorbance (8-43.4%) and UVQS (15.1-15.6%) [69].

Fenton and ozone treatments can be improved by combination with other therapies. Thus, solar photo-Fenton successfully removed 80-85% leachate UVQS [70]. Compared with the O<sub>3</sub>-alone process, the removal efficiencies of UVQS was improved in O<sub>3</sub>-combined processes (i.e., O<sub>3</sub>/H<sub>2</sub>O<sub>2</sub> and O<sub>3</sub>/UV) approximately by 7-15% [71].

Despite being a powerful and efficient chemical oxidation technique for removing UVQS, the Fenton treatment has disadvantages, such as chemical demand, sludge generation, and foaming during the first pH adjustment [72]. The significant amount of electricity used to generate ozone is the disadvantage of ozone-based treatment [73].

Subcritical water catalytic oxidation [74], supercritical water oxidation [75], and catalyzed wet oxidation [76] also demonstrated efficiency in the treatment of leachate UVQS, showing 66, 61, and 93% of HA removal, respectively. The elevated chemical cost generally restricts the possible use of these treatments [19].

### 3.4 Electrochemical Treatments

Electrochemical oxidation and electrochemical coagulation are the techniques primarily used in the electrochemical treatments of leachate UVQS. They can remove 57-80% of UVQS and 33.5-89.5% of UV absorbance [19].

Both direct and indirect oxidation can occur during electrochemical oxidation of leachate UVQS; in direct oxidation, a hydroxyl radical ( $\cdot\text{OH}$ ) is produced, which causes oxidation of UVQS on the anode surface; UVQS undergoes indirect oxidation when anodically created oxidants (e.g.,  $\cdot\text{OH}$ ,  $\text{O}_3$ ,  $\text{HClO}\dots$ ) are produced [77].

Electrochemical coagulation involves the production of ions by electricity-mediated electrolysis [78]; ions are then hydrolyzed and polymerized and absorb or aggregate UVQS to be removed as a floated or settled sludge [79]. Due to their better characteristics (accessibility, superior ability to remove contaminants, and continuous production of active ions), aluminum and iron are most frequently used as anodes in the electrochemical coagulation process [80].

Under optimal chemical oxygen demand removal conditions, the electrochemical coagulation process removed fractions of humic substances from the highest to the lowest: 69% HA, 63% FA, and 61% HPI [81]. These variations in removal seem to be due to the different molecular weights and surface loads of the three organic fractions [82]. In the electrochemical oxidation process, the removal of different fractions of humic substances occurred inversely to that of the electrochemical coagulation process, from the lowest to the highest: 40% removed HA, 55% removed FA, and 68% removed HPI [81]. This difference could be explained because in the electrochemical oxidation process, the elimination of UVQS was produced by the action of  $\cdot\text{OH}$  radicals, which more easily broke down the less complex molecules from less to more intricate structural complexity: HPI, FA, and HA [83].

Combined electrochemical processes have also been studied for treating landfill leachate UVQS with varied performances. For example, electrochemical methods combined with UV irradiation allowed the removal of 65% of humic compounds from biologically treated landfill leachate after 30 min of treatment [84]. Bioelectrochemical systems (a combination of anaerobic degradation and electrochemical process) achieved a 71-83% UVQS decrease and a 25.5-49.5% UV absorbance decrease after treatment [85, 86]. A membrane electrochemical reactor was developed explicitly for treating UVQS contained in landfill leachate; this process obtained significantly better reductions of both dissolved organic carbon (61.5%) and UV absorbance (63.4%) when compared to a control reactor without a membrane separator; this improved performance was most likely caused by the interactions between humic substances deposition and increased organic oxidation [87]. Electrochemical treatments are associated with important energy consumption [87].

## 4. Per- and Polyfluoroalkyl Substances (PFAS) Removal

Per- and poly-fluoroalkyl substances (PFAS), according to the Organisation for Economic Co-operation and Development (OECD), 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 ( $-\text{CF}_3$ ) or a perfluorinated methylene group ( $-\text{CF}_2-$ ) is a PFAS” [88].

PFAS can be divided into two broad categories: perfluoroalkyl acids (PFAA or terminal PFAS) and PFAA-precursors or PFAS precursors; compared with PFAA, PFAA-precursors are usually less stable

and degrade into related PFAA depending upon environmental conditions [89, 90]. PFAS can also be separated into two groups depending on their chain length: long- and short-chained; long-chain PFAS are perfluoroalkyl carboxylic acids (PFCA) with eight or more carbon atoms or perfluoroalkyl sulfonic acids (PFSA) with six or more carbon atoms [91]. Based on the tail ending of the compounds, terminal PFAS include but are not limited to perfluoroalkyl carboxylates (PFCA), perfluoroalkyl sulfonates (PFSA), and perfluorooctane sulfonamide (PFOSA). At the same time, PFCA and PFSA belong to perfluoroalkyl acids (PFAA) [31]. Concentrations of different PFAS groups in landfill leachate are included in Table 2.

**Table 2** Concentration range of the most frequently detected species of PFAS in landfill leachate in ng/L [14].

Specie	Concentration range	Specie	Concentration range
PFPeA	0-6500	PFBA	0-3050
PFHPA	0-5900	PFDA	0-1200
PFOS	0-5700	PFHxS	0-1150
PFOA	0-5000	PFNA	0-750

Since PFAS are discharged from wastes containing PFAS, landfills represent the last stage in the life cycle of PFAS [89]. Consequently, PFAS were detected in the landfill leachate, being the most frequently seen species of perfluorbutanoic acid (PFBA), perfluoropentanoic acid (PFPeA), perfluoroheptanoic acid (PFHPA), perfluorooctanoic acid (PFOA), perfluorononanoic acid (PFNA) perfluorodecanoic acid (PFDA), perfluorohexane sulfonate (PFHxS), perfluorooctane sulfonate (PFOS) [14, 92, 93].

#### 4.1 Biological Treatments

Leachate landfills containing PFAS can be treated using biological treatments [33, 94, 95]. When the conventional activated sludge process in wastewater treatment plants was used for landfill leachate [31], it failed in PFAS degradation [96]. Some bio-leachate treatment matrices, such as aerobic landfill reactors and membrane bioreactors, have even been found to have an overall increasing tendency in PFAS chemicals [33, 94, 95]. This increase could be explained by the persistence of perfluoroalkyl acids against biodegradation [97] and the possible biodegradation of fluorinated precursor compounds to perfluoroalkyl acids during biological processes, which was reported for biological treatment in wastewater treatment plants [98, 99].

However, a 61% decrease in the concentrations of PFAS after membrane bioreactor treatment has also been reported [94]; it was attributed to the loss of PFAS due to water-to-air transfer by bursting bubbles in the aeration tanks and the sorption of PFAS on the activated sludge as the concentrations of these PFAS in the sludge phase increased.

These different results could be due to the main factors that influence the biological degradation of PFAS:

- Microbial species and quantity: Different microorganisms have different degradation efficiencies and adaptabilities to PFAS. For instance, the PFAS contaminants can be degraded by ligninolytic fungi [100] or via biodegradation by aerobic bacteria [97] into non-toxic compounds such as carbon dioxide, water, and ammonia.



- Temperature and pH: Microbial activity and PFAS breakdown efficiency can be increased at the appropriate temperature and pH [94]. According to this, it is vital to choose the best environmental conditions based on the circumstances because different microorganisms have varied requirements.
- PFAS structure and concentration: The capacity of microorganisms to degrade PFAS can be influenced by its structure and concentration. Thus, long-chain PFAS are more challenging to break down than short-chain PFAS [101], and microorganisms may experience physiological changes and/or inhibitory effects because of PFAS exposure [102-104].
- Other environmental factors: Degradation of PFAS can be impacted by nutrients, organic matter, and oxygen. For example, PFOS and related PFAS mineralization should involve sequential aerobic and anaerobic degradation [105].

According to these factors, the biological treatment of PFAS has some disadvantages: it is a relatively slow process that needs time and conditions, and it can produce intermediates that require additional treatment to avoid secondary pollution [33, 90, 94, 95].

A promising approach to achieving appropriate leachate effluent values is the combination of physical, chemical, and biological treatments, for example, a biological treatment combined with reverse osmosis, activated carbon, or nanofiltration [31].

#### **4.2 Physical Treatments**

Most physical treatments rely on sorption and membrane separation-based techniques and remove between 61 and 100% of PFAS present in leachate [102, 106].

The sorption-based techniques include preferentially carbon as an adsorbent. Activated carbon can efficiently remove PFAA from leachate (70%-99% removal) [107]. Granular activated carbon effectively removed PFAS from leachate [108] but powdered activated carbon and activated carbon fiber showed better adsorption performance than granular activated carbon due to their highest surface area [109]. Biochar produced from biomass pyrolysis at 500-600°C can adsorb 80% of long-chain PFAS and is highly stable [110]; if biochar is created using a pyrolytic combustion integrated process, it may adsorb 90%, while short-chain PFAS performs poorly [111]. The adsorption of short-chain PFAS can be improved by using a modified coal fly ash as an adsorbent [112]. Coal-based magnetic activated carbon removed 72.8-89.6% PFCA with 8 g/150 mL adsorption dosage and 120 min contact time [113]. However, carbon sorption-based techniques have disadvantages, such as the tendency to become saturated and require regular replacement or regeneration [107].

Wetlands have also been used to remove PFAS from landfill leachate. Studies have shown that several plants can adsorb PFAS, and selecting suitable plant species can improve the adsorption effect [114]. Overall, the constructed wetland treatment system removed 61% of total PFAS and 50-96% of individual PFAS [106]. The removal of PFAS from landfill leachates in constructed wetlands can be enhanced using young plants or species with a high affinity for PFAS and substrates with high PFAS sorption capacity [115]. Despite being a green and sustainable method of PFAS remediation, phytoremediation has many drawbacks and restrictions, including a high time consumption, harmful effects on plants, and risk of entering PFAS into food chains, that restrict the use of wetlands in PFAS removal [116].

Studies carried out have proven that membrane-based treatment technology is successful in removing PFAS from leachate. Ultrafiltration and nanofiltration membranes have been utilized to

remove PFAS from leachate, exhibiting a success of more than 90% [95]. Systems fitted with reverse osmosis showed significant reductions (98-99%) of PFAS in the permeate [117]. An explanation for the different efficiency of these treatment systems in removing PFCA from the leachate is possibly in the characteristics of the membranes. Ultrafiltration, nanofiltration, and reverse osmosis are treatments working with semipermeable membranes, which separate the leachate into a clean permeate and a contaminated residue. The membrane pores of reverse osmosis are smaller than in ultrafiltration and nanofiltration and are, therefore, more effective for PFC removal than ultrafiltration and nanofiltration [118]. Although membrane filtration systems present significant advantages in PFAS removal (including low energy consumption, continuous separation, and enhanced treatment effectiveness), some issues can complicate the use of membranes, such as high levels of organic compounds, heavy metals, suspended solids, and salinity in leachate which can cause fouling of the membrane [119, 120], and concentrate management [121].

As opposed to membrane technology, membrane distillation uses hydrophobic membranes to keep non-volatile solutes out of the membrane pores [122, 123]. Because the regulated PFAS in the leachate are not volatile, theoretically, membrane distillation could eliminate them from the leachate with a rejection close to 100% [102]. Effectively, hybrid microporous materials with incorporated metal-organic framework showed a 98.4% rejection rate for total PFAS. However, it should be noted that the maximum rejection to PFOA was 65.6%, much lower than its rejection to total PFAS [100]. Despite its high PFAS rejection rate, membrane distillation has disadvantages such as energy-intensive water vaporization, low permeate flux, and membrane fouling issues [63, 107].

Foam fractionation is another technology that has been tested for leachate treatment. The efficiency of the foaming separation for concentrating PFAS changed by a compound with an average removal percentage (the percent difference between PFAS in leachate before and after foam removal) of 69% [124]; it was demonstrated that foam fractionation was successful in removing  $\geq 98.7\%$  PFOS,  $\geq 99.7\%$  PFOA, and  $\geq 98.8\%$  PFHxS from the feed stream without using absorbent media or chemical amendment consumables [125]. It was verified that long-chain PFAS were more effectively removed through foaming ( $>90\%$  on average) than short-chain PFAS, which had relatively low percent removal values ( $<30\%$  on average) [126]. Foaming can be utilized most efficiently in systems with high concentrations of long chains of PFAS [127]. Treatment efficiency decreased with decreasing contact time, air flow rate, and collected foam fraction [126]. The most significant advantage of the foam fractionation technology is its simplicity [126] and easy application at a relatively low cost at most landfills, where leachate aeration is now customary practice [124]. The main disadvantage is that PFAS released into the air instead of captured in the foam may still end up in the environment through long-range transport [128].

### **4.3 Chemical Treatments**

PFAS in leachate can be effectively removed by chemical treatments using advanced chemical oxidation (photocatalytic oxidation or plasma technology). These treatments have removed 84-99.9% of PFAS [43].

Heterogeneous photocatalytic oxidation with semiconductor-based catalysts effectively degrades PFAS in landfill leachate under ultraviolet irradiation [129, 130]. A 3DP photocatalyst fabricated using polylactic acid compounded with  $\text{TiO}_2$  (15 wt%) and 3D printed into tiles effectively decreased concentrations of PFAS in landfill leachate; photocatalytic degradation was achieved for

most of the PFAS evaluated: PFOS, PFOA, PFHPA, PFHxS, PFNA, PFDA, and PFOSAm; greater than 80% removal of PFOS, PFNA, PFDA, and PFOSAm was achieved with 24-h of photocatalysis [129]. An adsorptive photocatalyst of iron-doped, carbon-modified composite (Fe/TNTs@AC) was also able to remove PFAS selectively; at a dosage of 10 g/L, fresh Fe/TNTs@AC removed >95% of 13 PFAS from the leachate within 2 h, 86% after first regeneration, and 74% when reused three times; Fe/TNTs@AC degraded >92% of 18 PFAS in 8 h under the field conditions, and when the PFAS-laden solids were subjected to the UV-H<sub>2</sub>O<sub>2</sub> system, approximately 84% of 16 PFAS in the solid phase were degraded; nevertheless, Fe/TNTs@AC was less efficient for PFBA and PFPeA owing to the transformation of longer-chain homologs into these short-chain PFAS and competition of adsorption sites by the longer-chain PFAS [130]. Extremely low or high pH values may alter photocatalysts' stability and catalytic activity [107].

Non-thermal plasma technology was evaluated for PFAS destruction in landfill leachate. Plasma discharge in an Ar atmosphere destroyed PFAS compounds present in six leachate samples; the highest removal efficiency was achieved for long-chain PFAA (>99.9% in 40 min) followed by PFAS precursors (99.9% in 60 min) and short-chain PFAA (10-99.9% after 120 min of plasma treatment [43]; process efficiency was not affected by the presence of high concentrations of organic matter but was enhanced by addition of cationic surfactants [43]. Slower removal of short-chain PFAS compared to long-chain compounds can be attributed to differences in surface activity and their production as byproducts from longer-chain compound destruction [131]. Some aspects of contaminated water, such as pH and organic matter or nitrate concentrations, can directly lower the effectiveness of non-thermal plasma treatment [132]. One of the main concerns associated with plasma is its high energy consumption in some cases. However, producing non-thermal plasma with an efficient reactor type and power supply sources can substantially reduce the energy requirements [133].

Fenton's oxidation, sonochemistry, sub-critical and supercritical water oxidation, microwave-hydrothermal treatment, and wet air oxidation have proved to degrade more than 90% of the PFAS (especially long-chain PFAS like PFOA and PFOS) in water [31, 115, 134]. Still, the degradation of PFAS in landfill leachates have not been evidenced.

#### **4.4 Electrochemical Oxidation**

Electrochemical oxidation can efficiently eliminate PFAS (up to 99.7%) via the combined effects of direct and indirect oxidation [135].

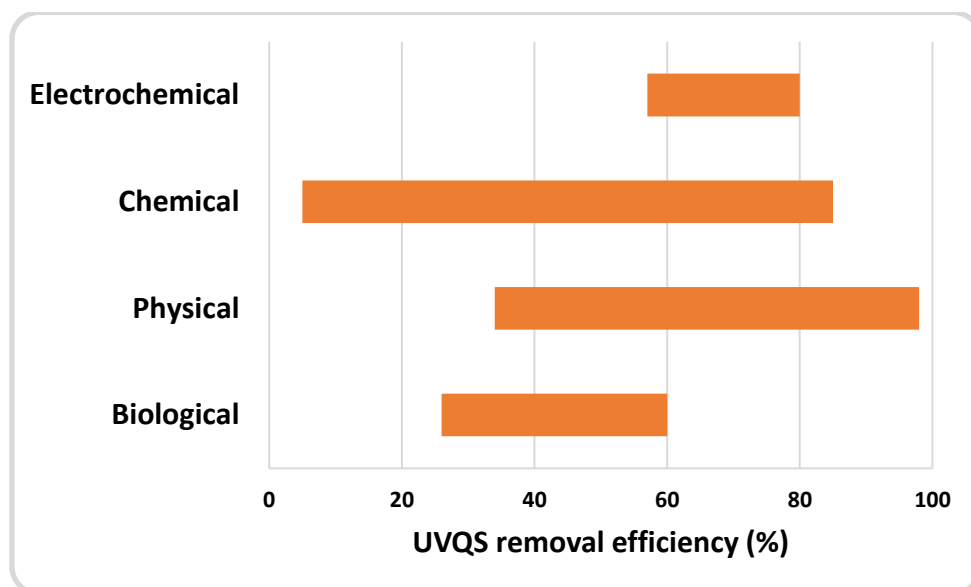
In electrochemical treatment of landfill leachates with boron-doped diamond electrodes, PFAS removal efficiency was high (99.7%) [107]; PFOA and PFOS removal were dependent on current density: at higher current density (75 mA/cm<sup>2</sup>) it was possible to obtain an average removal efficiency of 80% for PFOA and 78% for PFOS; at lower current density (25 mA/cm<sup>2</sup>), the efficiencies were reduced to half [136]; this is due to the competing reactions within a complex matrix [136]. In electrochemical treatment with Pd/Sn-doped electrodes (Pd (1%)-SnO<sub>2</sub>), long-chain PFAS in raw leachate were removed at an energy consumption of 20 Wh/L, but more than 100 Wh/L energy input was needed to remove about 80% of short-chain PFAS [43]; the required treatment time was inversely correlated with the chain lengths: PFOS and PFOA were rapidly removed at an energy input <20 Wh/L and a retention time of 18 min but the increase in short-chain PFAS (such as PFBA and PFBS) concentrations was prominent, probably due to the breakdown of PFAS precursors [43]; the

short-chain PFAS were further degraded by increasing the treatment duration [43]; a significant elimination of formed PFBA was achieved when energy input surpassed 400 Wh/L [43]. In electrochemical oxidation, the possible formation of toxic byproducts, such as HF, during treatment could be a problem [134]. Scaling up this technology is the major challenge for the full-scale use of electrochemical oxidation in PFAS treatment due to the low PFAS concentrations, high energy consumption, and costly acquisition of electrodes [137, 138]. Incorporating electrochemical oxidation after plasma treatment could be an effective strategy to remove short-chain PFAS without surfactant addition, although plasma alone with surfactant addition would be more energy effective [43].

## 5. Efficacy Comparison of the Optimal Treatments for UVQS and PFAS Removal

Figure 1 compares the removal efficiency of UVQS by the different treatments analyzed. According to the figure, the maximum removal efficacy sequence is:

Physical treatments > Chemical treatments > Electrochemical treatments > Biological treatments



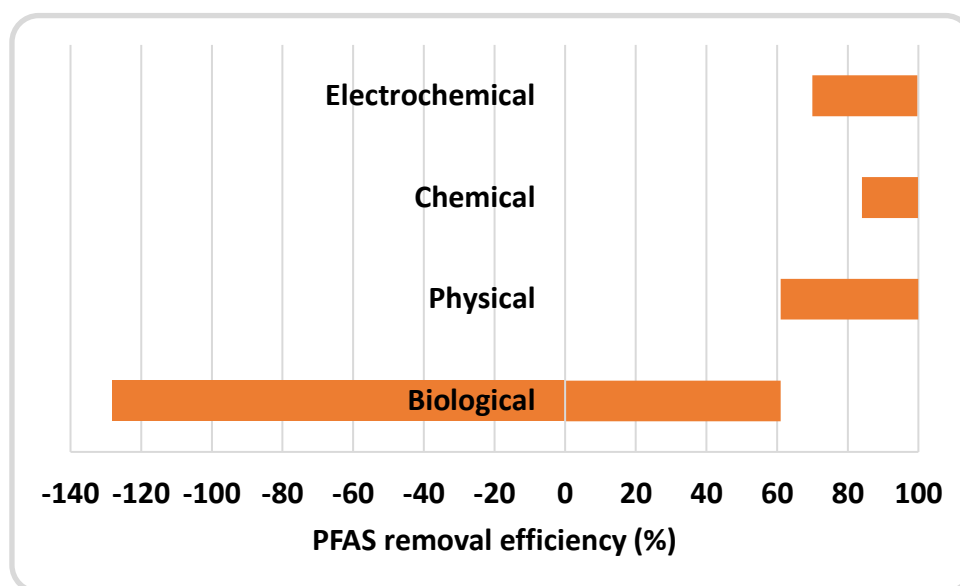
**Figure 1** UVQS removal efficiency (%) of the treatments.

In the best case, the biological treatments only removed 60% of the UVQS in landfill leachate [19]. However, electrochemical treatments worked efficiently when combined with biological treatments (anaerobic degradation + electrochemical process), achieving, on average, 80% UVQS removal [83, 84]. High removal efficiencies (85%) were obtained with chemical treatments like solar photo-Fenton system [70].

Physical treatments such as adsorption on powered active carbon [54] or laterite [55] resulted in the most efficient processes based on sorption with 95% and 97% UVQS removal, respectively. In comparison, osmosis was the most efficient membrane separation-based method, with 98% of UVQS elimination [56].

As shown in Figure 2, biological treatments are the least efficient option for PFAS removal. Significant variations in the elimination effectiveness of this treatment can be seen. For example, positive PFAS removal efficiency values were obtained in up-flow anaerobic sludge blankets (UASBs)

and denitrification units, whereas negative results were found in nitrification units [42]. The obtention of negative removal results, corresponding to the formation of PFAS in some biological systems, has been explained in section 4.1 [97, 99].



**Figure 2** PFAS removal efficiency (%) of the different treatments.

Similar maximum efficiency, nearly 100% (Figure 2), was observed for some physical, chemical, and electrochemical treatments:

- The adsorption on powdered active carbon [107], the reverse osmosis [117], and the membrane distillation [102] stand out as the best physical treatments for PFAS removal.
- The non-thermal plasma method was the most effective process among the chemical treatments [43].
- The boron-doped diamond electrodes contributed to the electrochemical treatment's highest PFAS degradation efficiency [107].

According to what has been discussed previously, two physical treatments (adsorption on powdered activated carbon and osmosis) can remove both UVQS and PFAS. This coincidence suggests that future landfill leachate studies should explore these treatment techniques to remove PFAS and UVQS simultaneously. As said in the Introduction Section, there are no studies on the simultaneous removal of PFAS and UVQS from landfill leachate; however, some research on adsorption or membrane-based processes has proved that these techniques are very efficient for the simultaneous removal of very different types of pollutants from wastewaters [139-144]. These observations justify the suggestion for future studies on the simultaneous removal of PFAS and UVQS from landfill leachate.

## 6. Conclusions

This review gives up-to-date information on recent findings in the UVQS and PFAS degradation field through physical, chemical, electrochemical, and biological treatments. Results from this current review showed that:

- Conventional biological treatments, like the activated sludge process, cannot remove UVQS or PFAS from the landfill leachate. Combinations of biological and physicochemical therapies, such as membrane bioreactors, can improve the efficiency of the process.
- Physical treatments show better effectiveness for emerging pollutants. Sorption-based processes result in an exciting option for removal, although the regeneration of sorbents limits their application once they are spent. Membrane-based processes have been revealed as a promising technology to remove UVQS and PFAS, having their performance controlled by the membrane's pore size and limited by the membrane fouling and concentrate management. Membrane distillation and foam fractionation are also remarkable options to eliminate PFAS restricted by membrane fouling and/or energy consumption.
- Chemical treatments, such as advanced oxidation-based processes, have also been capable of eliminating both UVQS and PFAS. The primary concerns associated with these procedures are often the high cost and complexity of operation that can restrict the potential use of these treatments.
- Electrochemical treatments, like electrochemical oxidation, can successfully remove UVQS and PFAS. The high energy consumption restricts the application of electrochemical processes. Combining electrochemical treatments with physical therapies, such as membranes, or chemical treatments, such as plasma processes, can improve the removal of UVQS or PFAS.

The overall evaluation of the treatments shows that technologies like adsorption or osmosis can efficiently remove PFAS and UVQS from landfill leachate. This conclusion suggests that both pollutants could be released simultaneously with a single treatment based on one of these simple, green, and efficient technologies. The simultaneous method would require lower costs and space compared to traditional methods that combine two or more techniques. In addition, the concurrent process could perhaps show synergistic removal performance. Therefore, future research in landfill leachate should be focused on the simultaneous removal of the UVQS and PFAS by one of these treatment methods, emphasizing the synergistic and economic effects of the process.

### Author Contributions

The author did all the research work of this study.

### Competing Interests

The author has declared that no competing interests exist.

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