Review

Pharmaceutical and Microplastic Pollution before and during the COVID-19 Pandemic in Surface Water, Wastewater, and Groundwater

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Abstract: Pharmaceuticals, microplastics, and oil spills are the most hazardous contaminants in aquatic environments. The COVID-19 pandemic enhanced pharmaceutical and microplastic contamination in aquatic environments. The present study aimed to investigate the prevalence of pharmaceutical and microplastic pollution on a global scale. This study assessed the results of pharmaceutical contamination in 25 countries and microplastic pollution in 13 countries. The findings show that pharmaceutical residues were detected in surface water, groundwater, and wastewater influents and effluents. In total, 43 types of pharmaceutical products were detected in 25 countries. Caffeine, acetaminophen, ibuprofen, sulfamethoxazole, and carbamazepine were the most abundant. In total, 32 types of polymers were detected in 13 countries. In the case of microplastics, polypropylene, polyethylene, polystyrene, and polyethylene terephthalate were the more abundant polymers. Particles with a size of 1–2.5 mm and 2.5–5 mm accounted for half of the microplastics present in 13 countries. This study provides new evidence of the importance of emerging pollutants in aquatic environments before and during the COVID-19 pandemic.

Keywords: pharmaceuticals; microplastics; COVID-19; water contamination; emerging pollutants

1. Introduction

The preservation of marine ecosystems has become a prominent environmental concern in the last 50 years [1], but particularly during the COVID-19 pandemic. The causes of water contamination can be classified as either natural or anthropogenic [2]; however, the effects of anthropogenic contamination on aquatic ecosystems are much more severe than those of natural contamination. Nevertheless, the impact of emerging pollutants resulting from the COVID-19 pandemic on the ecosystem is poorly understood [3,4]. Regarding the impact of lockdowns on river pollution, we must take into account the consumption of certain pharmaceuticals (PhACs) and personal care items for managing and preventing COVID-19 which spiked during the pandemic [5]. Additionally, the use of single-use plastics generated large volumes of waste, including hospital waste, post-used PPEs, confirmatory COVID-19 tests, and vaccination residues, as well as packaging in general [6].

PhACs and microplastics (MPs) are emerging pollutants that pose serious environmental risks [7–9]; thus, there is a challenge for scientists to identify approaches that will decrease the presence of pharmaceutical (PhAC) residues and microplastic (MP) particles in aquatic environments [10] given their hazardous impacts on aquatic ecosystems. However, their global release into aquatic ecosystems is far from understood. One of the primary reasons for this is that the MP and PhAC sources have been largely undocumented. Furthermore, it is difficult to determine the form of MP and PhAC in water. For example, MPs can be surface water pollutants, which are of concern, or they can be suspended in the water and be of less concern. However, the amount of MPs and PhACs in the water can be estimated using the volume of water that is polluted.

PhACs have been found in low concentrations in a variety of environmental samples, including sewage treatment plant effluents, surface water, seawater, and groundwater, in a number of countries [11,12]. Non-steroidal anti-inflammatory drugs, antibiotics, beta-blockers (-blockers), antiepileptic drugs, blood lipid-lowering agents, antidepressants, hormones, antihistamines, and X-ray contrast media are among the PhACs that are environmentally hazardous [13,14]. The consumption of PhACs increased during the COVID-19 pandemic, and a high volume of PhAC residues were released into the wastewater as well as to the sea and rivers because of hospital discharges and PhAC factories. It has been reported that the concentration of most of the PhACs used in the treatment of COVID-19 increased during the pandemic in water bodies [15]. Additionally, the PhACs from households, such as pills, capsules, and tablets, are released into wastewater. Through the wastewater, PhACs are released into natural ecosystems, such as rivers, lakes, and oceans. The PhACs from wastewater reach the environment after several transformations, from wastewater through hydrological pathways.

Plastic materials have many superior features compared to traditional materials due to their durability, malleability, low cost, versatility, and impermeability; nonetheless, their persistence in the environment has led to environmental concerns [16,17]. Plastic particles are categorized into three distinct size categories: (1) mesoplastics (500 μm–5 mm), (2) microplastics (MPs; 50–500 μm), and (3) nanoplastics (<50 μm) [10,18]. MPs are plastics with a primary diameter of less than 0.5 mm [19,20] that can be primary particulates or secondary fragments formed by weathering or degradation of primary plastics [20] and MP contamination is widespread due to the widespread use of plastics in society and industry, as well as the persistence and easy transport of sub-mm-sized primary and secondary particles [21–23]. The largest accumulation of plastic occurs in the ocean, which is estimated to have more than 500 million tons of plastic generally sourced from land-based emissions. These plastics have been accumulating in the ocean for decades, but the scale of plastic pollution in the ocean has only been realized in the last decade with the advent of oceanographic techniques to detect plastic, such as net tows and trawls. An estimated 80% of the world's population lives within 100 km of the ocean, making the ocean an important source of plastic pollution for humans, wildlife, and ecosystems. For this reason, the majority of the international scientific community's focus has thus far been on the research of plastic pollution in oceans [10]; however, the lack of information on the effects of plastic pollution on inland ecosystems is a major cause for concern [24].

This study's aim is to fill the knowledge gap about PhAC and MP pollution before and during COVID-19 in surface water, wastewater, and groundwater, as well as show the distribution of PhACs and MPs in aquatic environments, especially during the COVID-19 pandemic around the world. Moreover, the rate of PhAC residues before and after water treatment to provide a reference for the development of wastewater treatment plants (WWTPs). Based on the measured concentrations, the ecological risk of PhAC and MP pollution before and during COVID-19 in surface water, wastewater, and groundwater was assessed. The results of this work can be generalized to a large number of datasets, and the method can be applied to the classification of any dataset.

2. Distribution of Pharmaceutical Contamination in the Aquatic Environments

In recent years, PhACs and MPs have been identified in many aquatic habitats. Several types of PhACs have been detected in aquatic environments [25–57]. The most common PhACs were antibiotics (included penicillin, aminoglycoside, tetracycline, and erythromycin), cardiovascular drugs (including calcium channel blockers, angiotensin-converting enzyme inhibitors, and beta blockers), and psychiatric drugs (including antipsychotics, antidepressants, anxiolytics, and hypnotics). Consumption of antibiotics increased during the COVID-19 pandemic, and this has been a significant influence on the release of PhACs from wastewater of hospitals to aquatic environments over the last two years.

In aquatic environments, pH, temperature, time, enzymes, ionic strength, and water depth all play crucial roles in the adsorption, degradation, and transport of PhACs [10]. For instance, ionic strength and pH can influence the adsorption and transport of two antibiotics, sulfamethoxazole and ciprofloxacin [10,58]. Water content, pH, and temperature also have an effect on the decomposition of PhACs such as aspirin [10,59]. Degradation of PhACs in the summer is higher than in winter, which has an effect on the increasing spread of PhACs in aquatic environments [10]; this is because most of the PhACs transfer to surface water, and some of them go to deeper layers of water.

This study demonstrates fluctuations in PhAC and MP contamination in surface water, wastewater, and groundwater before and during the COVID-19 pandemic. In addition, the concentration of PhAC and MP contamination before and after water treatment was examined. Furthermore, this study provided information regarding PhAC and MP contamination in surface water, wastewater, and groundwater over the past 14 years. In all cases, PhACs were found, and 43 types of PhACs were detected in 25 countries. The highest concentration of PhACs from 25 countries was reported in Nigeria, with 129,000 ng/L of sulfamethoxazole and 111,000 ng/L of paracetamol in surface water, followed by 155,600 ng/L of caffeine in influent in Jordan, 140,000 ng/L of caffeine in the effluent in Iran, and 129,000 ng/L of sulfamethoxazole in surface water in Nigeria (Table S1). By contrast, the lowest concentration of PhACs across 25 countries was reported in Malaysia (4.4 ng/L of amoxicillin, 4.49 ng/L of diclofenac, and 5.01 ng/L of triclosan) in surface water, followed by effluent in Australia (5.04 ng/L of triclosan). As the results show, high concentrations of PhACs are widespread in Africa and Asia. This is due to the fact that the PhACs are widely used in these regions and that these regions have the highest population in the world. Hence, it is not surprising that the highest concentration of PhACs is located in these regions. The lowest concentration of PhACs is located in parts of Asia and Oceania. This is due to the fact that the use of the PhACs is limited in Oceania. The key sources of PhACs and their metabolites in the environment are the following: (1) the PhACs manufacturing sector, which includes industrial wastewater discharge and solid wastes containing PhACs, as well as stormwater runoff transporting powdered drugs [10,60]; (2) consumers/households, including PhACs excretion and inappropriate discharge to wastewater systems [10]; (3) hospitals, including the discharge of wastewater and solid wastes; and (4) agriculture and aquaculture including the using of hormones and other PhACs for poultry, livestock, shrimp, and fish [10] (Figure 1). Indeed, about half of the PhACs are excreted by humans and animals are released into the environment. One or more of these sources can be relevant to each case in our research. For instance, in India, the high density of the population corresponds with the high rate of consumption of PhACs. In Nigeria, high concentrations of PhACs were related to high consumption and the old technology used in WWTPs. Moreover, the COVID-19 pandemic has been the main reason for the increased global consumption of PhACs.
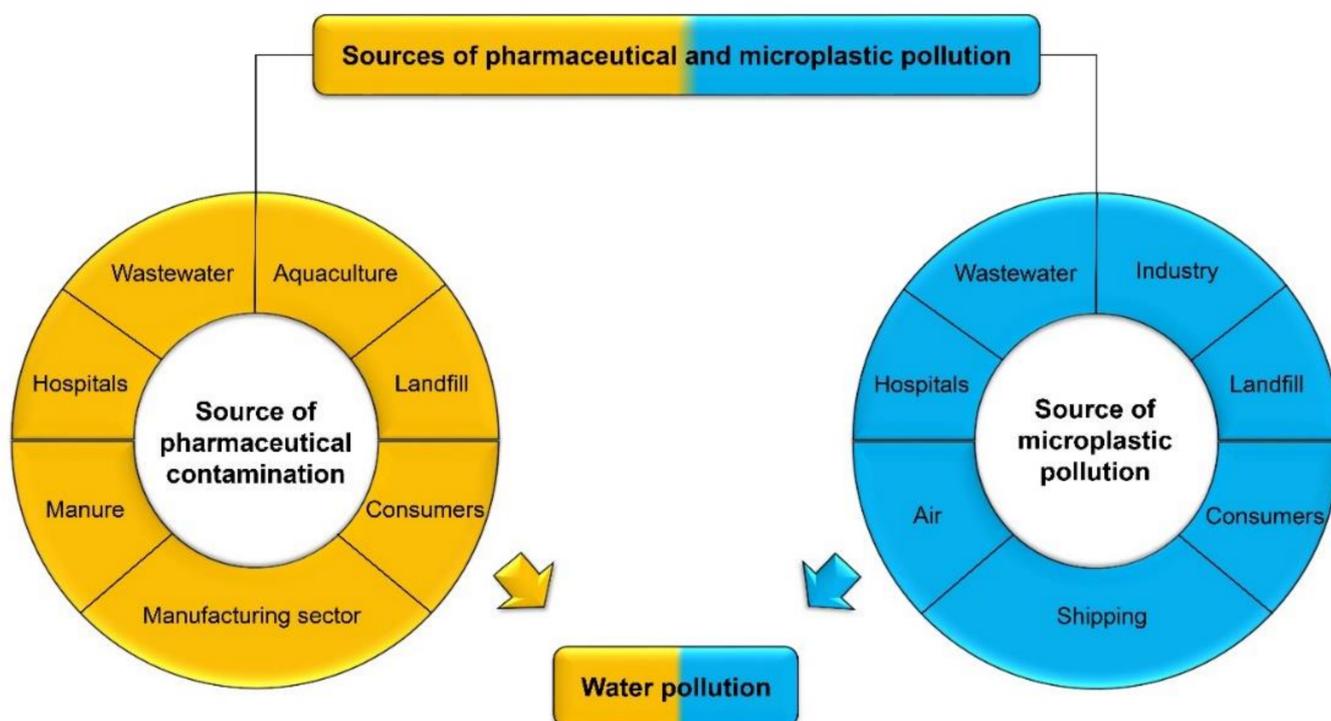
Figure 1. Sources of PhAC and MP pollution in aquatic environments.

2.1. Surface Water

The concentrations of PhAC residues in surface waters were generally low [61] but data shows that the concentration of PhAC residues in surface waters has increased in the past decade. For instance, [62] investigated the presence and distribution of 18 antibiotics in the surface water of Chaohu Lake in China. Based on the search topic of compound name and surface water, the data (Table S1) demonstrated that more than 25 of the compounds had already been extensively reported and reviewed in surface water. The concentration of PhACs in surface water and wastewater were found to be higher than that in groundwater. The highest range of PhACs was reported in surface water and wastewater. In total, 129,000 ng/L of sulfamethoxazole and 111,000 ng/L of paracetamol were detected in surface water in Nigeria (Table S1). It can be said that fifty percent of the PhACs contamination in this research was related to surface water. It was estimated that surface water pollution was largely caused by discharge from hospitals. The main issue is the lack of awareness of hospitals on the pollution of surface water. Comparing the concentration of PhACs before and during the COVID-19 pandemic in some statistics, showed that concentrations could either increase or decrease. For instance, the concentration of diclofenac in the surface waters of Ghana in 2019 was 30 ng/L, but it increased to 100.91 ng/L in 2021 [52,63] (Figure 2). Additionally, in surface water in China the concentration of venlafaxine in 2017 was 22.9 ng/L, while the concentration of venlafaxine in 2020 was 54.2 ng/L [64,65]. Conversely, the concentration of chloramphenicol in Ghana was lower in 2021 (41.36 ng/L) than in 2019 (180 ng/L) [52,63]. In Italy, high surface water concentrations of ketoprofen in 2015 (90 ng/L) were found to decrease to 5.84 ng/L in 2020 [66]. In addition, other research about China's surface water also shows reductions in PhAC concentrations. The concentration of tetracycline, sulfamerazine, doxycycline, ciprofloxacin, ofloxacin, and azithromycin in surface water in China in 2019 were 5.27 ng/L, 0.79 ng/L, 9.44 ng/L, 14.07 ng/L, 2.12 ng/L, and 0, respectively. The corresponding values in 2020 were 1.52 ng/L, 0.06 ng/L, 0, 0, 0, and 0.16 ng/L, respectively [67]. Except for azithromycin, the concentration of the other five PhACs in the surface water in China in 2020 were lower than the concentration in 2019.
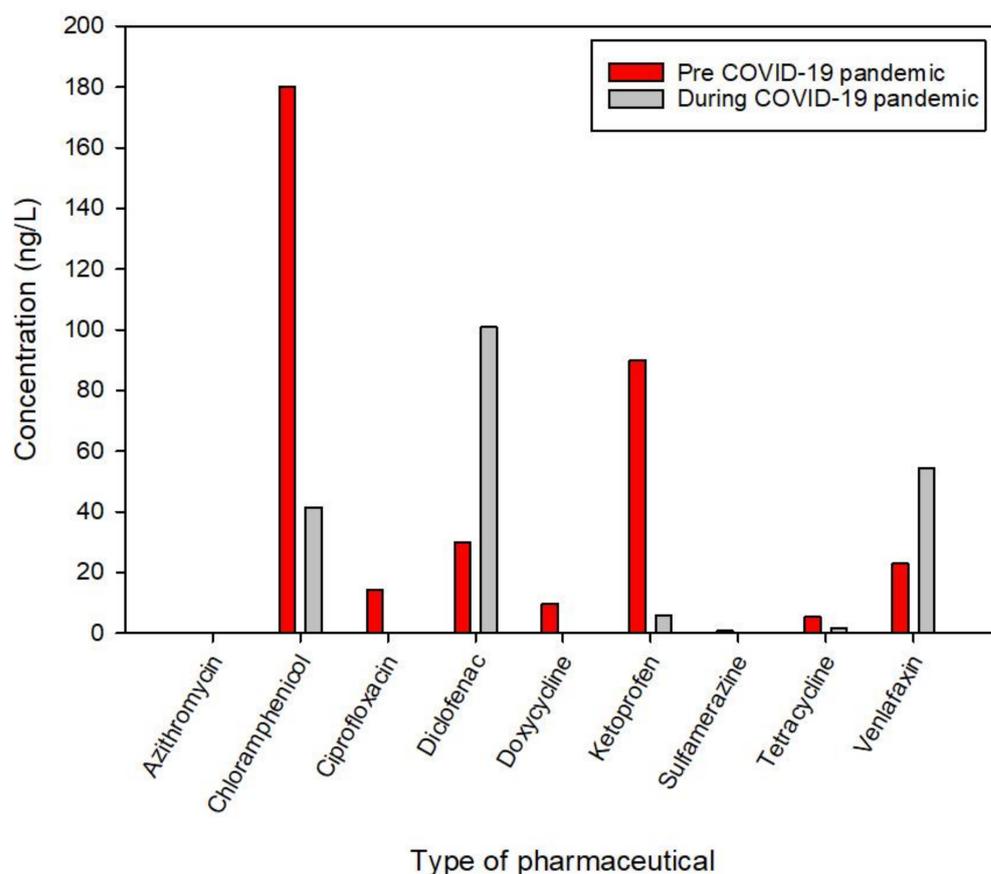
Figure 2. Concentrations of PhAC contamination in pre-COVID-19 period and during COVID-19 period in surface water.
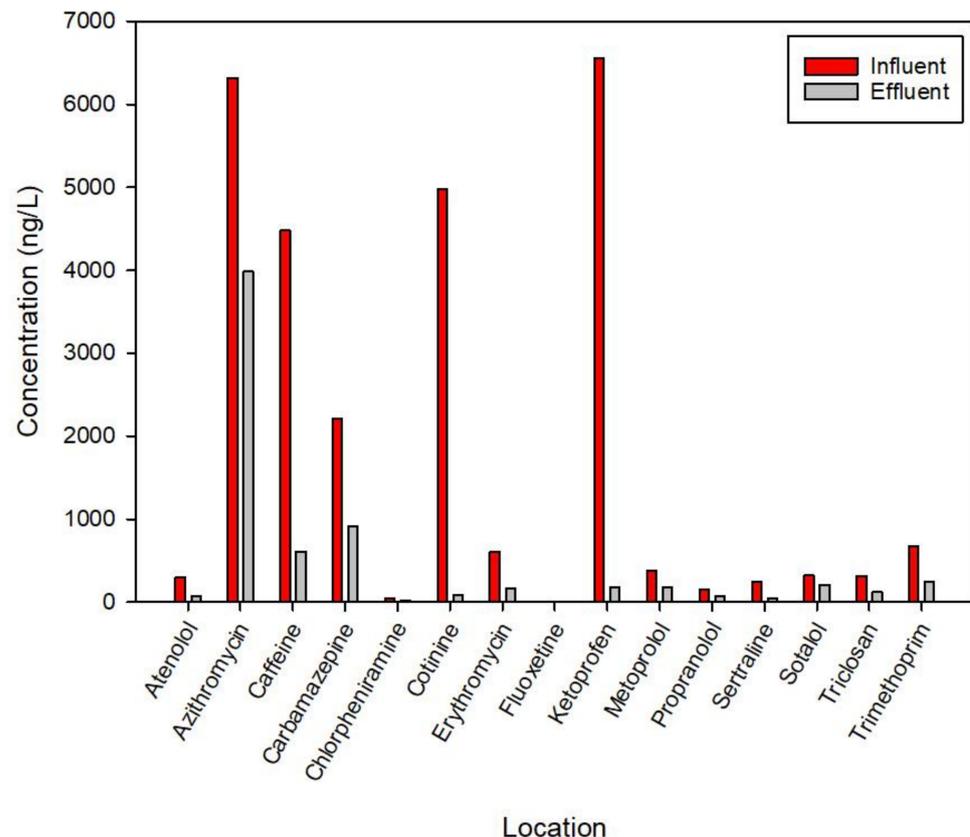
2.2. Wastewater Influent and Effluent

Anthropogenic activities, as well as irrational use of PhACs and antibiotics and their continual discharge from their manufacturing industries, have resulted in persistent and rising quantities in various wastewaters and aquatic environments [68]. Different types of pollution, especially PhAC contamination occur in influents (wastewater before treatment) and effluents (wastewater after treatment). The highest concentrations of PhACs in influents were reported in Jordan, France, and South Africa from three different continents. A total of 155,600 ng/L of caffeine, 96,700 ng/L of acetaminophen, and 62,820 ng/L of ibuprofen were detected in the surface water of Jordan, France, and South Africa, respectively (Table S2). Such surface water concentrations of some types of PhACs in wastewater were higher during the COVID-19 pandemic than before the pandemic. For instance, in China, the wastewater concentration of fluoxetine in influent and effluent before the pandemic was 2.6 ng/L and 1.4 ng/L, while during the pandemic, it was 4.25 ng/L and 1.05 ng/L, respectively [27,65].

PhAC residues were found in the effluent of these three countries, namely 86 ng/L of caffeine, 172 ng/L of acetaminophen, and 58,710 ng/L of ibuprofen which demonstrates that membrane filtration, active carbon filtration, ultraviolet radiation, and chlorination are not suitable for removing PhACs from WWTPs because even after wastewater treatment, PhACs can still be found in the effluent. In India 14,000,000 ng/L of ciprofloxacin and 2,100,000 ng/L of cetirizine were reported in effluents demonstrating that filtration was not effective at reducing the concentration of the PhACs. This illustrates that current technologies used in WWTPs are not suitable for the removal of PhACs. Particularly, in the case of South Africa, the concentration of ibuprofen after water treatment was very high (62,820 ng/L in the influent and 58,710 ng/L in the effluent) (Table S2). The common point

is that PhACs were detected in surface water and wastewater on all continents. It shows that PhACs are ubiquitous in aquatic environments.

The concentrations of PhACs in the effluent of WWTPs are presented in Figure 3. It demonstrates that ozonation is a promising method for removing contaminants from wastewater [69], as well as using biosilica for water treatment [70]. However, the efficiency of ozonation and biosilica depends on the physicochemical properties of the pollutant. However, in some cases, a filtration system could decrease the concentration of PhACs, as shown in Figure 3. For instance, the concentration of caffeine, cotinine, ketoprofen, and paraxanthine decreased significantly after water treatment.
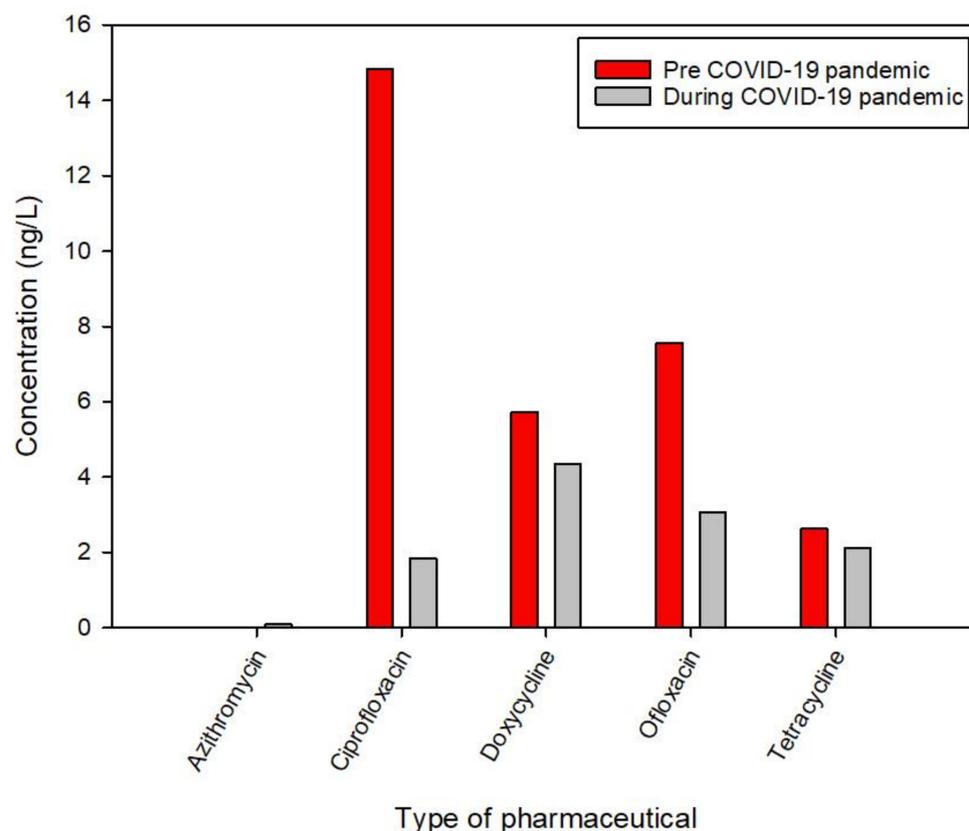

Figure 3. Concentrations of PhAC contamination in pre-COVID-19 period and during COVID-19 period in influent and effluent.

2.3. Groundwater

Groundwater is a critical global water resource that is being contaminated as a result of human activities [71]. The data highlight that the concentration of PhACs in groundwater was also high. Detecting PhACs in water in different types of water sources shows that PhACs are one of the biggest threats to human and marine organisms. It is a particularly significant that high concentrations of acetaminophen were found in the USA and Cameroon because it shows that PhACs can exist in advanced countries and developing countries. In fact, pollution is emerging everywhere in the world. The most crucial point is that the concentration of acetaminophen in the USA was much higher than in Cameroon in groundwater (1890 ng/L and 111 ng/L, respectively) [25,44] (Table S3). Conversely, in groundwater in Cameroon, the sulfamethoxazole concentration was seven times higher than in the USA. In general, groundwater has importance because it is the main supply for agriculture, and if groundwater has PhAC contamination, it can be transferred to the human food chain. The main source of PhAC contamination in groundwater is derived from wastewater from municipal and industrial WWTPs [72].

In groundwater, such as surface water and wastewater, concentrations of PhACs before and during the COVID-19 pandemic were analyzed, but the lack of data limits our understanding of changes that may have occurred. However, the concentration of tetracycline, sulfamerazine, doxycycline, ciprofloxacin, ofloxacin, and azithromycin in groundwater of China during the pandemic were 2.11 ng/L, 0.02 ng/L, 4.35 ng/L, 1.84 ng/L, 3.06 ng/L, and 0.10 ng/L, respectively and the corresponding concentrations before the pandemic were 2.63 ng/L, 0, 5.73 ng/L, 14.83 ng/L, 7.56 ng/L, and 0, respectively [66]. In most cases, there was a reduction in 2020 in comparison with 2019 concentrations (Figure 4). However, more research needs to be done in order to determine the concentrations of PhACs in groundwater before and during the COVID-19 pandemic.


Figure 4. Concentrations of PhAC contamination in pre-COVID-19 period and during COVID-19 period in groundwater.

3. Distribution of Microplastic Pollution in the Aquatic Environments

Polyethylene, polypropylene, polyethylene terephthalate, polyvinylchloride, polyester, and polystyrene are all common polymers found in aquatic environments [73–90] (Table S4). These materials are primarily used for packaging, plastic bottles, and other products. They are also used for food packaging and containers. The primary concern with these materials is their impact on the aquatic environments. They are non-biodegradable, so they accumulate in the environment and have the potential to harm aquatic organisms.

The effects of MPs on the environment are mainly related to their size, shape, charge, surface coating, agglomeration rate, density, and other properties [10,91]. The results focused on the most important factors of MPs in aquatic environments, namely size, shape, color, concentration, and kind of polymer. To compare the impact of MPs in surface water before and after the COVID-19 pandemic, it is useful to look at data from Turkey (Figure 5). Here, it was found that the concentration of MPs from 2009 to 2020 increased by more than threefold in the waters of the Black Sea [84]. Indeed, in 2009 the concentration of MPs was at its lowest level (0.331 Particles/m^3). Meanwhile, in the Black Sea from 2010 to 2020 the concentration of MPs increased. The highest concentration of

particles (0.944 Particles/m^3) was observed in 2020 [84]. In the surface water of Iran, the concentration of MPs increased during the COVID-19 pandemic (43 Particles/m^3 in 2021) by comparison with that before the pandemic (0.000061 Particles/m^3 in 2019) [79,92]. This was due to the fact that the COVID-19 pandemic led to an increase in the use of masks, gloves, and other disposable plastic.
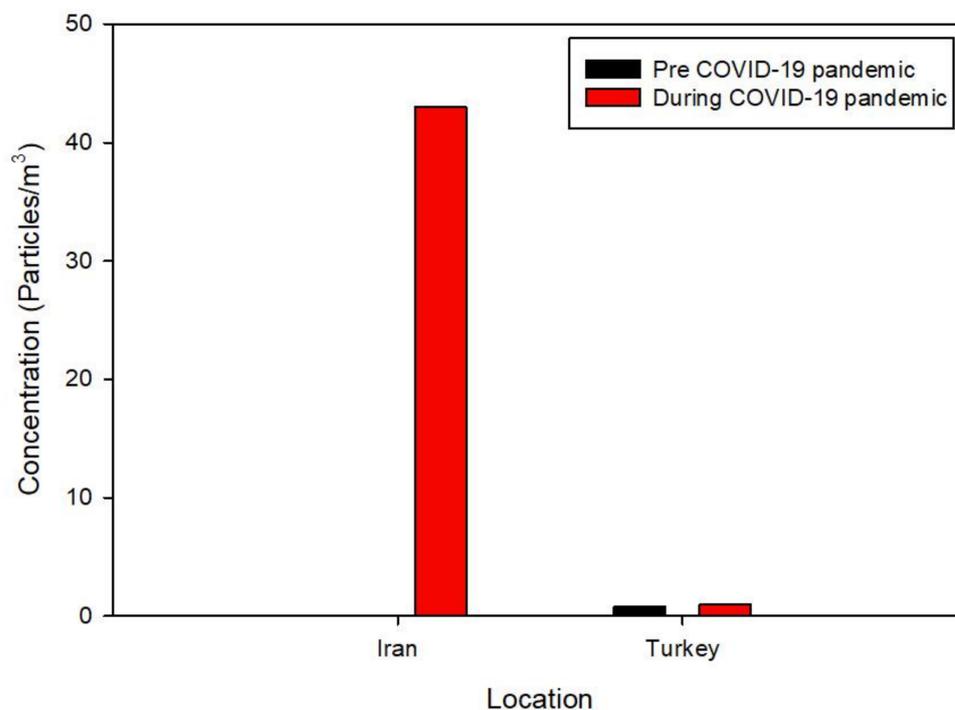
Figure 5. Concentrations of MP pollution in pre-COVID-19 period and during COVID-19 period in surface water.

In all the cases, MPs were found, and 32 types of polymers were found in 13 countries. The highest concentration of MPs across 13 countries was reported in South Korea (5242 Particles/m^3). The next highest concentration was in Portugal (1265 Particles/m^3), followed by China (967 Particles/m^3) and Tunisia (453 Particles/m^3) (Table S2). In contrast, the lowest concentration of MPs across 12 countries was reported in India (0.000004 Particles/m^3), followed by Iran (0.000061 Particles/m^3) and Norway (0.00084 Particles/m^3). PP, PE, PS, and PETE were the main polymers were found in 13 countries in surface waters. The results show high concentrations of MPs in locations in Asia, Europe, and Africa. These could be caused by many factors. For example, the high concentration of MPs in Asia and Africa could be caused by population density. The high concentration of MPs in Europe could be caused by high levels of industrialization and plastic use. The primary contributors to MP particles in aquatic ecosystems include the following: (1) direct disposal, intentional or unintentional (e.g., fishing gears, cargo ships, granules used for the production of larger products); (2) mechanical fragmentation of larger plastic debris already present in the environment [18]; (3) sewage and water treatments plants [93]; and (4) inefficient urban waste separation and disposal [10] (Figure 1). In the case of MPs, sources have a greater variety than PhACs because, nowadays, plastic is used everywhere, but the COVID-19 pandemic also had an impact on the increasing rate of plastic waste generation due to its use in everyday life. As a result of the COVID-19 pandemic, the use of personal protective equipment has shifted from specific use in confined settings (e.g., hospitals) to general use within the population, which contributes to increased MP consumption [94].

The data in Tables S1 and S4 indicate that surface waters are becoming increasingly polluted and that sea currents, waves, and major wind patterns are regarded as the main factors influencing the spread of MP particles in marine environments [88,95,96]. Addi-

tionally, ultraviolet radiation plays an important role in the degradation and spread of MP particles. However, the degradation and spread of MP particles are highly dependent on location and environmental conditions [10]. Indeed, the most important MP degradation pathways and transport processes are (1) physical degradation, (2) photodegradation, (3) chemical degradation, (4) biodegradation by organisms [97], and (5) wind and waves.

It is also important to consider the source of particles. The different colors, shapes, and sizes of MP particles are shown in Table S2, and demonstrate a variety of sources of MP particles. For example, the main source of MP particles in Urmia Lake in Northwest Iran is related to ship repair factories because most of the polymers are derived from ship bodies [79]. For example, blue and transparent MP particles are related to ferries that are used for transporting vehicles and people across Urmia Lake.

Between 700–1000 PhACs for treating COVID-19 were listed in the Drug Bank as of October 2021 [98], which is a very significant number. This amount of PhACs shows that there is a high potential for toxicity in aquatic systems because PhACs compounds remain biologically active in aquatic systems [10]. In particular the rate of consumption of PhACs in countries that lack access to vaccines, such as in developing countries, has increased. These circumstances indicate that the environmental risk of PhACs is increasingly threating water resources. The results of the analysis of the data from the monitoring of the water in the pre-pandemic and during pandemic showed that the concentration of MPs in the surface water increased in the same way as PhACs. However, in general, the quantity MPs represents a bigger threat to water quality. A key point here is that when PhAC residues are adsorbed by MP particles, they can exhibit increased toxicity within the water body [10].

4. Pharmaceutical and Microplastic Pollution before and during the COVID-19 Pandemic

4.1. Concentrations of Pharmaceutical Contamination before and during the COVID-19 Pandemic

Concentrations of PhAC contamination during the COVID-19 pandemic have increased in comparison with before the pandemic in aquatic environments (Figure 6). For instance, the concentration of diclofenac in Ghana's surface water in 2019 was 30 ng/L, but in 2021 during the COVID-19 pandemic, the concentration increased to 100.91 ng/L [52,55]. On the other hand, 90 ng/L of ketoprofen were found in the influent of water in Italy in 2013; however, the ketoprofen rate in 2020 was 5.84 ng/L which shows a remarkable decrease [43,66]. In China surface water data before and during the pandemic showed differences. Indeed, the number of PhACs before the pandemic was higher than that during the pandemic, but the PhACs concentrations before the pandemic were lower than during the pandemic. The concentrations of tetracycline (5.27 ng/L in 2019 and 1.52 ng/L in 2020), sulfamerazine (0.79 ng/L in 2019 and 0.06 ng/L in 2020), doxycycline (9.44 ng/L in 2019 and 0 in 2020), ciprofloxacin (14.0 ng/L in 2019 and 0 in 2020), and ofloxacin (2.12 ng/L in 2019 and 0 in 2020) before the pandemic were higher than during the pandemic. Conversely, the concentration of azithromycin in 2020 was 0.16, which was higher than 0 in 2019 [67]. Meanwhile, the concentrations of fluoxetine in influent and effluent before the pandemic were 2.6 ng/L and 1.4 ng/L, and during the pandemic, they were 4.25 ng/L and 1.05 ng/L, respectively [65,99]. Additionally, in groundwater in China, results in 2019 and 2020 showed differences before and during the pandemic. The concentration of tetracycline (2.63 ng/L in 2019 and 2.11 ng/L in 2020), doxycycline (5.73 ng/L in 2019 and 4.35 ng/L in 2020), ciprofloxacin (14.83 ng/L in 2019 and 1.84 ng/L in 2020), and ofloxacin (7.56 ng/L in 2019 and 3.06 ng/L in 2020) before the pandemic were higher than during the pandemic. Conversely, the concentration of sulfamethazine and azithromycin in 2020 were 0.02 ng/L and 0.10 ng/L, which shows a gain in comparison with 2019 [67].
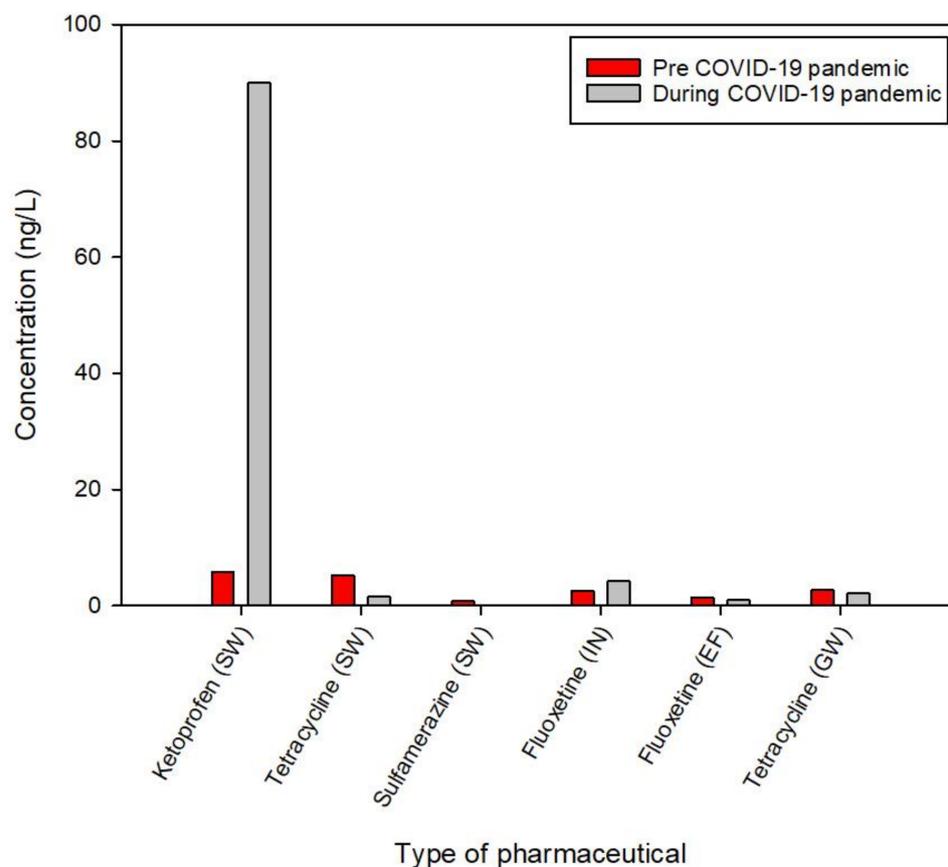
Figure 6.　　Concentrations of PhAC contamination in pre-COVID-19 period and during COVID-19 period.

4.2. Concentrations of MP Pollution before and during the COVID-19 Pandemic

Concentrations of MP pollution before and during the COVID-19 pandemic show that the quantity of MPs increased during the pandemic compared with the pre-COVID-19 period in surface water (Figure 5). For example, the concentration of MPs in the surface water in Turkey and Iran during the COVID-19 pandemic was higher than that pre-COVID-19 period. The concentration of MPs in Iran's surface water in 2016 was 0.000042 Particles/m^3, in 2019, 0.000061 Particles/m^3, but in 2021, during the COVID-19 pandemic, this rate increased to 0.246 Particles/m^3 [79,100]. Moreover, [92] detected 43 Particles/m^3 of MPs in Iran's surface water. In fact, the concentration of MPs in the surface water of Iran is increasing due to the presence of different types of plastics. Additionally, 0.750 Particles/m^3 of MPs were found in the surface water of Turkey in 2019; however, the MPs concentration in 2020 was 0.944 Particles/m^3 which shows an increase [84] (Figure 5).

5. Water Treatment Systems

For the removal of emerging contaminants from wastewater, natural water, and drinking water, several biological (for example, activated sludge, microalgae, membrane bioreactors) and chemical (for instance, chlorination, Fenton process, ozonation, photolysis [101,102], membrane filtration, active carbon filtration, advanced oxidation processes) procedures are being studied, but ozonation can enable the removal of a wide range of contaminants as well as water disinfection [102,103]. Ozonation can also decrease the concentration of PhAC residues in effluents [104]. Biosilica is also used for water treatment [70,105], but there is a lack of research on decreasing the concentration of PhAC residues in effluent by biosilica. Another system for PhAC residue removal in water is the combination of membrane filtration and advanced oxidation processes. There are several studies that have used membrane filtration for PhAC residue removal, and the efficacy

of advanced oxidation processes for this purpose has been demonstrated; however, the combination of these two technologies can complete the removal of PhAC residues from water in a more efficient way [106]. Overall, (1) ozonation and (2) the combination of membrane filtration with advanced oxidation processes are the most efficient and effective ways to remove heavy PhAC residues from water.

Filtration MPs by membrane filtration, active carbon filtration, ultraviolet radiation, chlorination, ozonation, and advanced oxidation processes alone is not suitable because they are not able to remove all of the MP residues from the water. For instance, ozonation changes the structures of polymers [107]. However, there is no improvement in microparticle removal due to ozonation [107]. In addition, studies based on membrane aging mechanisms and material attributes have demonstrated that membrane filtering systems could release MPs into drinking water distribution networks [108]. One technique that is very effective in MP removal is magnetic polyoxometalate-supported ionic liquid phases. Using magnetic polyoxometalate-supported ionic liquid phases, we can achieve remarkable removal efficiencies, and initial insights into a new technique of MPs removal via surface-binding of magnetic particles have been reported [109–111].

6. Discussion

This study has investigated the potential environmental risks of PhACs and MPs in pre-COVID-19 and during the COVID-19 pandemic in aquatic environments. Types of COVID-19 PhACs which are commonly used, belong to a wide range of categories, including (1) antibiotics, (2) analgesics, (3) nonsteroidal anti-inflammatory drugs, and (4) antiretrovirals. Commonly used PhACs for COVID-19 include ibuprofen, azithromycin, and paracetamol. During the COVID-19 pandemic consumption of other types of PhACs also increased. These PhACs have entered extensively into surface water, wastewater, and groundwater from various environmental sources. Moreover, the results of this study have shown that the MPs concentration in the surface water was significantly higher in the COVID-19 period than in the pre-COVID period because of the high consumption rates of masks and gloves. Further research is needed to estimate and measure PhAC and MP pollution during the COVID-19 pandemic in aquatic environments. The majority of the studies used to inform our estimate of PhAC and MP pollution during COVID-19 pandemic in aquatic environments were carried out in Africa, Asia, Europe, and Oceania. The results of several types of PhACs were detected in aquatic environments but caffeine, acetaminophen, ibuprofen, sulfamethoxazole, and carbamazepine were most prevalent in comparison with other types of PhACs because acetaminophen, ibuprofen, sulfamethoxazole, and carbamazepine had high consumption during the COVID-19 lockdown. Another key point of this research is that the filtration systems in WWTPs are not suitable for removing PhAC residues. Additionally, different types of polymers with different shapes, sizes, and colors were found in aquatic environments, but PP, PE, PS, and PETE were more abundant and were related to hospital waste, post-used PPEs, confirmatory COVID-19 tests, and vaccination residues, as well as packaging in general. Particles in the 1–2.5 mm and 2.5–5 mm size range accounted for half of the MPs present in 13 countries. In addition to the physical impacts of contaminated water on humans, we are also starting to see the impact on aquatic life. The contamination of an ecosystem can have a cascade of effects on other species in the ecosystem, including the removal of oxygen, the spread of disease, and the death of fish and other aquatic life. Therefore, growing awareness of emerging pollutants in aquatic environments is a critical priority for decreasing the impact of PhAC and MP pollution. This study provides significant information regarding the concentration of PhAC and MP in various regions of the world over the past 14 years, as well as the rate of PhAC residues before and after water treatment. Future researchers may find these data relevant for evaluating the rate of PhAC and MP contamination before and after the COVID-19 pandemic. Future research should address the technology of removal systems in WWTPs and the health risks of emerging pollution in aquatic environments.

Abbreviations

AR	Acrylate
ABR	Acrylonitrile butadiene rubber
ABS	Acrylonitrile butadiene styrene
AL	Alkyde
AN	Anthropogenic natural
ATR-FTIR	Attenuated total reflection-Fourier transform infrared
CA	Cellulose acetate
CP	Cellophane
COP	Copolymer
DP	Dibutyl phthalate
EA	Ethylenevinyl acetate
ELISA	Enzyme linked immunosorbent assay
ESI-MS/MS	Electrospray-tandem mass spectrometry
EVA	Ethylene vinyl acetate
FTIR	Fourier transform infrared
GC-MS	Gas chromatography–mass spectrometry
HFFR	Halogen-free flame retardant
HDPE	High density polyethylene
HPLC	High-performance liquid chromatography
HESI	Heated Electrospray
LC-MS/MS	Liquid chromatography with tandem mass spectrometry
LC-HRMS	Liquid chromatography with high resolution mass spectrometer
LDPE	Low-density polyethylene
MP	Microplastic
MPs	Microplastics
MS/MS	Tandem mass spectrometry
NI	Nitrile
NL	Nylon
PA	Polyamide
PAS	Poly(acrylate-styrene)
PAN	Polyacrylonitrile

PBPE	Poly(butyl methacrylate)-poly(ethylene glycol)
PC	Acrylic
PE	Polyethylene
PEA	Poly(ethylacrylate)
PES	Polyester
PETE	Polyethylene terephthalate
PEVA	Poly(ethylene-vinyl acetate)
PhAC	Pharmaceutical
PhACs	Pharmaceuticals
PMMA	Polymethylmethacrylate
PP	Polypropylene
PS	Polystyrene
PTFE	Polytetrafluoroethylene
PVA	Polyvinyl acetate
PUR	Polyurethane
PVC	Polyvinylchloride
Py-GC-MS	Pyrolysis–gas chromatography–mass spectrometry
RA	Rayon
SBR	Styrene butadiene rubber
SEM	Scanning electron microscopy
SI	Silicone
TGA	Thermal gravimetric analysis
TOF	Time-of-flight mass
TSQ	Triple quadrupole
UHPLC	Ultra-high performance liquid chromatography
WWTPs	Wastewater treatment plants

References

1. Pashaei, R.; Gholizadeh, M.; Iran, K.J.; Hanifi, A. The Effects of Oil Spills on Ecosystem at the Persian Gulf. *Int. J. Rev. Life Sci.* **2015**, *5*, 82–89. [CrossRef]
2. Abbasi, S.; Ashayeri, S.Y.; Jafarzadeh, N.; Sheikh Fakhradini, S.; Alirezazadeh, M.; Ebrahimi, P.; Peely, A.B.; Rezaei, N.; Mokhtarzadeh, Z.; Naraki, H.; et al. Hydrological and hydrogeological characteristics and environmental assessment of Hashilan Wetland, a national heritage in NW Iran. *Ecohydrol. Hydrobiol.* **2022**, *22*, 141–154. [CrossRef]
3. Bandala, E.R.; Kruger, B.R.; Cesarino, I.; Leao, A.L.; Wijesiri, B.; Goonetilleke, A. Impacts of COVID-19 pandemic on the wastewater pathway into surface water: A review. *Sci. Total Environ.* **2021**, *774*, 145586. [CrossRef] [PubMed]
4. Zambrano-Monserrate, M.A.; Ruano, M.A.; Sanchez-Alcalde, L. Indirect effects of COVID-19 on the environment. *Sci. Total Environ.* **2020**, *728*, 138813. [CrossRef] [PubMed]
5. Chen, X.; Lei, L.; Liu, S.; Han, J.; Li, R.; Men, J.; Li, L.; Wei, L.; Sheng, Y.; Yang, L.; et al. Occurrence and risk assessment of pharmaceuticals and personal care products (PPCPs) against COVID-19 in lakes and WWTP-river-estuary system in Wuhan, China. *Sci. Total Environ.* **2021**, *792*, 148352. [CrossRef]
6. De Sousa, F.D.B. Plastic and its consequences during the COVID-19 pandemic. *Environ. Sci. Pollut. Res.* **2021**, *28*, 46067–46078. [CrossRef]
7. Ebrahimi, P.; Abbasi, S.; Pashaei, R.; Bogusz, A.; Oleszczuk, P. Investigating impact of physicochemical properties of microplastics on human health: A short bibliometric analysis and review. *Chemosphere* **2022**, *289*, 133146. [CrossRef]
8. Pashaei, R.; Zahedipour-Sheshglani, P.; Dzingelevičienė, R.; Abbasi, S.; Rees, R.M. Effects of pharmaceuticals on the nitrogen cycle in water and soil: A review. *Environ. Monit. Assess.* **2022**, *194*, 105. [CrossRef]
9. Pashaei, R.; Dzingelevičienė, R.; Abbasi, S.; Szultka-Młyńska, M.; Buszewski, B. Determination of 15 human pharmaceutical residues in fish and shrimp tissues by high-performance liquid chromatography-tandem mass spectrometry. *Environ. Monit. Assess.* **2022**, *194*, 325. [CrossRef]
10. Pashaei, R.; Dzingelevičienė, R.; Abbasi, S.; Szultka-Młyńska, M.; Buszewski, B. Determination of the pharmaceuticals–nano/microplastics in aquatic systems by analytical and instrumental methods. *Environ. Monit. Assess.* **2022**, *194*, 93. [CrossRef]
11. Nikolaou, A.; Meric, S.; Fatta, D. Occurrence patterns of pharmaceuticals in water and wastewater environments. *Anal. Bioanal. Chem.* **2007**, *387*, 1225–1234. [CrossRef] [PubMed]
12. Halling-Sørensen, B.N.N.S.; Nielsen, S.N.; Lanzky, P.F.; Ingerslev, F.; Lützhøft, H.H.; Jørgensen, S.E. Occurrence, fate and effects of pharmaceutical substances in the environment-A review. *Chemosphere* **1998**, *36*, 357–393. [CrossRef]
13. Khetan, S.K.; Collins, T.J. Human pharmaceuticals in the aquatic environment: A challenge to green chemisty. *Chem. Rev.* **2007**, *107*, 2319–2364. [CrossRef] [PubMed]

14. Kanakaraju, D.; Glass, B.D.; Oelgemöller, M. Advanced oxidation process-mediated removal of pharmaceuticals from water: A review. *J. Environ. Manag.* **2018**, *219*, 189–207. [CrossRef] [PubMed]

15. Morales-Paredes, C.A.; Rodríguez-Díaz, J.M.; Boluda-Botella, N. Pharmaceutical compounds used in the COVID-19 pandemic: A review of their presence in water and treatment techniques for their elimination. *Sci. Total Environ.* **2021**, *2021*, 152691. [CrossRef]

16. Galgani, L.; Loiselle, S.A. Plastic pollution impacts on marine carbon biogeochemistry. *Environ. Pollut.* **2021**, *268*, 115598. [CrossRef]

17. Hale, R.C.; Seeley, M.E.; La Guardia, M.J.; Mai, L.; Zeng, E.Y. A Global Perspective on Microplastics. *J. Geophys. Rese. Oceans* **2020**, *125*, e2018JC014719. [CrossRef]

18. Andrady, A.L. Microplastics in the marine environment. *Mar. Pollut. Bull.* **2011**, *62*, 1596–1605. [CrossRef]

19. Arthur, C.; Baker, J.; Bamford, H. *Proceedings of the International Research Workshop on the Occurrence, Effects, and Fate of Microplastic Marine Debris*; University of Washington: Tacoma, WA, USA, 2009.

20. Abbasi, S.; Keshavarzi, B.; Moore, F.; Turner, A.; Kelly, F.J.; Dominguez, A.O.; Jaafarzadeh, N. Distribution and potential health impacts of microplastics and microrubbers in air and street dusts from Asaluyeh County, Iran. *Environ. Pollut.* **2019**, *244*, 153–164. [CrossRef]

21. Abbasi, S.; Turner, A. Human exposure to microplastics: A study in Iran. *J. Hazard. Mater.* **2021**, *403*, 123799. [CrossRef]

22. Rezaei, M.; Riksen, M.J.P.M.; Sirjani, E.; Sameni, A.; Geissen, V. Wind erosion as a driver for transport of light density microplastics. *Sci. Total Environ.* **2019**, *669*, 273–281. [CrossRef] [PubMed]

23. Waldschläger, K.; Lechthaler, S.; Stauch, G.; Schüttrumpf, H. The way of microplastic through the environment—Application of the source-pathway-receptor model (review). *Sci. Total Environ.* **2020**, *713*, 136584. [CrossRef] [PubMed]

24. Wagner, M.; Holzschuh, S.; Traeger, A.; Fahr, A.; Schubert, U.S. Asymmetric flow field-flow fractionation in the field of nanomedicine. *Anal. Chem.* **2014**, *86*, 5201–5210. [CrossRef] [PubMed]

25. Fram, M.S.; Belitz, K. Occurrence and concentrations of pharmaceutical compounds in groundwater used for public drinking-water supply in California. *Sc. Total Environ.* **2011**, *409*, 3409–3417. [CrossRef]

26. Matongo, S.; Birungi, G.; Moodley, B.; Ndungu, P. Pharmaceutical residues in water and sediment of Msunduzi River, KwaZulu-Natal, South Africa. *Chemosphere* **2015**, *134*, 133–140. [CrossRef]

27. Wu, C.; Witter, J.D.; Spongberg, A.L.; Czajkowski, K.P. Occurrence of selected pharmaceuticals in an agricultural landscape, western Lake Erie basin. *Water Res.* **2009**, *43*, 3407–3416. [CrossRef]

28. Kot-Wasik, A.; Jakimska, A.; Śliwka-Kaszyńska, M. Occurrence and seasonal variations of 25 pharmaceutical residues in wastewater and drinking water treatment plants. *Environ. Monitor. Assess.* **2016**, *2016*, 188. [CrossRef]

29. Fick, J.; Söderström, H.; Lindberg, R.H.; Phan, C.; Tysklind, M.; Larsson, D.G.J. Contamination of surface, ground, and drinking water from pharmaceutical production. *Environ. Toxicol. Chem.* **2009**, *28*, 2522–2527. [CrossRef]

30. Praveena, S.M.; Shaifuddin, S.N.M.; Sukiman, S.; Nasir, F.A.M.; Hanafi, Z.; Kamarudin, N.; Ismail, T.H.T.; Aris, A.Z. Pharmaceuticals residues in selected tropical surface water bodies from Selangor (Malaysia): Occurrence and potential risk assessments. *Sci. Total Environ.* **2018**, *642*, 230–240. [CrossRef]

31. Mokh, S.; El Khatib, M.; Koubar, M.; Daher, Z.; Al Iskandarani, M. Innovative SPE-LC-MS/MS technique for the assessment of 63 pharmaceuticals and the detection of antibiotic-resistant-bacteria: A case study natural water sources in Lebanon. *Sci. Total Environ.* **2017**, *609*, 830–841. [CrossRef]

32. Dehkordi, S.K.; Paknejad, H.; Blaha, L.; Svecova, H.; Grabic, R.; Simek, Z.; Otoupalikova, A.; Bittner, M. Instrumental and bioanalytical assessment of pharmaceuticals and hormone-like compounds in a major drinking water source—wastewater receiving Zayandeh Rood river, Iran. *Environ. Sci. Pollut. Res.* **2022**, *29*, 9023–9037. [CrossRef] [PubMed]

33. Nantaba, F.; Wasswa, J.; Kylin, H.; Palm, W.U.; Bouwman, H.; Kümmerer, K. Occurrence, distribution, and ecotoxicological risk assessment of selected pharmaceutical compounds in water from Lake Victoria, Uganda. *Chemosphere* **2020**, *239*, 124642. [CrossRef] [PubMed]

34. Muriuki, C.W.; Home, P.G.; Raude, J.M.; Ngumba, E.K.; Munala, G.K.; Kairigo, P.K.; Gachanja, A.N.; Tuhkanen, T.A. Occurrence, distribution, and risk assessment of pharmerciuticals in wastewater and open surface drains of peri-urban areas: Case study of Juja town, Kenya. *Environ. Pollut.* **2020**, *267*, 115503. [CrossRef] [PubMed]

35. Ogunbanwo, O.M.; Kay, P.; Boxall, A.B.; Wilkinson, J.; Sinclair, C.J.; Shabi, R.A.; Fasasi, A.E.; Lewis, G.A.; Amoda, O.A.; Brown, L.E. High Concentrations of Pharmaceuticals in a Nigerian River Catchment. *Environ. Toxicol. Chem.* **2020**, *41*, 551–558. [CrossRef] [PubMed]

36. Hossain, A.; Nakamichi, S.; Habibullah-Al-Mamun, M.; Tani, K.; Masunaga, S.; Matsuda, H. Occurrence and ecological risk of pharmaceuticals in river surface water of Bangladesh. *Environ. Res.* **2018**, *165*, 258–266. [CrossRef]

37. Botero-Coy, A.M.; Martínez-Pachón, D.; Boix, C.; Rincón, R.J.; Castillo, N.; Arias-Marín, L.P.; Manrique-Losada, L.; Torres-Palma, R.; Moncayo-Lasso, A.; Hernández, F. An investigation into the occurrence and removal of pharmaceuticals in Colombian wastewater. *Sci. Total Environ.* **2018**, *642*, 842–853. [CrossRef]

38. González-Alonso, S.; Merino, L.M.; Esteban, S.; López de Alda, M.; Barceló, D.; Durán, J.J.; López-Martínez, J.; Aceña, J.; Pérez, S.; Mastroianni, N.; et al. Occurrence of pharmaceutical, recreational and psychotropic drug residues in surface water on the northern Antarctic Peninsula region. *Environ. Pollut.* **2017**, *229*, 241–254. [CrossRef]

39. Ashfaq, M.; Li, Y.; Rehman, M.S.U.; Zubair, M.; Mustafa, G.; Nazar, M.F.; Yu, C.P.; Sun, Q. Occurrence, spatial variation and risk assessment of pharmaceuticals and personal care products in urban wastewater, canal surface water, and their sediments: A case study of Lahore, Pakistan. *Sci. Total Environ.* **2019**, *688*, 653–663. [CrossRef]

40. Guruge, K.S.; Goswami, P.; Tanoue, R.; Nomiyama, K.; Wijesekara, R.G.S.; Dharmaratne, T.S. First nationwide investigation and environmental risk assessment of 72 pharmaceuticals and personal care products from Sri Lankan surface waterways. *Sci. Total Environ.* **2019**, *690*, 683–695. [CrossRef]

41. Letsinger, S.; Kay, P.; Rodríguez-Mozaz, S.; Villagrassa, M.; Barceló, D.; Rotchell, J.M. Spatial and temporal occurrence of pharmaceuticals in UK estuaries. *Sci. Total Environ.* **2019**, *678*, 74–84. [CrossRef]

42. Reis-Santos, P.; Pais, M.; Duarte, B.; Caçador, I.; Freitas, A.; Vila Pouca, A.S.; Barbosa, J.; Leston, S.; Rosa, J.; Ramos, F.; et al. Screening of human and veterinary pharmaceuticals in estuarine waters: A baseline assessment for the Tejo estuary. *Marine Pollut. Bull.* **2018**, *135*, 1079–1084. [CrossRef] [PubMed]

43. Papagiannaki, D.; Morgillo, S.; Bocina, G.; Calza, P.; Binetti, R. Occurrence and human health risk assessment of pharmaceuticals and hormones in drinking water sources in the metropolitan area of turin in Italy. *Toxics* **2021**, *9*, 88. [CrossRef] [PubMed]

44. Branchet, P.; Ariza Castro, N.; Fenet, H.; Gomez, E.; Courant, F.; Sebag, D.; Gardon, J.; Jourdan, C.; Ngounou Ngatcha, B.; Kengne, I.; et al. Anthropic impacts on Sub-Saharan urban water resources through their pharmaceutical contamination (Yaoundé Center Region, Cameroon). *Sci. Total Environ.* **2019**, *660*, 886–898. [CrossRef] [PubMed]

45. Stasinakis, A.S.; Mermigka, S.; Samaras, V.G.; Farmaki, E.; Thomaidis, N.S. Occurrence of endocrine disrupters and selected pharmaceuticals in Aisonas River (Greece) and environmental risk assessment using hazard indexes. *Environ. Sci. Pollut. Res.* **2012**, *19*, 1574–1583. [CrossRef] [PubMed]

46. Daneshvar, A.; Svanfelt, J.; Kronberg, L.; Prévost, M.; Weyhenmeyer, G.A. Seasonal variations in the occurrence and fate of basic and neutral pharmaceuticals in a Swedish river–lake system. *Chemosphere* **2010**, *80*, 301–309. [CrossRef]

47. Komori, K.; Suzuki, Y.; Minamiyama, M.; Harada, A. Occurrence of selected pharmaceuticals in river water in Japan and assessment of their environmental risk. *Environ. Monitor. Assess.* **2013**, *185*, 4529–4536. [CrossRef]

48. Na, T.W.; Kang, T.W.; Lee, K.H.; Hwang, S.H.; Jung, H.J.; Kim, K. Distribution and ecological risk of pharmaceuticals in surface water of the Yeongsan river, Republic of Korea. *Ecotoxicol. Environ. Safety* **2019**, *181*, 180–186. [CrossRef]

49. Zhou, H.; Ying, T.; Wang, X.; Liu, J. Occurrence and preliminarily environmental risk assessment of selected pharmaceuticals in the urban rivers, China. *Sci. Rep.* **2016**, *6*, 16–18. [CrossRef]

50. Al-Mashaqbeh, O.; Alsafadi, D.; Dalahmeh, S.; Bartelt-Hunt, S.; Snow, D. Removal of selected pharmaceuticals and personal care products in wastewater treatment plant in Jordan. *Water* **2019**, *11*, 2004. [CrossRef]

51. Thiebault, T.; Boussafir, M.; Le Milbeau, C. Occurrence and removal efficiency of pharmaceuticals in an urban wastewater treatment plant: Mass balance, fate and consumption assessment. *J. Environ. Chem. Eng.* **2017**, *5*, 2894–2902. [CrossRef]

52. Asare, E.A. Status of pharmaceuticals in the Korle Lagoon and their toxicity to non-target organisms. *Ecotoxicology* **2022**, *31*, 299–311. [CrossRef] [PubMed]

53. Kermia, A.E.B.; Fouial-Djebbar, D.; Trari, M. Occurrence, fate and removal efficiencies of pharmaceuticals in wastewater treatment plants (WWTPs) discharging in the coastal environment of Algiers. *Comptes Rendus Chim.* **2016**, *19*, 963–970. [CrossRef]

54. Phonsiri, V.; Choi, S.; Nguyen, C.; Tsai, Y.L.; Coss, R.; Kurwadkar, S. Monitoring occurrence and removal of selected pharmaceuticals in two different wastewater treatment plants. *SN Appl. Sci.* **2019**, *1*, 789. [CrossRef]

55. Azanu, D.; Adu-Poku, D.; Saah, S.A.; Appaw, W.O. Prevalence of Pharmaceuticals in Surface Water Samples in Ghana. *J. Chem.* **2021**, *2021*, 477. [CrossRef]

56. Abdallah, M.A.E.; Nguyen, K.H.; Ebele, A.J.; Atia, N.N.; Ali, H.R.H.; Harrad, S. A single run, rapid polarity switching method for determination of 30 pharmaceuticals and personal care products in waste water using Q-Exactive Orbitrap high resolution accurate mass spectrometry. *J. Chromatogr. A* **2019**, *1588*, 68–76. [CrossRef] [PubMed]

57. Mhuka, V.; Dube, S.; Nindi, M.M. Occurrence of pharmaceutical and personal care products (PPCPs) in wastewater and receiving waters in South Africa using LC-OrbitrapTM MS. *Emerg. Contam.* **2020**, *6*, 250–258. [CrossRef]

58. Chen, H.; Gao, B.; Li, H.; Ma, L.Q. Effects of pH and ionic strength on sulfamethoxazole and ciprofloxacin transport in saturated porous media. *J. Contam. Hydrol.* **2011**, *126*, 29–36. [CrossRef]

59. Kornblum, S.S.; Zoglio, M.A. Pharmaceutical heterogeneous systems I. Hydrolysis of aspirin in combination with tablet lubricants in an aqueous suspension. *J. Pharm. Sci.* **1967**, *56*, 1569–1575. [CrossRef]

60. Gadipelly, C.; Pérez-González, A.; Yadav, G.D.; Ortiz, I.; Ibáñez, R.; Rathod, V.K.; Marathe, K.V. Pharmaceutical industry wastewater: Review of the technologies for water treatment and reuse. *Ind. Eng. Chem. Res.* **2014**, *53*, 11571–11592. [CrossRef]

61. Quesada, H.B.; Baptista, A.T.A.; Cusioli, L.F.; Seibert, D.; de Oliveira Bezerra, C.; Bergamasco, R. Surface water pollution by pharmaceuticals and an alternative of removal by low-cost adsorbents: A review. *Chemosphere* **2019**, *222*, 766–780. [CrossRef]

62. Zhou, Q.; Liu, G.; Arif, M.; Shi, X.; Wang, S. Occurrence and risk assessment of antibiotics in the surface water of Chaohu Lake and its tributaries in China. *Sci. Total Environ.* **2022**, *807*, 151040. [CrossRef] [PubMed]

63. Gyesi, J.N.; Nyaaba, B.A.; Darko, G.; Mills-Robertson, F.C.; Miezah, K.; Acheampong, N.A.; Frimpong, F.; Gyimah, G.; Quansah, B.; Borquaye, L.S. Occurrence of pharmaceutical residues and antibiotic-resistant bacteria in water and sediments from major reservoirs (owabi and barekese dams) in Ghana. *J. Chem.* **2022**, *2022*, 204. [CrossRef]

64. Ma, L.D.; Li, J.; Li, J.J.; Liu, M.; Yan, D.Z.; Shi, W.Y.; Xu, G. Occurrence and source analysis of selected antidepressants and their metabolites in municipal wastewater and receiving surface water. *Environ. Sci. Processes Impacts* **2018**, *20*, 1020–1029. [CrossRef] [PubMed]

65. Chen, Y.; Wang, J.; Xu, P.; Xiang, J.; Xu, D.; Cheng, P.; Wang, X.; Wu, L.; Zhang, N.; Chen, Z. Antidepressants as emerging contaminants: Occurrence in wastewater treatment plants and surface waters in Hangzhou, China. *Front. Public Health.* **2022**, *2022*, 10. [CrossRef]

66. Patrolecco, L.; Capri, S.; Ademollo, N. Occurrence of selected pharmaceuticals in the principal sewage treatment plants in Rome (Italy) and in the receiving surface waters. *Environ. Sci. Pollut. Res.* **2015**, *22*, 5864–5876. [CrossRef]

67. Ma, N.; Tong, L.; Li, Y.; Yang, C.; Tan, Q.; He, J. Distribution of antibiotics in lake water-groundwater-Sediment system in Chenhu Lake area. *Environ. Res.* **2022**, *204*, 112343. [CrossRef]

68. Chandel, N.; Ahuja, V.; Gurav, R.; Kumar, V.; Tyagi, V.K.; Pugazhendhi, A.; Kumar, G.; Kumar, D.; Yang, Y.-H.; Bhatia, S.K. Progress in microalgal mediated bioremediation systems for the removal of antibiotics and pharmaceuticals from wastewater. *Sci. Total Environ.* **2022**, *825*, 153895. [CrossRef]

69. Szabová, P.; Hencelová, K.; Sameliaková, Z.; Marcová, T.; Staňová, A.V.; Grabicová, K.; Bodík, I. Ozonation: Effective way for removal of pharmaceuticals from wastewater. *Mon. Chemie.* **2020**, *151*, 685–691. [CrossRef]

70. AL Saoud, H.A.; Sprynskyy, M.; Pashaei, R.; Kawalec, M.; Pomastowski, P.; Buszewski, B. Diatom biosilica: Source, physical-chemical characterization, modification, and application. *J. Sep. Sci.* **2022**, *45*, 3362–3376. [CrossRef]

71. Reberski, J.L.; Terzić, J.; Maurice, L.D.; Lapworth, D.J. Emerging organic contaminants in karst groundwater: A global level assessment. *J. Hydrol.* **2022**, *604*, 127242. [CrossRef]

72. Khan, H.K.; Rehman, M.Y.A.; Junaid, M.; Lv, M.; Yue, L.; Haq, I.U.; Xu, N.; Malik, R.N. Occurrence, source apportionment and potential risks of selected PPCPs in groundwater used as a source of drinking water from key urban-rural settings of Pakistan. *Sci. Total Environ.* **2022**, *807*, 151010. [CrossRef] [PubMed]

73. Wakkaf, T.; El Zrelli, R.; Kedzierski, M.; Balti, R.; Shaiek, M.; Mansour, L.; Tlig-Zouari, S.; Bruzaud, S.; Rabaoui, L. Characterization of microplastics in the surface waters of an urban lagoon (Bizerte lagoon, Southern Mediterranean Sea): Composition, density, distribution, and influence of environmental factors. *Mar. Pollut. Bull.* **2020**, *160*, 111625. [CrossRef] [PubMed]

74. Zhao, S.; Zhu, L.; Li, D. Microplastic in three urban estuaries, China. *Environ. Pollut.* **2015**, *206*, 597–604. [CrossRef] [PubMed]

75. Lestari, P.; Trihadiningrum, Y.; Wijaya, B.A.; Yunus, K.A.; Firdaus, M. Distribution of microplastics in Surabaya River, Indonesia. *Sci. Total Environ.* **2020**, *726*, 138560. [CrossRef] [PubMed]

76. Eo, S.; Hong, S.H.; Song, Y.K.; Han, G.M.; Shim, W.J. Spatiotemporal distribution and annual load of microplastics in the Nakdong River, South Korea. *Water Res.* **2019**, *160*, 228–237. [CrossRef]

77. Han, M.; Niu, X.; Tang, M.; Zhang, B.T.; Wang, G.; Yue, W.; Kong, X.; Zhu, J. Distribution of microplastics in surface water of the lower Yellow River near estuary. *Sci. Total Environ.* **2020**, *707*, 135601. [CrossRef]

78. Huang, D.; Li, X.; Ouyang, Z.; Zhao, X.; Wu, R.; Zhang, C.; Lin, C.; Li, Y.; Guo, X. The occurrence and abundance of microplastics in surface water and sediment of the West River downstream, in the south of China. *Sci. Total Environ.* **2021**, *756*, 143857. [CrossRef]

79. Pashaei, R.; Loiselle, S.A.; Leone, G.; Tamasi, G.; Dzingelevičienė, R.; Kowalkowski, T.; Gholizadeh, M.; Consumi, M.; Abbasi, S. Determination of nano and microplastic particles in hypersaline lakes by multiple methods. *Environ. Monit. Assesst.* **2021**, *193*, 668. [CrossRef]

80. Napper, I.E.; Baroth, A.; Barrett, A.C.; Bhola, S.; Chowdhury, G.W.; Davies, B.F.R.; Duncan, E.M.; Kumar, S.; Nelms, S.E.; Hasan Niloy, M.N.; et al. The abundance and characteristics of microplastics in surface water in the transboundary Ganges River. *Environ. Pollut* **2021**, *274*, 116348. [CrossRef]

81. Park, T.J.; Lee, S.H.; Lee, M.S.; Lee, J.K.; Lee, S.H.; Zoh, K.D. Occurrence of microplastics in the Han River and riverine fish in South Korea. *Sci. Total Environ.* **2020**, *708*, 134535. [CrossRef]

82. Rodrigues, M.O.; Abrantes, N.; Gonçalves, F.J.M.; Nogueira, H.; Marques, J.C.; Gonçalves, A.M.M. Spatial and temporal distribution of microplastics in water and sediments of a freshwater system (Antuã River, Portugal). *Sci. Total Environ.* **2018**, *633*, 1549–1559. [CrossRef] [PubMed]

83. Jiang, C.; Yin, L.; Li, Z.; Wen, X.; Luo, X.; Hu, S.; Yang, H.; Long, Y.; Deng, B.; Huang, L.; et al. Microplastic pollution in the rivers of the Tibet Plateau. *Environ. Pollut.* **2019**, *249*, 91–98. [CrossRef] [PubMed]

84. Eryaşar, A.R.; Gedik, K.; Şahin, A.; Öztürk, R.Ç.; Yılmaz, F. Characteristics and temporal trends of microplastics in the coastal area in the Southern Black Sea over the past decade. *Mar. Pollut. Bull.* **2021**, *173*, 112993. [CrossRef] [PubMed]

85. Zeri, C.; Adamopoulou, A.; Koi, A.; Koutsikos, N.; Lytras, E.; Dimitriou, E. Rivers and wastewater-treatment plants as microplastic pathways to eastern mediterranean waters: First records for the aegean sea, Greece. *Sustainability* **2021**, *13*, 328. [CrossRef]

86. Campanale, C.; Stock, F.; Massarelli, C.; Kochleus, C.; Bagnuolo, G.; Reifferscheid, G.; Uricchio, V.F. Microplastics and their possible sources: The example of Ofanto river in southeast Italy. *Environ. Pollut.* **2020**, *258*, 113284. [CrossRef]

87. Bikker, J.; Lawson, J.; Wilson, S.; Rochman, C.M. Microplastics and other anthropogenic particles in the surface waters of the Chesapeake Bay. *Mar. Pollut. Bull.* **2020**, *156*, 111257. [CrossRef]

88. Aigars, J.; Barone, M.; Suhareva, N.; Putna-Nimane, I.; Deimantovica-Dimante, I. Occurrence and spatial distribution of microplastics in the surface waters of the Baltic Sea and the Gulf of Riga. *Mar. Pollut. Bull.* **2021**, *172*, 112860. [CrossRef]

89. Jiang, Y.; Yang, F.; Zhao, Y.; Wang, J. Greenland Sea Gyre increases microplastic pollution in the surface waters of the Nordic Seas. *Sci. Total Environ.* **2020**, *712*, 136484. [CrossRef]

90. Lenaker, P.L.; Baldwin, A.K.; Corsi, S.R.; Mason, S.A.; Reneau, P.C.; Scott, J.W. Vertical Distribution of Microplastics in the Water Column and Surficial Sediment from the Milwaukee River Basin to Lake Michigan. *Environ. Sci. Technol.* **2019**, *53*, 12227–12237. [CrossRef]

91. Awet, T.T.; Kohl, Y.; Meier, F.; Straskraba, S.; Grün, A.L.; Ruf, T.; Jost, C.; Drexel, R.; Tunc, E.; Emmerling, C. Effects of polystyrene nanoparticles on the microbiota and functional diversity of enzymes in soil. *Environ. Sci. Europe* **2018**, 30. [CrossRef]

92. Vayghan, A.H.; Rasta, M.; Zakeri, M.; Kelly, F.J. Spatial distribution of microplastics pollution in sediments and surface waters of the Aras River and reservoir: An international river in Northwestern Iran. *Sci. Total Environ.* **2022**, *843*, 156894. [CrossRef] [PubMed]

93. Lusher, A.L.; McHugh, M.; Thompson, R.C. Occurrence of microplastics in the gastrointestinal tract of pelagic and demersal fish from the English Channel. *Mar. Pollut. Bull.* **2013**, *67*, 94–99. [CrossRef] [PubMed]

94. Lee, M.; Kim, H. COVID-19 Pandemic and Microplastic Pollution. *Nanomaterials* **2022**, *12*, 851. [CrossRef] [PubMed]

95. Liubartseva, S.; Coppini, G.; Lecci, R.; Creti, S. Regional approach to modeling the transport of floating plastic debris in the Adriatic Sea. *Mar. Pollut. Bull.* **2016**, *103*, 115–127. [CrossRef]

96. Zhang, Z.; Wu, H.; Peng, G.; Xu, P.; Li, D. Coastal Ocean dynamics reduce the export of microplastics to the open ocean. *Sci. Total Environ.* **2020**, *713*, 136634. [CrossRef]

97. Klein, S.; Dimzon, I.K.; Eubeler, J.; Knepper, T.P. Analysis, Occurrence, and Degradation of Microplastics in the Aqueous Environment. In *Freshwater Microplastics*; Springer: Cham, Switzerland, 2018; pp. 51–67.

98. Gwenzi, W.; Selvasembian, R.; Offiong, N.A.O.; Mahmoud, A.E.D.; Sanganyado, E.; Mal, J. COVID-19 drugs in aquatic systems: A review. *Environ. Chem. Lett.* **2022**, *2022*, 1–20. [CrossRef]

99. Wu, M.; Xiang, J.; Que, C.; Chen, F.; Xu, G. Occurrence and fate of psychiatric pharmaceuticals in the urban water system of Shanghai, China. *Chemosphere* **2015**, *138*, 486–493. [CrossRef]

100. Manbohi, A.; Mehdinia, A.; Rahnama, R.; Dehbandi, R. Microplastic pollution in inshore and offshore surface waters of the southern Caspian Sea. *Chemosphere* **2021**, *281*, 130896. [CrossRef]

101. Ahmed, M.B.; Zhou, J.L.; Ngo, H.H.; Guo, W.; Thomaidis, N.S.; Xu, J. Progress in the biological and chemical treatment technologies for emerging contaminant removal from wastewater: A critical review. *J. Hazard. Mater.* **2017**, *323*, 274–298. [CrossRef]

102. Gomes, J.; Costa, R.; Quinta-Ferreira, R.M.; Martins, R.C. Application of ozonation for pharmaceuticals and personal care products removal from water. *Sci. Total Environ.* **2017**, *586*, 265–283. [CrossRef]

103. Von Gunten, U. Ozonation of drinking water: Part II. Disinfection and by-product formation in presence of bromide, iodide or chlorine. *Water Res.* **2003**, *37*, 1469–1487. [CrossRef]

104. Andreozzi, R.; Canterino, M.; Marotta, R.; Paxeus, N. Antibiotic removal from wastewaters: The ozonation of amoxicillin. *J. Hazard. Mater.* **2005**, *122*, 243–250. [CrossRef] [PubMed]

105. D'Agostini, F.; Vadez, V.; Kholova, J.; Ruiz-Pérez, J.; Madella, M.; Lancelotti, C. Understanding the Relationship between Water Availability and Biosilica Accumulation in Selected C 4 Crop Leaves: An Experimental Approach. *Plants* **2022**, *11*, 1019. [CrossRef] [PubMed]

106. Ganiyu, S.O.; Van Hullebusch, E.D.; Cretin, M.; Esposito, G.; Oturan, M.A. Coupling of membrane filtration and advanced oxidation processes for removal of pharmaceutical residues: A critical review. *Sep. Purif. Technol.* **2015**, *156*, 891–914. [CrossRef]

107. Cherniak, S.L.; Almuhtaram, H.; McKie, M.J.; Hermabessiere, L.; Yuan, C.; Rochman, C.M.; Andrews, R.C. Conventional and biological treatment for the removal of microplastics from drinking water. *Chemosphere* **2022**, *288*, 132587. [CrossRef] [PubMed]

108. Ding, H.; Zhang, J.; He, H.; Zhu, Y.; Dionysiou, D.D.; Liu, Z.; Zhao, C. Do membrane filtration systems in drinking water treatment plants release nano/microplastics? *Sci. Total Environ.* **2021**, *755*, 142658. [CrossRef]

109. Misra, A.; Zambrzycki, C.; Kloker, G.; Kotyrba, A.; Anjass, M.H.; Franco Castillo, I.; Mitchell, S.G.; Güttel, R.; Streb, C. Water purification and microplastics removal using magnetic polyoxometalate-supported ionic liquid phases (magPOM-SILPs). *Angew. Chem. Int. Ed.* **2020**, *59*, 1601–1605. [CrossRef]

110. Dilshad, A.; Taneez, M.; Younas, F.; Jabeen, A.; Rafiq, M.T.; Fatimah, H. Microplastic pollution in the surface water and sediments from Kallar Kahar wetland, Pakistan: Occurrence, distribution, and characterization by ATR-FTIR. *Environ. Monit. Assess.* **2022**, *194*, 1–16. [CrossRef]

111. Roberts, J.; Kumar, A.; Du, J.; Hepplewhite, C.; Ellis, D.J.; Christy, A.G.; Beavis, S.G. Pharmaceuticals and personal care products (PPCPs) in Australia's largest inland sewage treatment plant, and its contribution to a major Australian river during high and low flow. *Sci. Total Environ.* **2016**, *541*, 1625–1637. [CrossRef]