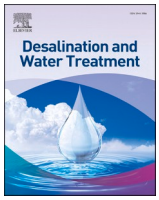
Desalination and Water Treatment 324 (2025) 101530

Contents lists available at ScienceDirect

Desalination and Water Treatment

journal homepage: www.sciencedirect.com/journal/desalination-and-water-treatment/



# A critical review, and up-to-date research progress on the occurrence, and removal of antiretroviral drugs in water

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ARTICLE INFO

*Keywords:*
Antiretroviral drugs
Toxicity
Contaminated water
Remediation
Degradation products

ABSTRACT

In the last decade, research on identification of antiretroviral drugs in water has intensified, especially in the developing countries. Concentrations reaching 33 μg/L for efavirenz have been reported in wastewater effluent which is released into the surface water. Some antiretroviral drugs have been found to possess toxic effects towards the aquatic organisms, and plants. This threatens the sustainability of high-quality vegetables as the water scarcity necessitates the practise of crop irrigation with used or treated water which is often contaminated with pharmaceuticals. This article reviewed the progress made on the removal of antiretroviral drugs in contaminated water, identified challenges, and gaps while outlining the possible future research direction. It observed that various degradation processes for antiretroviral drugs have been explored with limited information on the characterization of the degradation by-products, and their toxic effects. Adsorption process is also dominating for removal of antiretroviral drugs in water, while wetland removal, and plant uptake are least explored. An adsorption capacity as high as 833 mg/g has been reported for lopinavir using biochar modified with incorporating layered double hydroxide. Research on the removal of antiretroviral drugs in water is expected to continue with the inclusion of studies focussing on metabolites of these chemicals.

## 1. Introduction

In the past 40 years, antiretroviral drugs have been in existence as medications used for the treatment of human immunodeficiency virus (HIV) [1,2]. Their presence in source-separated urine serves as an indication of their excretion from humans as part of urine [3]. The concentration of lamivudine in source-separated urine from Lusaka (Zambia) was reported to reach 10010 μg/L [3]. Several antiretroviral drugs are excreted unchanged from the human body with the excretion rates for lamivudine, zidovudine, and nevirapine reported at 70, 16, and 2.7 %, respectively [4]. This means the geographical areas with high prevalence of HIV, and high consumption rates of antiretroviral drugs will have high quantities of these drugs in their environment. In source-separated urine from eThekwini (South Africa), emtricitabine was detected in 40 % of studied urine samples [5].

In addition to the detection of antiretroviral drugs in urine, their presence in wastewater treatment plants (WWTPs), and their discharge into the surface water through the effluent discharges has been reported in literature [6,7]. Due to their polar functional groups, and water solubility [8], it is difficult to remove antiretroviral drugs from water during the wastewater treatment process. For example, the water solubilities for zidovudine, lamivudine, and nevirapine are 20100, 70000, and 0.7046 mg/L, respectively [8]. Hence, these drugs have been found present in surface water [8,9]. Therefore, due to the limited removal of antiretroviral drugs in water, several removal strategies have been proposed. Such processes include the removal of antiretroviral drugs in water using adsorption [10], various forms of degradation [11–13], plant uptake [14], and wetland-based processes [15]. The removal of these drugs in water bodies is important as they pose risks in the environment. For example, the environmentally relevant concentrations for lamivudine have been found to pose ecological health risk at different trophic levels, to both flora, and fauna [16]. More concerning is the synergistic effects caused by the mixtures of antiretroviral drugs on Cyanobacterium *Microcystis novacekii*, where they presented increasing toxicity compared to individual drugs [17]. One of the most detected antiretroviral drugs, efavirenz, has been found to be highly toxic to two aquatic organisms, *Ceriodaphnia dubia*, and *Raphidocelis subcapitata* with $EC_{50}$, and $IC_{50}$ values of 26 μg/L, and 34 μg/L, respectively [18].

Research on the occurrence of antiretroviral drugs in water bodies intensified in the last decade. Similarly, studies investigating the

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https://doi.org/10.1016/j.dwt.2025.101530
Received 16 September 2025; Received in revised form 14 October 2025; Accepted 24 October 2025
Available online 25 October 2025

removal of these drugs in water increased in the last decade with great focus on degradation, and adsorption. As this is a relatively new study area, there are still very few review articles that extensively critique the available information on the occurrence, and removal of antiretroviral drugs in water. The review article published by our research group in the last three years focused on the contamination of water bodies in Kenya, and South Africa by antiretroviral drugs, and their removal strategies [19]. This was motivated by the high number of studies that report the occurrence of these drugs in Kenyan, and South African waters than anywhere else in the world. Other relevant reviews in this subject focussed on the contamination of water resources in the African continent at large [20], and Brazil [21]. Some studies on remediation reviewed pharmaceuticals in general rather than solely focussing on antiretroviral drugs [22–24]. Additional review articles directed towards the antiretroviral drugs limited their scope to the treatment of wastewater with algae-mediated processes [25], and photocatalytic degradation [26].

Therefore, there is a need for the availability of a detailed review article in open literature which focus on the critical assessment of available information reporting the occurrence, and removal of antiretroviral drugs in contaminated water. Hence, the aim of this article was to provide a comprehensive assessment of all the removal strategies proposed for the removal of antiretroviral drugs in contaminated water. This was necessary to establish the explored processes, their successes, and shortfalls, while also analysing research gaps to inform the direction for future studies. The scope of the present review included discussion on environmental occurrence of antiretroviral drugs, and toxicity of these drugs to emphasise on the importance of their removal in water bodies.

## 2. Occurrence of antiretroviral drugs in water

The research focussing on the determination of antiretroviral drugs in the aquatic environment has intensified in recent years. Several antiretroviral drugs have been found in high concentrations in water from African countries, especially Kenya, and South Africa, which are known for the high prevalence of HIV, and AIDS [27]. In fact, one of the first studies to establish the occurrence of antiretroviral drugs in African water bodies (Nairobi, Kenya) found three antiretroviral drugs: lamivudine, zidovudine, and nevirapine, with higher concentrations than those in the literature at the time [28]. As a result, there are articles in the literature which solely critically reviewed the prevalence of antiretroviral drugs in water bodies of the African countries [27,29,30]. The examination of the prevalence of these drugs in African waters was possibly necessitated by their frequent detection with high concentrations when compared to other pharmaceuticals.

As expected, the quantities of antiretroviral drugs reaching the WWTPs are large, with one study reporting the efavirenz concentration reaching 34 μg/L in a WWTP-influent from South Africa [31]. Other antiretroviral drugs reported by the same study with high concentrations include zidovudine, darunavir, and raltegravir with the influent levels reaching 53, 43, and 17 μg/L, respectively. Even worse, the same study reported poor removal of the efavirenz during the wastewater treatment with up to 33 μg/L found in the effluent of a WWTP. Interestingly, a different South African-based study established the occurrence of the metabolite of efavirenz, 8,14-dihydroxy-efavirenz, in wastewater at concentrations sometimes similar to those of the parent compound [32]. This points to a possibility of metabolization of efavirenz in the environment with a potential of metabolites being transformed into the parent compound at a later stage. Similarly, a metabolite of nevirapine, 2-hydroxy-nevirapine, has also been detected in wastewater [32]. The inability to efficiently remove antiretroviral drugs in WWTPs contributes to their high concentrations in surface water. For example, lopinavir, and efavirenz have been reported with concentrations of up to 38 μg/L, and 24 μg/L in surface water, respectively [33].

Concerningly, antiretroviral drugs have also been detected in drinking water sources [34]. In South Africa, efavirenz was the most dominant contaminant of emerging concern among others in raw drinking water with concentrations reaching 1.1 μg/L in raw water [34]. The other antiretroviral drug found in drinking water by the same study was emtricitabine, with both compounds detected in all the stages of the drinking water treatment process including post post-chlorination stage [34]. Antiretroviral drugs have also been detected in groundwater with nevirapine once reported with concentrations of 1.2–1.6 μg/L [35]. Furthermore, the escape of antiretroviral drugs into the oceans has been reported with compounds such as efavirenz, and lamivudine being detected in seawater [36,37]. These discoveries showcase a need to investigate the toxicity antiretroviral drugs towards aquatic organisms, and any species that utilize the surface water, while also establishing effective procedures for their elimination in water.

## 3. Toxic effects, and health risk associated with the occurrence of antiretroviral drugs in water

Antiretroviral drugs have been found to pose environmental risks, and linked to toxic effects towards aquatic organisms [17,18,38]. In this context, efavirenz has been found with a risk quotient exceeding the value of 1 as an indication of presenting high ecological risks towards both *Ceriodaphnia dubia*, and *Raphidocelis subcapitata* [18]. In the same study, the risk quotient for lamivudine only exceeded 1 in the case of *Ceriodaphnia dubia*. A different study observed a DNA damage, and oxidative stress when *Ceriodaphnia dubia* was exposed to μg/L of ribavirin, and tenofovir [39]. When lamivudine was exposed to different species, the findings suggested that it poses an ecological health risk at different trophic levels to both flora, and fauna [16]. This means plants exposed to the environment contaminated with antiretroviral drugs may also be at risk as presented elsewhere [40]. Such reports are translated to a need for the removal of antiretroviral drugs in contaminated water. The other study found that efavirenz exposure to mice might induce genotoxicity in the brain [41], thus, further demonstrating the importance of continuous environmental monitoring of antiretroviral drugs, and the urgent need to establish procedures for the removal of these drugs in the environment. In fact, efavirenz is reported to have more harmful effects than other antiretroviral drugs such as tenofovir [42], and nevirapine [41]. Oxidative stress alterations were observed for Crustacean *Daphnia*, and magna exposed in river water contaminated with efavirenz, and tenofovir [42]. Suggestions on the toxic effects due to the presence of antiretroviral drugs in seawater have also been documented [43]. This summary emphasises the need to seek procedures for the removal of antiretroviral drugs in water. In addition, the detection of antiretroviral drugs in drinking water sources (as discussed earlier) suggests a need to conduct the human health risk assessment due to the unintentional exposure and possible consumption of these drugs through drinking water by humans. As research focussing on investigating the occurrence of antiretroviral drugs in drinking water sources is still minimal, its execution should be performed alongside the human health risk assessment to understand human health effects due to their presence in drinking water.

## 4. Removal of antiretroviral drugs in wastewater

In recent years, it has become a common knowledge that WWTPs are unable to remove pharmaceuticals during the sewage wastewater treatment processes. Factors influencing the removal efficiency are complex, and sometimes depend on the physicochemical properties of the respective pharmaceuticals, and the wastewater treatment process. The removal efficiencies reported for selected antiretroviral drugs in selected WWTPs are presented in Table 1. These removal efficiencies are determined based on the concentration of each drug detected in the wastewater effluent in comparison to the amount in the influent of a WWTP. Three different WWTPs in Durban (South Africa) reported negative removal efficiencies for an antiretroviral drug, lopinavir [31].

**Table 1**
Removal efficiency of antiretroviral drugs in WWTPs.

| Antiretroviral drug | WWTP location | Removal efficiency (%) | Reference |
|---|---|---|---|
| Abacavir | KwaZulu-Natal (South Africa) | 75–91 | [44] |
| Efavirenz | KwaZulu-Natal (South Africa) | 6–53 | [44] |
| | Western Cape (South Africa) | −38–40 | [32] |
| | Limpopo, South Africa | 47–82 | [47] |
| | Eastern Cape, South Africa | 2–12 | [48] |
| | Gauteng (South Africa) | 0–70 | [49] |
| | Gauteng (South Africa) | 27–71 | [45] |
| | KwaZulu-Natal (South Africa) | 19–92 | [50] |
| Emtricitabine | Western Cape (South Africa) | 76–100 | [32] |
| Lamivudine | Western Cape (South Africa) | 100 | [32] |
| | Gauteng (South Africa) | > 70 | [49] |
| | Juju Town (Kenya) | −56 | [51] |
| | Kisumu City (Kenya) | 24–59 | [35] |
| Nevirapine | KwaZulu-Natal (South Africa) | 44–87 | [44] |
| | Western Cape (South Africa) | 3–100 | [32] |
| | Kisumu City (Kenya) | 11–49 | [35] |
| Zidovudine | Juju Town (Kenya) | 41 | [51] |
| | Kisumu City (Kenya) | > 95 | [35] |

This means the concentration of this drug in the effluent exceeded the amount found in the influent. However, antiretroviral drugs such as zidovudine, and lamivudine were sufficiently removed by the same WWTPs [31]. A different study conducted within the same geographical location reported removal efficiencies of 44–87 %, 6–53 %, and 75–91 %, for nevirapine, efavirenz, and abacavir, respectively (Table 1) [44]. The limited removal of these drugs in wastewater, and their increase from the influent to the effluent is linked to several processes. For example, the increase in nevirapine concentrations from the influent to the effluent of a certain WWTP was linked to the de-conjugation of the hydroxylated metabolites of nevirapine in the WWTP, its resistance to degradation, and the lack of binding of the nevirapine to the sludge [45]. In some cases, the treatment processes do not remove antiretroviral drugs in water but transform these drugs into other forms. It has been reported that the chlorination process which is usually the last step of the wastewater treatment process only transforms antiretroviral drugs such as nevirapine [46].

## 5. Removal strategies of antiretroviral drugs in water

### 5.1. Adsorption

Based on Table 2, numerous kinds of adsorbents have been explored for the adsorption of antiretroviral drugs in water. Such adsorbents range from the synthetic materials such as molecularly imprinted polymers (MIPs) to the modified agricultural waste such as macadamia nutshells. Table 2 is indicative that many studies investigate the adsorption of single drugs in water, rather than the multi-drug adsorption demonstrated in few cases. Antiretroviral drugs are consumed simultaneously in groups of 2–3 active ingredients as demonstrated through the introduction of the combination therapy. Studies in literature also showed the co-existence of various antiretroviral drugs in water bodies [7,52]. This immediately showcase a need to establish strategies for the simultaneous removal of several antiretroviral drugs in water. Some adsorption studies have already started investigating the multi-drug (antiretroviral drugs) adsorption from contaminated water [53,54]. Although such studies encounter the competitive binding of adsorbates into the adsorbents, promising results showcasing efficient adsorption of antiretroviral drugs have been reported. For example, co-adsorption of didanosine, ritonavir, efavirenz, and other pharmaceuticals onto nanofibers fabricated from *Mondia whitei* root extract yielded adsorption capacities in the range of 75–320 mg/g [55]. Another study reported the removal of three antiretroviral drugs; abacavir, nevirapine, and efavirenz, with exfoliated graphite which resulted in adsorption capacities reaching 237 mg/g [56].

**Table 2**
Adsorption of antiretroviral drugs in water with different adsorbents.

| Antiretroviral drug | Adsorbent | Adsorption mechanism | Adsorption capacity (mg/g) | Reference |
|---|---|---|---|---|
| Abacavir | MIP | Hydrophobic, and electrostatic interactions | 157 | [59] |
| Abacavir | MIP | Electrostatic interactions | 5.98 | [62] |
| Emtricitabine | MIP | Electrostatic interactions | 4.65 | [63] |
| Efavirenz | Graphene wool | Hydrogen bonding, covalent bonding, hydrophobic interaction, and π-π interactions | 4.41 | [53] |
| Nevirapine | | | 48.3 | |
| Efavirenz | Calcined layered double hydroxide clay | Hydrogen bonding | 2.73 | [64] |
| Nevirapine | | | 2.93 | |
| Lamivudine | Coal discards, and sewage sludge derived-hydrochar | Not clearly defined | 41–52 | [10] |
| Nevirapine | | | 42–50 | |
| Lamivudine | Jamun seed (*Syzygium cumini*) biochar | Not clearly defined | 400 | [57] |
| Lopinavir | Biochar modified by incorporating layered double hydroxide | Hydrogen bonding, π–π interactions, and minimal electrostatic forces | 833 | [58] |
| Tenofovir | Activated carbon produced from maize cobs | Van der Waals interactions, and electrostatic attraction | 145.2 | [65] |
| Tenofovir | Sodium alginate-encapsulated activated carbon | Electrostatic interactions | 8.40 | [66] |
| Zidovudine | Tea-waste-based biochar | Electrostatic interactions | 20.2 | [67] |
| Ritonavir, and lopinavir | Sewage sludge | Van der Waals forces, coordination bonding, hydrogen bonding, electrostatic interactions, hydrophobic, and π-π, π-n interactions | 7.55–8.71 | [54] |
| Abacavir | Exfoliated graphite | Hydrogen bonding, intra-particle diffusion, and electrostatic attraction | 1.66–197 | [56] |
| Nevirapine | | | 1.66–233 | |
| Efavirenz | | | 1.65–237 | |
| Abacavir | Activated macadamia nutshells | Electrostatic interaction, and π–π interactions | 14.25–27.44 | [68] |
| Nevirapine | | | 10.79–13.67 | |
| Efavirenz | | | 20.8–38.2 | |
| Didanosine, ritonavir, efavirenz, and other pharmaceuticals | Nanofibers fabricated from *Mondia whitei* root extract | Intraparticle diffusion, hydrogen bonding, electrostatic attraction or/and peptide bond interaction | 75–320 | [55] |

Among the reported adsorbents, the biochar-based adsorbents gave promising adsorption capacities reaching 800 mg/g. The explored adsorbents in this case are jamun seed (*Syzygium cumini*) biochar, and biochar modified by incorporating layered double hydroxide prepared for the removal of lamivudine [57], and lopinavir [58], respectively. For the latter, with the maximum adsorption capacity of 833 mg/g, different mechanisms which include hydrogen bonding, π–π interactions, and minimal electrostatic forces were reported to have an influence on the adsorption process [58]. Other adsorption mechanisms driving the adsorption of different antiretroviral drugs are given in Table 2. Furthermore, some of the adsorption processes are explained in Fig. 1, with different functional groups in the antiretroviral drugs promoting the different forms of interactions such as hydrogen bonding, with the adsorbent [53].

Among the other reported adsorbents for antiretroviral drugs, MIPs formed part of the first groups of adsorbents to be evaluated for the adsorption of antiretroviral drug, abacavir, in water [59]. MIPs are known in literature for their selectivity [60,61]which is induced through their synthetic procedures. These materials have been applied for the selective adsorption of abacavir [62], and emtricitabine [63] in water. Due to their selectivity, perhaps this type of adsorbent could be more useful for removal of selected drugs in industrial wastewater rather than the municipal wastewater which contains a concoction of different drugs.

### 5.2. Degradation

#### 5.2.1. Chlorination, and ozonation

Degradation process is one of the most explored approaches for the elimination of antiretroviral drugs in water, with some applications presented in Table 3. In this process, antiretroviral drugs present in water are transformed into other products (supposedly safer chemicals).

Both chlorination, and ozonation are important processes in the purification systems which are often used for disinfection. As described in an earlier section, the conventional water treatment processes are unable to completely remove antiretroviral drugs in water. Hence, various research groups have investigated the potential of employing both ozonation, and chlorination for the elimination of these drugs in water [69–73]. The success of such processes would mean that disinfection, and antiretroviral drug removal occur at the same step, thus avoiding the implementation of additional step in the water/wastewater treatment just for the purpose of dealing with the presence of antiretrovirals. However, a detailed investigation of such removal strategies is important as processes such as chlorination in water/wastewater treatment when residues of organics are present in water have been linked to the formation of disinfection by-products which could be dangerous than the parent compounds [74].

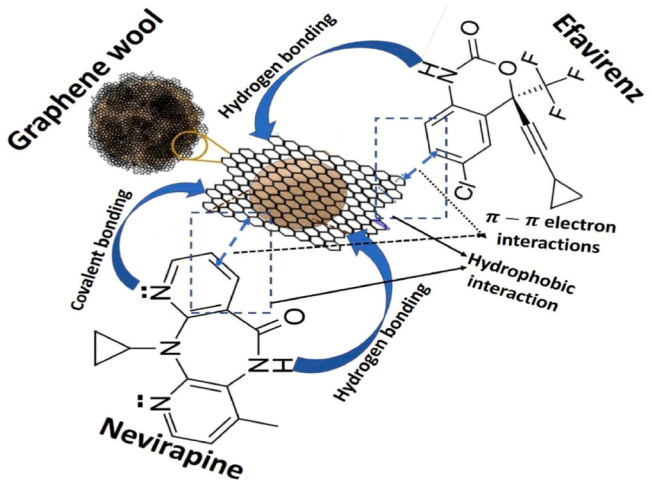


**Fig. 1.** Some mechanisms showing the interactions between graphene wool (adsorbent), and efavirenz as well as nevirapine as representatives of antiretroviral drugs [53]. Reused with permission from Elsevier.

The use of chlorine for the removal of antiretroviral drugs in contaminated water has been reported in literature [69–71,73]. One of the antiretroviral drugs often found in surface water, nevirapine, was reported to be resistant to degradation through the use of chlorination [69], thus necessitating a need to establish more robust procedures for elimination of this drug in the water system. Furthermore, its chlorination by-products were found to retain antiviral activity, although they were less toxic than the parent compound [69]. Advanced oxidation processes which combine the chlorine treatment with UV showed promising results for the removal of selected antiretroviral drugs [73]. A different study established that the degradation of antiretroviral drugs (nevirapine, and efavirenz) is influenced by the chlorine dosage, and lead to the formation of trihalomethanes [71]. These results show that a simple disinfection step cannot necessarily eliminate antiretroviral drugs in water without producing a secondary pollution.

On the other hand, ozonation of antiretroviral drugs has been less studied when compared to chlorination. One study that investigated the ozonation of zidovudine reported the formation of nine transformation products [72]. Three of these transformation products were identified as hydroperoxides. This means both the ozonation, and chlorination are unable to completely degrade antiretroviral drugs into carbon dioxide, and water which could be much safer than the transformation products which are organic chemicals themselves. Therefore, more research work is still needed to achieve a complete degradation, and removal of these chemicals, and their transformation products in water. In general, literature suggests that the degradation of organic contaminants including pharmaceuticals (i.e. antiretroviral drugs) is described by the pseudo-first-order kinetic model [12]. This was observed for the degradation of several antiretroviral drugs discussed in this paper which include nevirapine, lamivudine, and zidovudine [12].

#### 5.2.2. Photocatalytic degradation

As demonstrated by several review articles in literature [91,92], photocatalytic degradation is well investigated for the removal of pharmaceuticals in contaminated water. However, there still seems to be very few research articles focussing on antiretroviral drugs. Most of the available information seem to investigate photocatalytic degradation of single antiretroviral drugs [75,83], despite the available information suggesting the co-occurrence of many antiretroviral drugs in water, and wastewater samples [7,52]. Some applications of photocatalytic degradation of antiretroviral drugs in water are summarized in Table 3. The available information shows the breakdown of antiretroviral drugs into smaller molecules. For example, the photocatalytic degradation of nevirapine with a heterostructure of few-layer black phosphorus coupled with niobium (V) oxide nanoflowers resulted in the formation of 7 smaller degradation products which included *n*-butanol [83]. A different study discovered 46 intermediates during the photocatalytic degradation of abacavir arising from different transformation routes [75]. The same investigation reported the transformation routes to be influenced by the sample matrix, with additional information suggesting different transformation rates in different matrices [82]. While one study established four major transformation products for the same antiretroviral drug, abacavir [75]. This could be translated to the formation of different transformation products, and different routes for their formation as a result of different experimental routes such as the application of different photocatalysts. Overall, the degradation process for these chemicals is influenced by several factors (Table 3) which include the nature of the catalyst, and the concentration of antiretroviral drug in the system [93].

In the case where photocatalysis was investigated for simultaneous degradation of lamivudine, and zidovudine, the degradation products (not characterized) were linked to inhibition of the growth of lettuce,

**Table 3**
Degradation methods for removal of selected antiretroviral drugs in water.

| Antiretroviral drug | Degradation material | Optimized degradation process, and outcomes | Degradation efficiency (%) | Degradation products | Reference |
|---|---|---|---|---|---|
| Abacavir | Titania-graphene oxide nanocomposites | 2 % graphene oxide within 20 min | 100 | Forty-six intermediates | [75] |
| Abacavir | Electrochemical degradation with penetration flux porous Ti/$SnO_2$-Sb anode | 10 min at a current density of 0.2 mA/$cm^2$ | > 97 | Not named | [76] |
| Efavirenz | Immobilized *Trametes versicolor* laccase on $Ti_2NT_x$ MXene | 240 min | 65 | Six by-products given | [77] |
| Efavirenz | Dielectric barrier discharge plasma | Discharge power of 59.3 W, and pH 9 | 93 | Six degradation by-products | [78] |
| Lamivudine | Photo-peroxidation degradation | Degradation in the presence of UV-C radiation, and hydrogen peroxide | 95 | Not named | [79] |
| Lamivudine | Graphene oxide, and Yb co-modified $PbO_2$ electrodes | Initial concentration of 100 mg/L, current density of 30 mA/$cm^2$, electrolyte concentration of 0.1 M, and pH of 9 | 100 | Carbon dioxide, water, and other inorganic ions | [80] |
| Lamivudine | $Bi_2O_3$−$TiO_2$ supported by powdered activated carbon under a Xe lamp | Initial concentration of 10 mg/L, pH of 7, and catalyst dose of 250 mg/L, 120 min | 57 | Propionamide, 2-mercaptoethanol, and diethylene glycol | [81] |
| Maraviroc | Peroxymonosulfate, and $TiO_2$ photocatalytic oxidation | Light-activated peroxymonosulfate halved the drug concentration after 7 min of irradiation, when combined with $TiO_2$, the recorded half-life was decreased to 0.47 min | | Twenty-two transformation products | [82] |
| Nevirapine | Heterostructure of few-layer black phosphorus coupled with niobium (V) oxide nanoflowers | 3 hrs with 5 mg/L initial concentration solution of nevirapine, at a working pH of 3, and 15 mg of photocatalyst | 68 | Butanol | [83] |
| Nevirapine | Dielectric barrier atmospheric non-thermal plasma discharge | 40 min, discharge power of 63.5 W, and plasma working gas of atmospheric air | > 94 | Six transformation products | [84] |
| Nevirapine | UV/$TiO_2$/$H_2O_2$ system | 60 min of irradiation, pH of 3 | 89 | Not detected by LC-MS | [85] |
| Nevirapine | Immobilised cross-linked laccase, and tyrosinase embedded within a membrane | 5 mg/L nevirapine at pH 7 | 99 | 5,6-dihydroxyindoline−2-carboxylic acid, 1-ethylquinolium, 3-hydroxy benzodiazepine, and N-(4-phenoxyphenyl)−1,3,5-triazine−2,4-diamine | [11] |
| Tenofovir disoproxil | Biodegradation with cyanobacteria/bacterial culture | Initial concentration of 25 mg/L, 16 days | 94 | Tenofovir isoproxil monoester | [86] |
| Tenofovir | Electro-persulfate degradation process | 30 min at pH 3 | 99 | Not mentioned | [87] |
| Efavirenz, Nevirapine | Visible light-activated Ag-AgBr-LDH nanocomposite catalyst | 8–12 hrs at neutral pH, under visible light irradiation | 80<br>100 | Not reported | [88] |
| Lamivudine, Zidovudine | Brass plates calcined at 500 °C, and coated with $TiO_2$ under UV-C irradiation | Coating ratio of 0.6 mg/$cm^2$, 300 min (15 mg/L of each drug) | 38–71<br>37–60 | Not mentioned | [89] |
| Lamivudine, Zidovudine | Heterogeneous photo-Fenton process applying pyrite as a catalyst, and artificial solar radiation | Aqueous solution treated for 210 min, and synthetic effluent treated for 420 min | 94–97<br>> 99 | Not identified | [90] |
| Nevirapine, Lamivudine, Zidovudine | Electro-Fenton degradation using iron-supported carbon-cloth | pH 3 | 25<br>25<br>31 | Not identified | [12] |
| Emtricitabine, Lamivudine, Tenofovir | UV/$TiO_2$/$H_2O_2$ hybrid system | Optimized parameters were pH, pollutant concentration, $TiO_2$ loading, $H_2O_2$ dosage, and time | 93<br>92<br>95 | Not detected by LC-MS | [13] |

carrot, and tomato seeds; thus, suggesting superior toxicity of degradation products when compared to the main drugs [89]. Although the simultaneous degradation of antiretroviral drugs is a great initiative, such processes still need major consideration. For example, the origin of toxicity caused by the degradation products reported elsewhere [89] is not clear. In fact, the toxicity has also been observed after the degradation of single drugs such as lamivudine [79]. It is not known if such toxic products originate from lamivudine or zidovudine or its degradation products that are common from both drugs. At the same time, the degradation process might be more efficient under certain experimental conditions for one drug over the other. For example, it was found that neutral pH was more efficient for the degradation of lamivudine, and tenofovir, whereas the same study observed acidic pH to be more ideal for degradation of emtricitabine when using UV–Vis/$TiO_2$/$H_2O_2$ hybrid system [13].

### 5.2.3. *Electrochemical degradation*

Electrochemical degradation of antiretroviral drugs in contaminated water has been evaluated [87,94–97]. An electro-Fenton system previously discussed elsewhere [98] was reported to remove 100 % of lamivudine within an hour, with five identified transformation products [99]. This system is showing promising results as it was also reported to partially transform zidovudine (31 %), nevirapine (25 %), and lamivudine (25 %), with degradation facilitated on iron-modified carbon-cloth electrode [12]. In a different context, the degradation of lamivudine was reported to be influenced by the type of the electrochemical process [100]. In this case, peroxi-photoelectrocoagulation process was found to be the most suitable for electrochemical degradation when compared to photoelectrocoagulation, and peroxi-electrocoagulation [100]. In electrochemical oxidation of lamivudine, its removal efficiency was 99.6 % when pollutant concentration in the system was 100 mg/L, with the current density, electrolyte concentration, and the initial pH of 30 mA/$cm^2$, 0.1 M, and 9.0, respectively [80]. This means there are many factors that could influence the electrochemical degradation of antiretroviral drugs. Notably, the initial concentration of the antiretroviral drugs is one of the factors with a

potential to influence the degradation process. However, one study reported that the initial concentration of nevirapine was not a limiting factor during its degradation using dielectric barrier atmospheric non-thermal plasma discharge [84]. When the same degradation process was applied for efavirenz, six degradation products were identified [78]. Furthermore, there seems to be ongoing research focussing on investigating the suitable electrode materials that promote the degradation process, with materials such as Ti/$SnO_2$-Sb/Ce-$PbO_2$ [76,96,97], and boron-doped diamond [101] already proposed.

The performance of electrochemical degradation has been compared to chemical degradation methods for selected antiretroviral drugs [102, 103]. In this context, electrochemical degradation of abacavir on platinum, and boron-doped diamond was found to yield the same two degradation products obtained when the degradation was performed with 3 % hydrogen peroxide [102]. However, the advantage of electrochemical degradation in this case was the rapid formation of the degradation products which was established within 10 min [102]. Although the degradation products have not been elucidated in these case studies, one study reported abacavir degradation to form an unknown degradation product with an *m/z* value of 299 [103]. Such information opens research opportunities for the structural elucidation of degradation products which need to be followed by investigating their toxicity towards aquatic organisms, and humans.

#### 5.2.4. Biocatalytic degradation processes

There is other degradation processes proposed in literature, although less explored for removal of antiretroviral drugs in water. For example, a biocatalytic degradation of nevirapine in water with immobilised cross-linked laccase, and tyrosinase embedded within a membrane has been reported [11]. In this application, 99 % of 5 mg/L nevirapine was removed from water solution with a pH of 7. Although the nature of degradation products from this process is still unknown, this could be an interesting solution to antiretroviral pollution as the removal process is conducted in realistic pH conditions expected in the environment. Also, the removal of tenofovir disoproxil with the cultures of *Microcystis novacekii* with the bacteria *Pseudomonas pseudoalcaligenes* has been investigated [86]. In this case, the biodegradation process lasted 16 days with a maximum removal efficiency of 94 % (starting concentration range was 12.5–50 mg/L) [86]. In a different study reporting a biocatalytic degradation of efavirenz with immobilized *Trametes versicolor* laccase on $Ti_2NT_x$ MXene, six degradation products (shown in Fig. 2) were identified within 240 min which corresponded to 65 % removal of the parent compound [77]. Similarly to the other degradation studies, more research studies still need to be conducted to establish efficient processes for the biocatalytic degradation of antiretroviral drugs in water. Ideally, suitable enzymes that could speed-up the degradation process which produce environmentally friendly by-products are needed.

### 5.3. Plant uptake

The plant uptake of pharmaceuticals from contaminated water, and soil is well studied with reports suggesting the accumulation in different plant species [104]. Antiretroviral drugs have been detected in water hyacinth plants sampling from surface water, thus indicating the accumulation of these chemicals in plants growing in the contaminated environment [52]. Although the same antiretroviral drugs (emtricitabine, tenofovir disoproxil, and efavirenz) were detected in aerial tissues, it was roots found with highest concentrations of these drugs from 7.4 to 29.6 μg/kg [52]. The accumulation of these antiretroviral drugs in this invasive species which has also been reported to absorb other organics [105] serves as indication to investigate plants that are suitable for removal of antiretroviral drugs in water through phytoremediation. To further demonstrate the plant uptake of antiretrovirals, nevirapine, lamivudine, and efavirenz were found to accumulate in lettuce grown hydroponically [14].

The plant uptake of these drugs suggests their possible removal in contaminated water through phytoremediation. *Lemna minor* has been found to have potential to remove tenofovir, lamivudine, and efavirenz from contaminated water [106]. However, extensive research studies

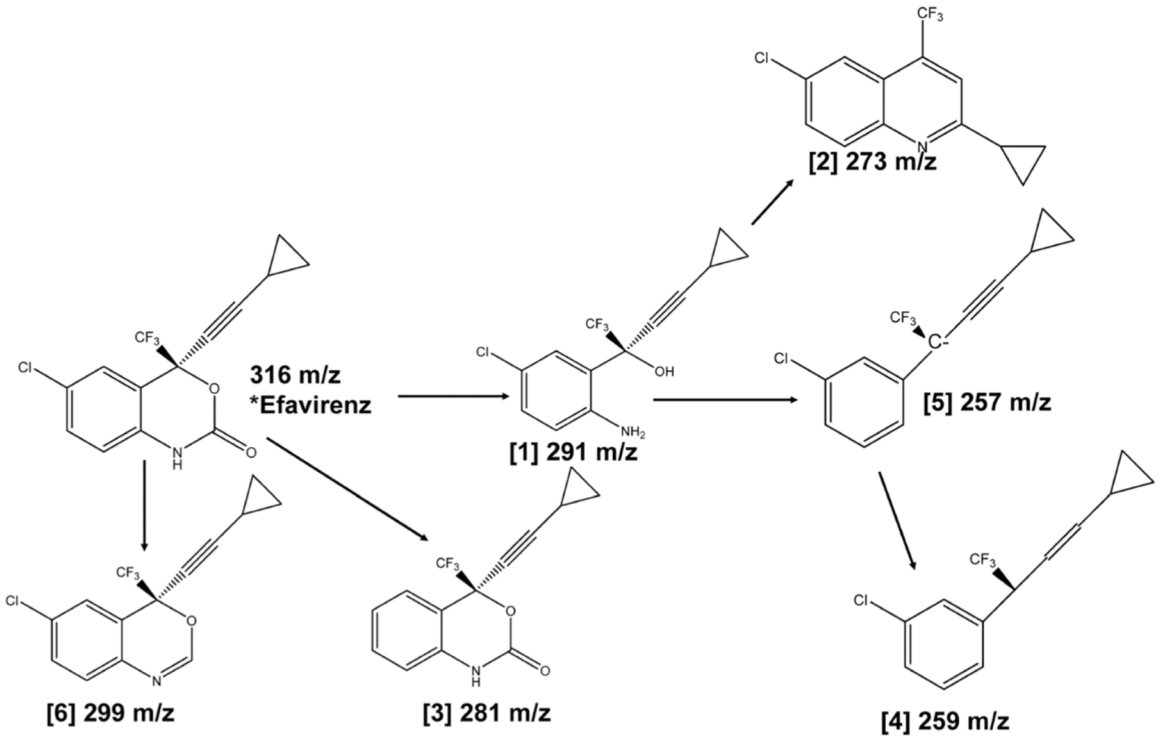


**Fig. 2.** Degradation products of efavirenz identified from its removal with immobilized *Trametes versicolor* laccase on $Ti_2NT_x$ MXene [77]. Permission for reuse is not required as the Figure is published in an open access article distributed under the terms of the Creative Commons CC-BY license which permits unrestricted use.

are still required to find suitable plant(s) for remediation of antiretroviral drugs in water. Thus far, most research studies in this subject largely focus on the antiretroviral drug uptake by vegetables [107] with potential outcomes suggesting food contamination than remediation.

### 5.4. Wetland removal

Although limited studies have been conducted, the potential of using wetlands in the format of open-water unit process wetlands for the removal of antiretroviral drugs in water has been explored [15,108]. In this case, the removal of abacavir, emtricitabine, lamivudine, and zidovudine in water was investigated in laboratory microcosm experiments simulating an open-water unit process wetland receiving municipal wastewater effluent [108]. The findings showed photo-transformation as the main removal mechanism for abacavir, zidovudine, and emtricitabine, while lamivudine was mostly removed through slower microbial processes [108]. Similarly, in a different study, photo-transformation was found to be responsible for the removal of other organic contaminants which included pharmaceuticals such as antiretroviral drugs, and antibiotics [15]. However, other processes such as biotransformation, and ozone treatment assisted in improving the removal of organic contaminants in water [15]. The processes, and mechanisms explaining the removal of emerging contaminants in general using the wetlands are described in a review article presented by Overton, and colleagues [109]. As shown in Fig. 3, removal of organics in wetlands could be based on the combination of different mechanisms which include adsorption, plant uptake, degradation, etc. This means this removal strategy which has not received much attention has a potential provided that suitable plant species for the removal of antiretroviral drugs are established. Limited attention on the removal of antiretroviral drugs in water through wetlands could be a result of fewer studies reporting these contaminants (as compared to other pharmaceuticals such as antibiotics) in water bodies across the world. Research on the environmental occurrence of these drugs intensified in the last decade with more studies performed in Africa. It is South Africa and Kenya at the forefront of this research where the major focus is still on understanding the fate of these drugs and their spread in the environment. Other African countries are lagging behind which means it is Kenya and South Africa which should intensify their research on remediation studies which include investigations on the potential of wetlands to remove antiretroviral drugs in water.

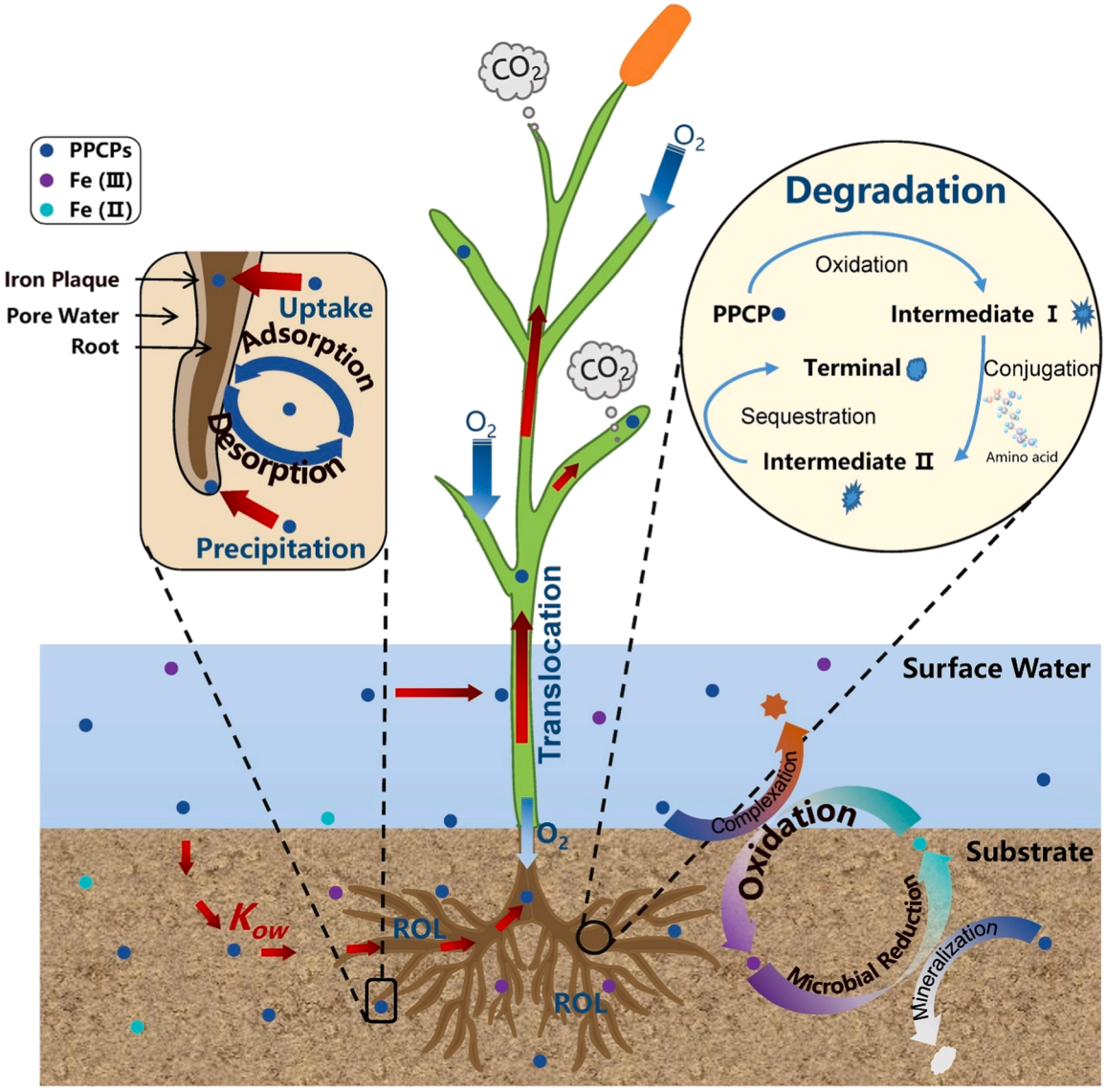


**Fig. 3.** The role of plants in removal of pharmaceuticals, and personal care products (PPCPs) by a constructed wetland [110]. Reused with permission from Elsevier.

## 6. Future perspectives

In the case of adsorptive removal, reviewed literature suggests that the most applied adsorbents were designed for the removal of single antiretroviral drugs in contaminated water. While few cases have been presented for the removal of two drugs or more (Table 3). This probably show these adsorbents as potential tools to eliminate antiretroviral drugs from water, and were designed as the proof-of-concept that they can target drugs with certain functionalities. However, in most cases, the selectivity of these adsorbents has not been tested. Selectivity is the crucial functionality of the adsorbents as it gives indication on the preferential recognition of adsorbate(s), failing which could result in early saturation of the adsorption sites. Thus, leading to poor performance towards the intended drugs. Research on multi-drug removal is needed for the elimination of a wide range of antiretroviral drugs, and other pharmaceuticals. This is necessary as these compounds are found as group of water contaminants in the same groups of samples.

Challenges related to the degradation of pharmaceuticals in water sometimes include the inability to identify the degradation products. The identification of the degradation products in water requires extensive analytical skills with the analyst expected to be able to operate, and interpret the analytical data from the chromatographic hyphenated techniques such as GC-MS, and LC-MS. At the same time, a deep understanding of organic chemistry is required to understand the transformation process of pharmaceuticals taking place during their degradation. Furthermore, matrix effects have been reported to influence the degradation process. For example, the photocatalytic degradation of abacavir with titania-graphene oxide was found to occur faster in ultra-pure water (20 min duration) than in leachate which lasted 3 hrs [75]. This means sufficient optimization of the degradation process on contaminated real water matrix is needed to ensure the success of antiretroviral drug removal.

To reduce costs related to the removal of antiretroviral drugs in water, it is necessary to apply re-usable procedures including the establishment of reusable photocatalysts, and adsorbents. It was noted that some of the investigated photocatalysts, and adsorbents have not been evaluated for their ability to be reused. This has been done to a limited extent where some of these materials have been found to be reusable [13,85,88,111].

The present literature shows various strategies for the removal of antiretroviral drugs in water bodies. However, the conducted studies thus far focussed largely on the elimination of these drugs in water using the laboratory scale set-up. There is currently still a lack of scale-up research to explore the removal of antiretroviral drugs in large scale which can be accommodated by the municipal WWTPs which have been found to contain large quantities of these drugs. Therefore, most of the conducted research has not determined if the removal of such contaminants in water can be performed at an industrial or large scale. In addition, the conducted research studies have not evaluated the cost-effectiveness of their approach for the removal of antiretroviral drugs in water. This is important when considering the financial position of the developing countries that already have their financial resources allocated to the fight against the spread of HIV/AIDS.

Research on plant uptake of antiretroviral drugs from contaminated environment is still limited. Selected antiretroviral drugs have already been detected in invasive species such as water hyacinth which presence an opportunity to investigate the potential to utilize these plants for phytoremediation. Other plants as well with potential to remove pharmaceuticals in water bodies can be investigated. Thus far, these plants should be investigated if they can serve as the polishing step in WWTPs where holding times can be applied for treated water to allow plant growth, while also removing pharmaceutical residues.

Some studies have reported the occurrence of metabolites of antiretroviral drugs in water bodies [112,113]. But there is still title focus on studying the potential toxicity of these metabolites towards aquatic organisms. The same is observed for their removal in contaminated water. This could be a result of still limited information on their presence in water. Therefore, future research is expected to extensively monitor the occurrence of the metabolites of antiretroviral drugs in water, the toxic effects, and removal strategies in contaminated water.

## 7. Conclusion

In this paper, a comprehensive review focussing on the occurrence, and removal of antiretroviral drugs in water is presented. The article included a detailed summary on the occurrence of antiretroviral drugs in environmental water bodies, and their potential toxic effects. Both adsorption, and degradation processes were found to be dominant initiatives employed for the removal of antiretroviral drugs in water. Although promising results have been presented in literature for the degradation of antiretroviral drugs, there is still not sufficient information on the identification of the degradation products, and evaluation of the effects for the identified products. To some extent, the removal of antiretroviral drugs has been reported to be influenced by the nature of the water sample, and its complexity. In this case, the removal of antiretroviral drugs in complex water samples such as leachates seem to be prolonged when compared to simpler water matrices. The research summary, gaps, future research opportunities identified in this review are expected to advance the research area.

## Declaration of Competing Interest

The author declares that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

## Acknowledgments

The author thanks the National Research Foundation of South Africa (Grant number: MCR240409212966) for funding his related research on environmental monitoring, and removal of antiretroviral drugs in contaminated water.

## Data availability

No data was used for the research described in the article.

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