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Review

Occurrence of Pharmaceuticals and Personal Care Products in the Water Environment of Poland: A Review

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Abstract: The issue of pharmaceuticals and personal care products (PPCPs) in the water environment has gained increasing interest worldwide. To determine the nature and extent of this problem for Poland, this paper presents a review of research on the presence of PPCPs in Poland, looking at results for different water samples, including wastewater (before and after treatment), landfill leachate, surface water (standing water bodies and rivers), seawater, groundwater and drinking water. The review is based on over 50 scientific articles and dissertations referring to studies of PPCPs. It also briefly outlines possible sources and the fate of PPCPs in the aquatic environment. The review of Polish research has revealed that studies have previously covered at least 39 PPCP groups (270 compounds in total). These studies focused mainly on wastewater and rivers, and only a few concerned landfill leachate and seawater. They most often reported on nonsteroidal anti-inflammatory drugs and antibiotics. The highest concentrations of the analysed PPCPs were found mainly in raw wastewater (e.g., naproxen, up to 551,960 ng/L), but they were also occasionally found in surface water (e.g., azithromycin, erythromycin, irbesartan and metoprolol) and in groundwater (e.g., N,N-diethyl-meta-toluamide, known as DEET, up to 17,280 ng/L). Extremely high concentrations of bisphenol A (up to 2,202,000 ng/L) and diclofenac (up to 108,340 ng/L) were found in landfill leachate. Although numerous substances have been detected, PPCPs are still not monitored regularly, which makes it difficult to obtain a clear understanding of their incidence in the water environment.

Keywords: PPCPs; emerging contaminants; wastewater; surface water; groundwater; water quality

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1. Introduction

The problem of pharmaceuticals and personal care products (PPCPs) in the water environment has gained increasing interest worldwide in the past two decades [1–4]. PPCPs belong to a large group of emerging contaminants, comprising organic chemical compounds occurring in small quantities in environmental samples, mostly in µg/L or ng/L. Among these substances, various medicines, cosmetics, disinfectants and even their metabolites are included. These compounds have been found in different water environments, such as wastewater, surface water, groundwater and tap water [5–8]. Their presence in the environment mostly results from excretion of compounds by organisms and the passing of these compounds into wastewater. Yet, they may also derive from improper disposal of expired pharmaceutical drugs and cleaning supplies [9]. Advanced treatment technologies and attenuation methods are currently tested against organic micro-contaminants occurring in sewage and water, e.g., ozonation, nanofiltration and reverse osmosis [10–16]. Unfortunately, these are not always effective when it comes to removal of PPCPs, as their efficiencies often vary depending on the compounds involved and their chemical properties [17,18]. Therefore, slowly degradable contaminants enter the natural environment and migrate to water bodies, including those which serve as drinking water supplies. In this regard, PPCPs may pose a threat to aquatic organisms and may be ingested by humans in tap water [7,19].

According to current Polish legislation, the monitoring of pharmaceutical and cosmetic residues in water is not obligatory. This concerns all types of water samples [20]. Limit values have not been set for drinking water either [21]. The Polish regulation regarding the list of priority substances (in force from 2019) [22] includes chemical compounds that may cause toxicity to organisms and accumulate in ecosystems. Thus, efforts must be made to prevent large quantities of these pollutants from entering the water environment. This list contains substances such as pesticides, polycyclic aromatic hydrocarbons and heavy metal compounds, among other things. Unfortunately, PPCPs are not addressed, even though the toxicity of some compounds within this group has been confirmed in previous studies [23–27].

Although the monitoring of PPCPs in water bodies is not mandatory, pilot surveys have been implemented in some countries to examine water quality and contamination with pharmaceutical and cosmetic residues [7,28–32]. Positive results from PPCP analyses have led to greater interest in the problem of water pollution by such compounds. This issue has also been raised at EU level, and the European Commission (EC) has released the European Union Strategic Approach to Pharmaceuticals in the Environment. In its communication from 2019 [33], the EC pointed to a severe problem involving pollution caused by certain drugs, and it highlighted the need to prevent the adverse effects of pharmacological substances on the natural environment. Furthermore, in 2020, a watch list of substances for Union-wide monitoring in the field of water policy was amended [34]. Currently, the list includes several pharmacological substances to be monitored, e.g., antibiotics, antifungal agents, veterinary drugs and one antidepressant, along with its metabolite. Despite this, PPCPs are still not included in regular monitoring in Poland. Furthermore, these contaminants have not yet been adequately addressed in other European countries [35,36]. In addition, in the recent proposal for a Directive of the European Parliament and of the Council on the quality of water intended for human consumption [37], new parameters were proposed and recommended for inclusion by the WHO (World Health Organization), including, among others, three substances classified as endocrine-disrupting compounds (EDCs): beta-oestradiol, nonylphenol and bisphenol A. In light of the above, it may be assumed that the monitoring strategies of EU countries and the scope of analysis of water samples will be extended in the future.

Many recent reviews have focused on the presence of PPCPs in water, on both a worldwide and a regional scale [1,3,8,38–41]. However, to the best of the authors' knowledge, to this day, such research has not been conducted for Poland. This paper presents a comprehensive review of research into conditions in Poland in terms of the presence of PPCPs in the water environment. It summarizes research conducted to date, in Poland, involving different environmental samples: raw and treated wastewater, landfill leachate, surface water (standing water bodies and flowing streams), seawater, groundwater and drinking water. It also briefly outlines possible sources, pathways and release mechanisms of these microcontaminants in the water environment. This paper presents the most common PPCP groups detected in water samples and ranges of concentrations for analysed compounds. An attempt has also been made to outline the overall scale of water contamination with PPCPs in Poland and to identify the gaps in current knowledge.

2. Sources and Fate of PPCPs in the Water Environment

Most compounds belonging to PPCP groups are derived from anthropogenic sources, except for some hormones and metabolites, which may have a natural origin [42]. There are numerous sources and pathways for these microcontaminants in the environment, which may lead to drinking water contamination (Figure 1). PPCPs are released into the environment due to human activities in different sectors of industry and urban life. As the considered substances are residues of medicines and personal care products, their major sources include pharmaceutical and chemical industries, households, hospitals, landfills, animal farming and veterinary clinics. Given the mentioned sources, contaminants enter water bodies primarily through wastewater discharges. This relates to both raw and treated

wastewater from wastewater treatment plants (WWTPs), because many of the treatment methods that are currently applied are ineffective in removing organic micropollutants. Therefore, microcontaminants are transported with treated wastewater from WWTPs to recipients, mostly rivers and streams [43,44]. Regarding raw sewage, PPCPs may be released into water or soil by leaky septic tanks, sewage pipelines or even illegal spilling in rural areas [45,46].

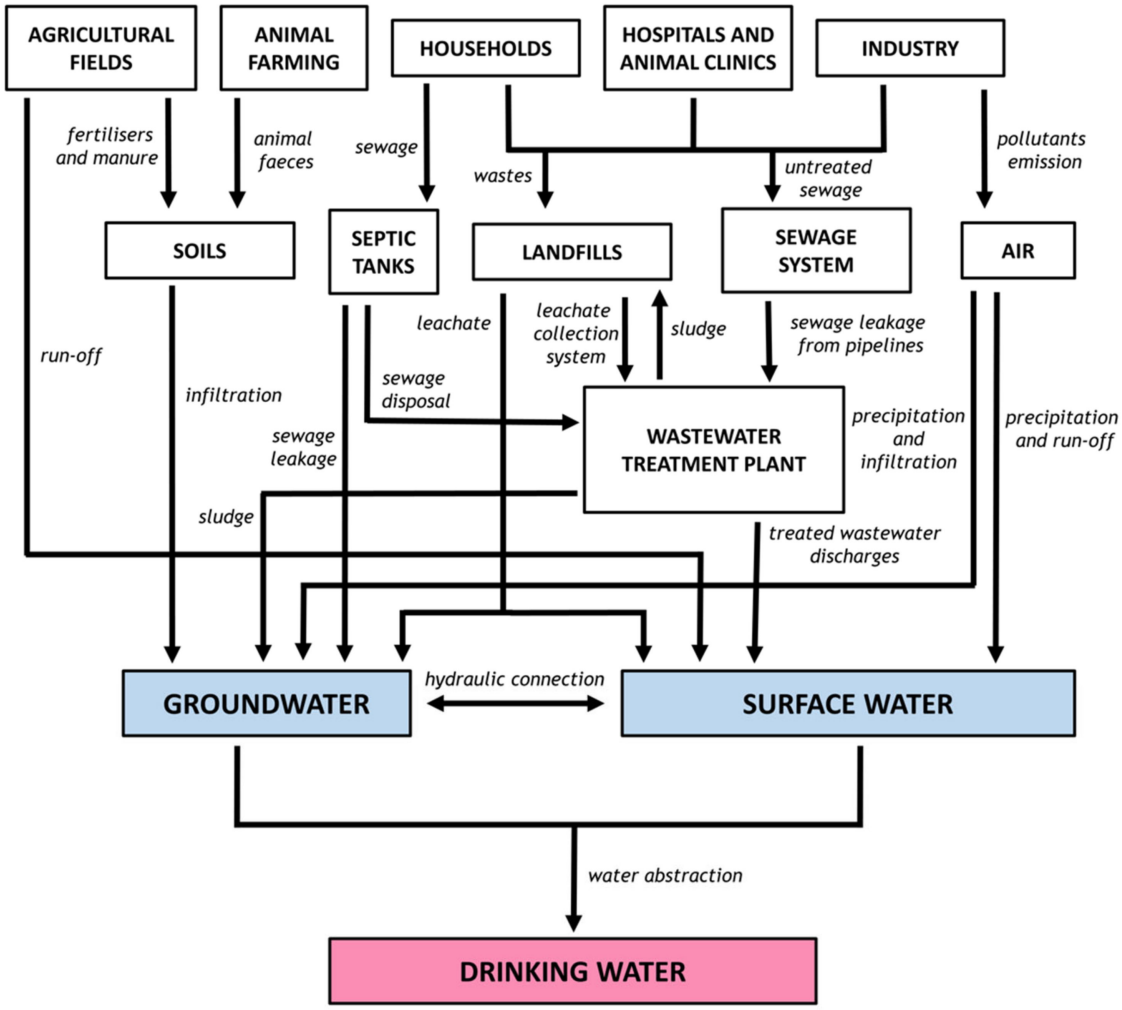

Figure 1. Possible sources and pathways of PPCPs in the natural environment.

Municipal and hospital sewage may carry a significant load of pharmaceuticals and their metabolites from urine, faeces and expired drugs, disposed via sinks and toilets, which then find their way into wastewater and WWTPs [47,48]. Some residues from expired medications are deposited along with domestic waste in municipal landfills, where harmful compounds are rinsed from waste by rainwater, passing to leachates, and then, through infiltration or run-off, to groundwater or surface water. Apart from municipal landfills, industrial waste lagoons are also related to potential sources of PPCPs, especially those associated with chemical and pharmaceutical industries [18,49,50]. This problem mainly concerns landfills without base sealing and illegal dumps [17,49]. Regarding landfills with base sealing, leachate is collected by a leachate drainage system and then directed to WWTPs.

It is noteworthy that pharmaceutical residues also include veterinary products, which occur in wastewater from veterinary clinics. Yet, they may also enter the environment through aquaculture, agricultural activity and animal farming (including concentrated animal feeding operations; CAFOs). Veterinary antibiotics, hormones and growth promoters present in manure are often spread on farmlands as fertilizers. Along with sewage sludge, which is also used for soil fertilization and may contain accumulated PPCP residues, they are the cause why agricultural activities are an additional source of these microcontaminants, which migrate to soil and groundwater or reach surface water due to the run-off [17,42,47,51–53]. Interestingly, PPCPs may also be released after the death of an organism, from its decaying remains. Thus, cemeteries and burial sites are taken into account as sources of medicine residues as well [14].

In the group of personal care products, various cosmetics and detergents are included: shampoos, toothpaste, sprays, dyes, perfumes, deodorants, sunscreens, soaps, washing powders, etc. Since they are applied to skin and on external surfaces, they may pass not only to wastewater but also directly to surface water as a result of tourist activities in summer. These substances can be washed from the skin as residues of lotions and other cosmetics or released into water along with human saliva, sweat and body fluids [54–56]. This also concerns seawater, which may receive additional quantities of PPCPs along with contaminated rivers [57].

It should be noted that compounds qualified as endocrine-disrupting chemicals (EDCs) are also regarded as PPCPs, even though, strictly speaking, they might be of industrial origin, e.g., those used as additives in the production of plastic, dyes, rubber, paper, packages, etc. Thus, industrial sewage and operations other than chemical and pharmaceutical industries may be responsible for PPCP contamination to some extent [58–61]. Moreover, some compounds are emitted into the atmosphere during industrial processes. Subsequently, they reach the surface through precipitation and contaminate surface water and shallow groundwater [62]. Due to hydraulic connection, surface water infiltration may influence groundwater quality, and thus PPCPs migrate to aquifers. This has been observed, for instance, in the vicinity of riverbank filtration sites [63]. Conversely, if PPCP pollution sources, such as unsealed landfills or leaky septic tanks, are situated within zones of groundwater discharge to rivers, then flow direction and migration of compounds are inverse, and surface water may be contaminated by groundwater.

As mentioned above, PPCPs originate primarily from anthropogenic sources. However, when present in water, they undergo numerous natural processes, which lead to their transformation, degradation or subsequent migration. The most important are photolysis, biodegradation, chemical transformations, hydrolysis, volatilization, sorption of contaminants on fine particles or organic matter, and dilution resulting from hydrodynamic dispersion and mixing with clean water or rainwater [14,64]. Their rates are conditioned by many factors, including the presence and activity of microorganisms, redox potential, oxygen content, depth, water temperature, pH and other physicochemical properties of water and soil [65]. All these processes partially result in natural attenuation and reduction of PPCP concentrations in water. Nevertheless, due to incomplete degradation of PPCPs under natural conditions and low effectiveness of treatment methods for PPCP removal, these microcontaminants may still appear in drinking water provided by waterworks [25,47].

3. Selection of Materials for the Review

In order to compile information on PPCPs in the water environment of Poland, scientific publications were searched via Google Scholar and ResearchGate, using the following keywords: pharmaceuticals and personal care products, PPCP, emerging contaminants, EC and microcontaminants. Since the number of related studies was limited, all research conducted in Poland was considered, regardless of publication date. Apart from scientific works such as papers, chapters in monographs and PhD theses, related project materials and legal acts were studied so that we could get acquainted with current trends and legislation in this field.

In total, 153 works related to PPCPs were identified. The extent of environmental contamination with PPCPs was our main interest, and only information on PPCP concentrations in real water samples was taken into account. During screening of the gathered materials, 42 works were excluded since they reported results for synthetic solutions in laboratory experiments. Of the remaining 111 publications and documents dealing with the general problem of PPCPs in water or legislation issues, 51 contained specific data on concentrations revealed during Polish research involving at least one type of water sample: raw and treated wastewater, landfill leachate, surface water, seawater, groundwater and drinking water. As most of the publications were not supplemented with raw databases, information such as sampling sites, type of water sample, analysed compounds and concentrations were extracted from the published texts.

4. Short Overview of PPCP Groups

A significant proportion of PPCP compounds constitutes of medicine residues. There are numerous groups of pharmaceutical substances used in the treatment of various diseases. According to the Central Statistical Office [66], over 70% of the Polish population takes pharmaceuticals, and this has remained constant in recent years. Almost half of all Poles use over-the-counter (OTC) medicines, including nonsteroidal anti-inflammatory drugs (NSAIDs) and some analgesics. Prescription drugs are less prevalent but are taken regularly, primarily by the elderly. Among these substances, the most commonly prescribed drugs are painkillers, which are taken by approximately 32% of adults. Other medicated groups of drugs are as follows: antihypertensives (prescribed for 21% of adults), cholesterol-lowering drugs (10%), drugs for cardiovascular diseases (9%), antidiabetic drugs (6%), hormones (5%), medicines for gastrointestinal disorders (4%), antibiotics (4%), antihistamines (3%), hypnotics (3%), anti-asthmatic agents (3%), antidepressants (3%) and other pharmaceuticals (10%). Furthermore, production of pharmaceutical preparations in Poland has increased over the past decade (from approximately 2140 million EUR in 2010 to 2315 million EUR in 2019) [67]. This also applies to personal care products, including all kinds of cosmetics, fragrances, detergents, and other substances used for hygiene purposes. As in the case medicaments, consumption of soap, detergents and washing preparations is higher than in previous years (increasing from approximately 609,000 t in 2017 to 652,000 t in 2019) [67,68]. Unfortunately, this fact may contribute to higher loads of relevant microcontaminants being released into the natural environment.

The conducted review of Polish research in water contamination from pharmaceuticals and personal care products revealed that studies covered at least 39 different PPCP groups. Among these, 270 compounds were distinguished in total. However, thanks to the latest analytical techniques, together with rising interest in PPCP occurrence in the environment, more emerging contaminants are being discovered in water samples. To date, the most common groups are antibiotics, NSAIDs, painkillers, stimulants, antidepressants, antihypertensives, hormones and PPCP metabolites. Nevertheless, some groups with few representative compounds are also being detected in relatively high concentrations, e.g., repellents (DEET, the only repelling substance considered in all studies) and alkylphenols (the main representative of which is bisphenol A, regarded as particularly dangerous due to its endocrine-disrupting properties). A summary of the characteristics of all PPCP groups considered in the reviewed Polish studies is presented in Table 1.

Table 1. The list of PPCP groups analysed in the water environment of Poland.

PPCP Groups (Number of Compounds)	Description	Compounds	Dominant PPCP in the Review
Alkylphenols (4)	Organic compounds obtained by the alkylation of phenols; used in industry, e.g., as detergents and additives in making fuels, lubricants, fragrances, fire retardant materials, etc.	Bisphenol A, Bisphenol S, Nonylphenol, Octylphenol	Bisphenol A up to 2,202,000 ng/L in landfill leachate [69]
Alpha-1 blockers (2)	Drugs blocking the effect of alpha-1-adrenergic receptors; used to treat prostate enlargement, hypertension and post-traumatic stress disorder	Alfuzosin, Carvedilol	not detected
Antiarrhythmic drugs (2)	Medications used to treat cardiac arrhythmias	Amiodarone, Propafenone	Propafenone up to 87 ng/L in surface water [70]
Antibacterial agents (2)	Antimicrobial substances applied to destroy bacteria and reduce the possibility of infection; previously used in personal care products, e.g., soaps, lotions, toothpaste, detergents, etc.	Triclocarban, Triclosan	Triclosan up to 6721.7 ng/L in raw wastewater [71]
Antibiotics (75)	Medications killing or inhibiting the growth of bacteria; used for fighting and preventing bacterial infections	Amoxicillin, Ampicillin, Azithromycin, Carbadox, Cefadroxil, Cefalonium, Cefapirin, Cefazolin, Cefoperazone, Cefotaxime, Cefquinome, Ceftazidime, Ceftiofur, Cephalexin, Chloramphenicol, Chlorotetracycline, Ciprofloxacin, Clarithromycin, Clindamycin, Cloxacillin, Danofloxacin, Dicloxacillin, Difloxacin, Dihydrostrepromycin, Doxycycline, Enoxacin, Enrofloxacin, Erythromycin, Fleroxacin, Flumequine, Fluraltadone, Josamycin, Levofloxacin, Lincomycin, Lomefloxacin, Marbofloxacin, Metronidazole, Nafcillin, Nalidixic acid, Neomycin, Nitrofurantoin, Norfloxacin, Ofloxacin, Oleandomycin, Oxacillin, Oxolinic acid, Oxytetracycline, Penicillin G, Rifampicin, Roxithromycin, Sarafloxacin, Spectinomycin, Spiramycin, Streptomycin, Sulfachloropiridazine, Sulfadiazine, Sulfadoxine, Sulfadimethoxine, Sulfamerazine, Sulfamethazine, Sulfamethizole, Sulfamethoxazole, Sulfamethoxypyridazine, Sulfamonomethaxine, Sulfanilamide, Sulfapyridine, Sulfasalazine, Sulfathiazole, Sulfisoxazole, Tetracycline, Tiamulin, Tilmicosin, Trimethoprim, Tylosin, Vancomycin	Metronidazole up to 7400 ng/L in raw wastewater [72]

Table 1. *Cont.*

PPCP Groups (Number of Compounds)	Description	Compounds	Dominant PPCP in the Review
Antidementia agents (1)	Medications used to treat dementia and Alzheimer's disease	Memantine	not detected
Antidepressants (12)	Medications used in the treatment of depression, anxiety disorders, chronic pains and some addictions	Amitriptyline, Citalopram, Clomipramine, Desvenlafaxine, Diazepam, Doxepin, Fluoxetine, Fluvoxamine, Imipramine, Mirtazapine, Sertraline, Venlafaxine	Diazepam up to 531 ng/L in raw wastewater [17]
Antidiabetic drugs (2)	Drugs used in the treatment of diabetes, altering the glucose level in the blood	Gilbenclamide, Metformin	Metformin up to 16,790.7 ng/L in raw wastewater [73]
Antiepileptics (6)	Pharmaceutical drugs used in the treatment of epileptic seizures, neuropathic pain and as mood stabilizers	Carbamazepine, Gabapentin, Lamotrigine, Lorazepam, Phenytoin, Primidone	Carbamazepine up to 5127.8 ng/L in treated wastewater [73]
Antifungal agents (2)	Medications for fungal infections or preservatives used to control mould and fungal diseases	Fluconazole, Thiabendazole	Thiabendazole up to 104 ng/L in surface water [72]
Antihistamines and histamine receptor modulators (3)	Drugs opposing the activity of histamine receptors in the body and used to treat allergies	Cimetidine, Fexofenadine, Ranitidine	Rantidine up to 5702.2 ng/L in raw wastewater [73]
Antihypertensives (11)	Drugs lowering blood pressure; used to treat hypertension	Almodipine, Enalapril, Irbesartan, Labetalol, Losartan, Nifedipine, Quinapril, Ramipril, Telmisartan, Valsartan, Verapamil	Valsartan up to 92,532.7 ng/L in raw wastewater [71]
Antiischemic agents (1)	Medications used to treat insufficient blood flow, e.g., for angina	Trimetazidine	Trimetazidine up to 826.7 ng/L in raw wastewater [73]
Antiparasitics (5)	Substances destroying or inhibiting the growth of parasites; used for the treatment of parasitic diseases	Flubendazole, Ivermectin, Mebendazole, Sulfaquinoxalline, Tinidazole	Mebendazole up to 9.8 ng/L in surface water [74]
Antiretroviral drugs (1)	Medications inhibiting viruses development; used for treating viral infections	Darunavir	not detected
Antithrombotics (1)	Drugs reducing and preventing the formation of blood clots	Warfarin	not detected
Beta-blockers (9)	Medications used to manage abnormal heart rhythms, prevent heart attacks and treat high blood pressure	Acebutolol, Atenolol, Bisoprolol, Celiprolol, Metoprolol, Nadolol, Pindolol, Propranolol, Sotalol	Sotalol up to 2120 ng/L in surface water [75]

Table 1. *Cont.*

PPCP Groups (Number of Compounds)	Description	Compounds	Dominant PPCP in the Review
Beta-2-adrenergic agonists (2)	Drugs acting on the beta-2-adrenergic receptor and causing smooth muscle relaxation; used to treat asthma and other pulmonary disorders	Salbutamol, Terbutaline	not detected
Chemotherapeutic agents (2)	Substances used in chemotherapy for cancer	Cyclophosphamide, Ifosfamide	Cyclophosphamide up to 33.3 ng/L in raw wastewater [73]
Contrast agents (5)	Substances used to increase the contrast of structures within the body in medical imaging	Diatrizoate, Iohexol, Iomeprol, Iopamidol, Iopromide	Iopromide up to 27,000 ng/L in raw wastewater [76]
Corrosion inhibitors (2)	Compounds decreasing the corrosion rate of material into contact with the fluid; used in industry, over-the-counter products and as additives to water to prevent leaching of lead or copper from pipes	1H-Benzotriazole, Tolyltriazole	1H-Benzotriazole up to 180 ng/L in groundwater [63]
Corticosteroids (3)	Synthetic analogues of steroid hormones produced in the adrenal cortex; used as pharmaceutical drugs in the treatment of various conditions, e.g., skin diseases, asthma, cancers, tumours, allergies, inflammation and others	Dexamethasone, Flumethasone, Prednisolone	not detected
Cosmetic and pharmaceutical preservatives (4)	Substances added to pharmaceutical drugs and cosmetics, preventing decomposition by microbial growth or by undesirable chemical changes, mostly parabens	Butylparaben, Ethylparaben, Methylparaben, Propylparaben	Methylparaben up to 40,898.6 ng/L in raw wastewater [71]
Diuretics (3)	Medications increasing the production of urine and the excretion of water from a body	Chlorothiazide, Furosemide, Hydrochlorothiazide	Hydrochlorothiazide up to 5072.3 ng/L in raw wastewater [73]
Fibrates (6)	Medications used for metabolic disorders, mainly hypercholesterolemia	Bezafibrate, Ciprofibrate, Clofibric acid, Etofibrate, Fenofibrate, Gemfibrozil	Bezafibrate up to 1000 ng/L in raw wastewater [76]

Table 1. *Cont.*

PPCP Groups (Number of Compounds)	Description	Compounds	Dominant PPCP in the Review
Hormones (11)	Compounds responsible for the regulation of physiological processes and behavioural activities in organisms, some of which used as medications with a dose far greater than naturally occurs in a body, especially for hormonal contraception or hormone replacement therapy	17α-Ethynyloestradiol, 17β-Oestradiol, Androstenedione, Diethylstilboestrol, Oestriol, Oestrone, Equilin, Levonorgestrel, Norethisterone, Progesterone, Testosterone	Levonorgestrel up to 1529.8 ng/L in raw wastewater [73]
Hypnotics (2)	Psychoactive drugs used in the treatment of insomnia and for surgical anaesthesia	Nitrazepam, Temazepam	Temazepam up to 257 ng/L in treated wastewater [71]
Immunosuppressive drugs (2)	Medicines used to inhibit or prevent the activity of the immune system	Mycophenolic acid, Tacrolimus	Mycophenolic acid up to 179.6 ng/L in surface water [77]
Lipid-modifying agents (4)	Statin medications used to treat elevated lipid levels, preventing cardiovascular disease	Atorvastatin, Lovastatin, Pravastatin, Simvastatin	Atorvastatin up to 2756.5 ng/L in raw wastewater [71]
Metabolites (33)	Products of metabolism, degradation and elimination of parent compounds	17α-hydroxyprogesterone, 4-*n*-nonylphenol, 4-*n*-octylphenol, ATH, Benzoylecgonine, Carbamazepine 10,11-dihydroxy, 2-hydroxy Carbamazepine, Cocaethylene, Cotinine, Creatinine, DAMI (N-Deethylamiodarone), Diclofenac-4′-hydroxy, Dihydrocodeine, Dihydromorphine, Dimethyl-aminophenazone, EDDP, Erythromycin-H_2O, HMMA, Ibuprofen-2-hydroxy, Ibuprofen-carboxy, N-acetyl-sulfamethoxazole, Naproxene-o-desmethyl, Nordiazepam, Norephedrine, Norketamine, Normorphine, Noroxycodone, O-desmethyltramadol, Oxazepam, Oxcarbazepine, Oxypurinol, Paraxanthine, Salicylic acid	Paraxanthine up to 174,336.8 ng/L in raw wastewater [71]
Nonsteroidal anti-inflammatory drugs (14)	Drugs used for the treatment of acute or chronic conditions, reducing pain, decreasing fever and inflammation and preventing blood clots	5-Aminosalicylic acid, Acetylsalicylic acid, Diclofenac, Diflunisal, Fenoprofen, Flurbiprofen, Ibuprofen, Indomethacin, Ketoprofen, Ketorolac, Naproxen, Phenazone, Propyphenazone, Tolmetin	Naproxen up to 551,960 ng/L in raw wastewater [78]

Table 1. *Cont.*

PPCP Groups (Number of Compounds)	Description	Compounds	Dominant PPCP in the Review
Painkillers (11)	Drugs and substances used to achieve analgesia and relief from pain	Codeine, Hydrocodone, Ketamine, Metamizole, Methadone, Morphine, Oxycodone, Oxymorphone, Paracetamol, Pentoxifylline, Tramadol	Paracetamol up to 51,400 ng/L in raw wastewater [79]
PDE5 inhibitors (2)	Vasodilating drugs used to treat erectile dysfunction	Sildenafil, Vardenafil	Sildenafil up to 81.17 ng/L in surface water [80]
Repellents (1)	Substances applied for the determent of insects to prevent insect-borne diseases	DEET	DEET up to 2620 ng/L in raw wastewater [50]
Stimulants (16)	Drugs increasing the activity of the central nervous system and substances with pleasurable and invigorating effects	4-MEC, Amphetamine, Benzylpiperazine, Caffeine, Cocaine, mCCP, MDA, MDEA, MDMA, MDPV, Mephedrone, Methamphetamine, MPD, Nicotine, PMA, Theophylline	Nicotine up to 423,978 ng/L in raw wastewater [71]
Sugar substitutes (2)	Artificial, non-nutritive food additives providing a sweet taste	Saccharin, Sucralose	Saccharin up to 360 ng/L in surface water [81]
Synthetic musks (2)	Synthetic aroma compounds emulating the scent of animal musks; used as flavourings and fixatives in cosmetics, detergents, perfumes and foods	Galaxolide, Tonalide	Galaxolide up to 1200 ng/L in raw wastewater [76]
UV filters (3)	Compounds blocking or absorbing ultraviolet light; used, e.g., in sunscreens, cosmetics and as additives in flavourings, perfumes and plastic packaging	3-(4-ethylbenzylidene)camphor, Benzophenone, Oxybenzone	3-(4-Methylbenzylidene)camphor up to 18,010 ng/L in raw wastewater [50]
Vasodilators (1)	Drugs widening blood vessels; used, e.g., in the treatment of peripheral and cerebral vascular disorders	Nafronyl	Nafronyl up to 27.9 ng/L in raw wastewater [73]

5. PPCPs in the Water Environment of Poland

The following review of conditions in Poland was based on scientific articles and dissertations referring to studies of pharmaceuticals and personal care products in different water samples, including wastewater before and after treatment, landfill leachate, surface water (flowing streams and standing water bodies), seawater, groundwater and drinking water. Information on the occurrence of PPCPs in Polish water was found in over 50 publications. The first information on PPCP occurrence in environmental samples was mentioned in a study from 2003, and the latest papers were published at the beginning of 2021. The studies focused mainly on wastewater and rivers (Figure 2). Little research concerned PPCP occurrence in landfill leachate and seawater. In the case of drinking water and

groundwater (which is an essential source of drinking water in many regions of Poland), analyses were performed less often, compared with types of samples considered to be more vulnerable to PPCP residues. Seemingly, water types less likely to be contaminated are rarely subjected to such analyses because the presence of PPCPs in these samples is not always apparent. Nevertheless, as this review shows, none of the water types may be regarded as utterly free from microcontaminants.

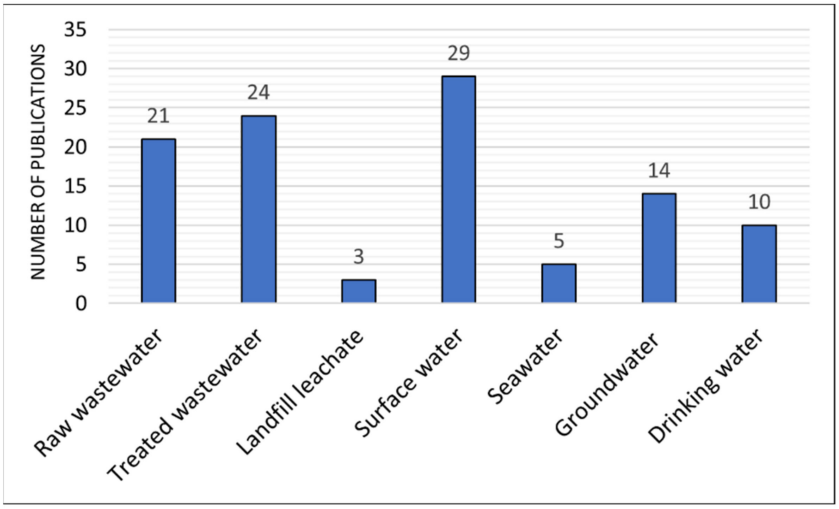

Figure 2. Number of publications of Polish research on PPCPs in water for different types of samples (in the years 2003–2021).

The studies covered a wide range of contaminants from different PPCP groups (Figure 3). In general, the most frequently analysed groups were non-steroidal anti-inflammatory drugs (NSAIDs) and antibiotics, with information on their determination found in 36 and 21 scientific papers, respectively. Other compounds of greater interest were anti-epileptics, metabolites, stimulants, beta-blockers, hormones and antidepressants. The other PPCP groups were examined rarely or only occasionally.

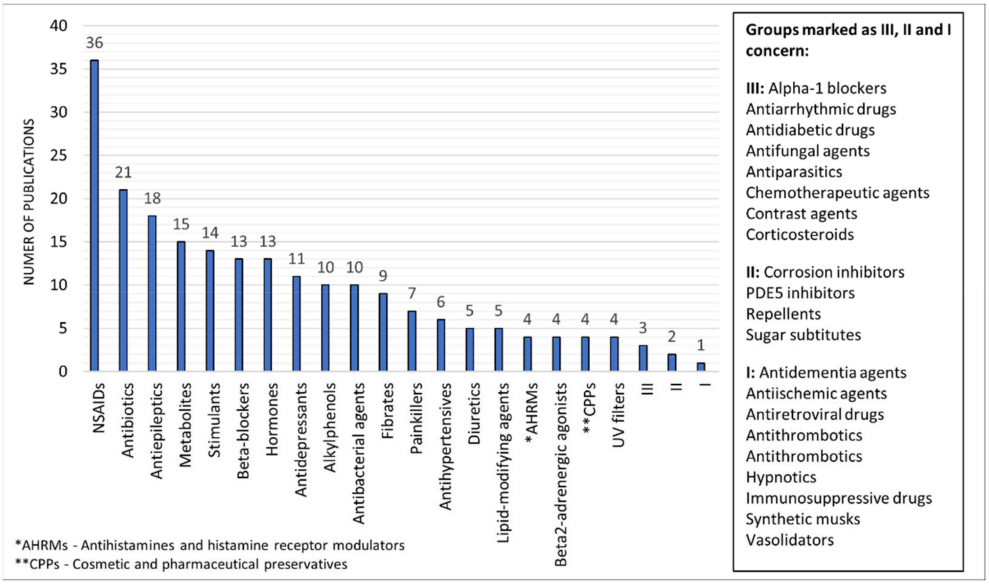

Figure 3. Number of publications of Polish research concerning compounds from the considered PPCP groups.

In Polish research, two analytical methods have dominated. The most frequently applied method has been gas chromatography coupled with mass spectrometry (GC-MS) and, less often, the liquid chromatography–mass spectrometry (LC-MS) method. According to the studies that we considered, many of the analysed compounds were in concentrations below the limit of detection (<LOD) or limit of quantification (<LOQ) established during the research. Such results were reported especially for groundwater, drinking water, some streams and lakes. Although these outcomes may seem irrelevant, the mere fact of performing such analyses suggests a growing interest in the problem concerning PPCPs in this country. Moreover, knowledge about the list of previously analysed substances (and their presence or absence in the environment) allows us to focus on the substances (and water bodies) which pose the greatest issue in terms of pollution levels. Apart from <LOD and <LOQ results, in several cases, only the fact of detection of PPCPs was mentioned, without any indication being given of specific values (i.e., above the limit of detection, >LOD). The other results were diversified in terms of concentrations, depending on the particular compound and the type of water sample.

The number of PPCP groups taken into account in the reviewed studies was found to be similar for wastewater, surface water, groundwater and drinking water (Table 2). This number was significantly lower for landfill leachate and seawater. However, the number of compounds analysed for each of these groups varied considerably, and some groups contained only one or two substances. Hence, our comparison does not specifically indicate the scale of the problem associated with PPCP contamination. However, it does provide an insight into the general trend in current Polish research focusing on PPCP occurrence in the water environment.

Table 2. PPCP groups studied in considered types of water samples in the review (marked as X).

PPCP Groups	Raw Wastewater	Treated Wastewater	Landfill Leachate	Surface Water	Seawater	Groundwater	Drinking Water
Alkylphenols	X	X	X	X		X	X
Alpha-1 blockers		X		X		X	
Antiarrhythmic drugs				X		X	X
Antibacterial agents	X	X	X	X		X	X
Antibiotics	X	X		X	X	X	X
Antidementia agents				X		X	
Antidepressants	X	X		X		X	X
Antidiabetic drugs	X	X		X		X	X
Antiepileptics	X	X	X	X	X	X	X
Antifungal agents	X	X		X		X	X
Antihistamines and histamine receptor modulators	X	X		X		X	X
Antihypertensives	X	X		X		X	X
Antiischemic agents	X	X				X	X
Antiparasitics				X		X	X
Antiretroviral drugs				X			X
Antithrombotics				X		X	
Beta-blockers	X	X		X		X	X

Table 2. *Cont.*

PPCP Groups	Raw Wastewater	Treated Wastewater	Landfill Leachate	Surface Water	Seawater	Groundwater	Drinking Water
Beta-2-adrenergic agonists				X		X	X
Chemotherapeutic agents	X	X		X		X	X
Contrast agents	X			X		X	
Corrosion inhibitors				X		X	
Corticosteroids		X		X			
Cosmetic and pharmaceutical preservatives	X	X	X	X		X	
Diuretics	X	X		X		X	X
Fibrates	X	X		X		X	X
Hormones	X	X	X	X		X	X
Hypnotics	X	X		X			
Immunosuppressive drugs				X			X
Lipid-modifying agents	X	X		X		X	X
Metabolites	X	X	X	X		X	X
Nonsteroidal anti-inflammatory drugs	X	X	X	X	X	X	X
Painkillers	X	X		X	X	X	X
PDE5 inhibitors	X	X		X			
Repellents	X	X	X	X		X	
Stimulants	X	X		X	X	X	X
Sugar substitutes				X		X	
Synthetic musks	X						
UV filters	X	X	X	X		X	
Vasodilators	X	X				X	X
IN TOTAL	**28**	**28**	**9**	**36**	**5**	**33**	**27**

The current situation with regard to water environment contamination with PPCPs in Poland is summarized for the above-mentioned types of water samples (raw wastewater, treated wastewater, landfill leachate, surface water, seawater, groundwater and drinking water) in the following sections. We have made an effort to present the most important outcomes achieved in the reviewed research, highlighting compounds and water types characterized by the highest concentrations and level of contamination with PPCPs. All the detailed results found in the studied literature are included in the Supplementary Material (Table S1). It should be noted that many of the results are single values and the sole information on PPCP concentrations in a given water sample. Unfortunately, the analysed literature frequently did not provide meaningful information concerning, for example, the exact location of sampling points. As for works on wastewater samples before and after treatment, more often than not, there was a lack of details on applied treatment technologies and their stages. Therefore, it was impossible to assess the treatment efficiency

or attenuation rate for these compounds in natural conditions. Moreover, the limited number of results did not allow us to perform a statistical analysis, and we only took into account ranges of reported PPCP concentrations (minimum and maximum values). Hence, the outcomes from this review provide only an overall view of the level of water contamination from PPCPs in Poland. Despite this, the review did identify gaps in current knowledge and revealed key water types and substances, which could become interesting subjects for future research.

5.1. Raw Wastewater

After surface water, wastewater was the second most studied water environment in Poland in terms of PPCPs. Study results relating to the presence of these compounds in untreated wastewater were found in 21 publications and included 136 compounds, mainly metabolites (23), antibiotics (22), nonsteroidal anti-inflammatory drugs (11), stimulants (11) and painkillers (9). The authors of the publications did not always report in detail the number and location of wastewater treatment plants from which the wastewater samples were collected for analysis; therefore, it is not possible to present detailed data on the sources of wastewater samples analysed in Poland. The full list of PPCPs investigated in untreated wastewater is provided in Table S1. Here, only the most interesting findings are described.

The most frequently tested PPCPs in raw wastewater in Poland were non-steroidal anti-inflammatory drugs [17,44,50,71,73,76,78,82–87]. The maximum concentration of naproxen (551,960 ng/L) turned out to be the highest among all the PPCPs analysed. Four other compounds from this group were found in high concentrations as well: ketoprofen (up to 233,630 ng/L), diclofenac (up to 40,570.2 ng/L), ibuprofen (up to 31,250 ng/L), and flurbiprofen (up to 4952 ng/L). As many as 22 compounds belonging to the antibiotic group were found in raw wastewater [17,71–73,76,88]. The highest concentrations were found for metronidazole (7400 ng/L), but five other compounds were also found with their maximum concentrations exceeding 1000 ng/L: ciprofloxacin, vancomycin, sulfamethoxazole, sulfasalazine and clarithromycin. Of the nine painkillers analysed, paracetamol was the most frequently tested in raw wastewater. Its concentration ranged from <LOD to 51,400 ng/L [73,79,83,86,87]. Pentoxifylline [76], codeine, morphine, methadone and ketamine were also found [71]. Among 11 investigated stimulants, the most frequently studied was caffeine, with concentrations reaching 89,544.5 ng/L [17,71,73,76]. Even higher concentrations were found in the case of nicotine, from 19,035 ng/L to 423,978 ng/L [71]. The other stimulants detected were amphetamine, cocaine, MDMA (midomafetamine), PMA (para-Methoxyamphetamine) and mephedrone [71].

Metabolites, degradation products of other compounds, represent the largest group of compounds (23) studied in untreated wastewater. Most metabolites were analysed in one project [71]. Compounds detected in the highest concentrations included paraxanthine [71], salicylic acid [73,84,87], ibuprofen-2-hydroxy [76], cotinine [71], and N-acetyl-sulfamethoxazole [88].

Among the other PPCPs investigated in raw wastewater in Poland, high concentrations were detected for metformin (up to 16,790.7 ng/L) (representing anti-diabetic drugs) [73], atorvastatin (up to 2756.5 ng/L) (representing lipid-modifying agents) [71], valsartan (up to 92,532.7 ng/L), irbesartan (up to 3142.3 ng/L) and losartan (up to 1708.8 ng/L) (a form of antihypertensive medication) [71,73] or, in the case of diuretics, furosemide (up to 3372.5 ng/L) and hydrochlorothiazide (up to 5072.3 ng/L) [73,82]. Of the seven hormones investigated in untreated wastewater in Poland, three were not detected in any sample (oestriol, 17α-ethynyloestradiol and diethylstilboestrol) [50,87], while the rest was found in various concentrations: levonorgestrel (up to 1529.8 ng/L) [73], 17β-oestradiol (max. 1067.8 ng/L) [17,50,71,87], oestrone (max. 773.3 ng/L) [14,50,71,87] and progesterone (up to 89.3 ng/L) [73].

Relatively high concentrations of cosmetic and pharmaceutical preservatives were found in raw wastewater, with the highest values detected for methylparaben (max.

40,898.6 ng/L) [50,71]. High concentrations were also found for another group of compounds, alkylphenols [50,69,71,78,85,89,90]. In this group, bisphenol A (up to 12,060 ng/L) and nonylphenol (up to 102,540 ng/L) were found at the highest concentration. One study of raw wastewater in Poland included two UV filters, benzophenone and 3-(4-methylbenzylidene)camphor, as well as the commonly used repellent DEET [50]. Apart from these, carbamazepine (up to 3217.1 ng/L) was a relatively frequently tested PPCP, a form of anti-epileptic medication [50,73,76,84,89].

5.2. Treated Wastewater

The results of PPCP studies of treated wastewater in Poland were published in 24 publications. The compounds tested in treated wastewater were almost the same as those tested in untreated water. In total, 127 PPCP compounds were analysed in treated wastewater. In general, significantly lower concentrations of studied PPCPs were observed in comparison with untreated wastewater. Only a few substances were present at similar levels, and even more rarely, higher concentrations were observed in comparison with raw wastewater. The last case was found to relate almost exclusively to metabolites and may also have resulted from the limited number of samples tested and from the fact that sampling procedures did not include sampling of the same wastewater before and after treatment processes. As a result, the wastewater samples were collected at random, and the results do not represent the effectiveness of wastewater treatment.

The most commonly investigated PPCPs were non-steroidal anti-inflammatory drugs [17,44,71,73,75,78,79,82–87,91–93]. The highest concentrations were detected in the case of ibuprofen (up to 22,610 ng/L), but elevated values were also found for naproxen, diclofenac and ketoprofen. As many as 21 antibiotics were tested in treated wastewater in Poland [71–73,88], but most of these were tested only once per study. The highest concentrations were found for sulfamethoxazole (508–770 ng/L). The presence of nine painkillers was also studied in treated wastewater [71,73,75,83,86,87,91], and paracetamol was found in the highest concentrations (max. 3824 ng/L). The other compounds, metamizole, codeine, morphine, methadone and ketamine, were found in much lower concentrations.

A fairly frequently tested PPCP group was that of alkylphenols [50,71,78,85,89,90,93], with bisphenol A reaching the highest concentration (up to 10,840 ng/L). Among cosmetic and pharmaceutical preservatives, the most commonly investigated was methylparaben (max. 1440 ng/L). Benzophenone (representing UV filters) was found to have a maximum concentration of 1400 ng/L [50], and DEET was found in the range of <LOD–1150 ng/L [50].

The studies indicated that some stimulants were found in treated wastewater but at much lower concentrations, compared with raw wastewater. The highest concentrations were observed for caffeine (up to 2868.5 ng/L) and nicotine (up to 2222.5 ng/L), while no evidence of amphetamine, methamphetamine, MDA (tenamfetamine), MDEA (3,4-Metylenodioksy-N-etyloamfetamina), mCCP (metachlorophenylpiperazine) or MDPV (3,4-Metylenodioksypirowaleron) was found [17,71,73,94].

Among the 19 investigated metabolites in treated wastewater [50,71,73,75,84,87,88,91,92], the highest concentrations were detected for paraxanthine (up to 1881.3 ng/L). High concentrations in treated wastewater were observed also for carbamazepine (max. 5127.8 ng/L) [50,73,75,84,89,91], venlafaxine and diazepam (antidepressants) [17,71,95]. Metformin (an anti-diabetic drug) was found in low concentrations (max. 62.9 ng/L) [73], as well as trimetazidine (an anti-ischemic agent, up to 457.8 ng/L) [73].

It is also important to mention that four compounds from the antihypertensives group have been found in treated wastewater investigated to date: valsartan, irbesartan, losartan and ramipril [71,73]. High concentrations were found for two compounds representing diuretics: hydrochlorothiazide (up to 4313.7 ng/L) and furosemide (up to 1879.2 ng/L) [73,82].

Of the seven hormones tested, four were not found in treated wastewater, and the remaining three were detected in low concentrations: 17β-oestradiol (<LOD–432 ng/L), oestrone (<LOD–249 ng/L), and progesterone (9.4–13.4 ng/L) [17,50,71,73,87]. Numerous studies [50,71,78,84,85] reported evidence of one antibacterial agent (triclosan), found at

low concentrations (up to 910 ng/L). Studies of wastewater treated in Gdańsk [73] and Cracow [71] revealed presence of two compounds belonging to the group of antihistamines and histamine receptor modulators: ranitidine and fexofenadine. From the beta-blockers group, metoprolol and atenolol were found [17,75,84,91]. Only two compounds belonging to fibrates were detected: bezafibrate and clofibric acid [44,71,84,96]. Thiabendazole (an antifungal agent) was tested only twice, and the results showed slightly higher concentrations compared with raw wastewater [72].

Three PPCP groups analysed in treated wastewater were not detected at all: alpha1-blockers [75,91], corticosteroids [75,91] and PDE5 inhibitors [71].

5.3. Landfill Leachate

To date, landfill leachate in Poland has been studied only rarely in comparison with wastewater. Nevertheless, landfill leachate is also considered to be wastewater, which may strongly influence the quality of the water environment. It particularly concerns groundwater in the vicinity of both industrial and municipal dumps without base sealing, where leachate may infiltrate shallow aquifers along with microcontaminants. Although there have been few results for PPCP occurrence in leachate in Poland, they have confirmed the presence and high concentrations of some PPCPs in such samples.

Landfill leachate studies revealed the presence of nine PPCP groups: alkylphenols, antibacterial agents, antiepileptics, cosmetic and pharmaceutical preservatives, hormones, metabolites, NSAIDs, repellents and UV filters. In total, these included 16 compounds reported in three publications between 2016 and 2019. Almost all the results showed concentration values above the detection limit. The published results mainly referred to landfills located in north-eastern Poland. The most common group was cosmetic and pharmaceutical preservatives (4 compounds), among which methylparaben dominated (up to 17,150 ng/L). However, in this area, the maximum PPCP concentrations in landfill leachate were found for other compounds, e.g., DEET (up to 202,420 ng/L) and diclofenac (up to 108,340 ng/L) [49,50]. The remaining analysed PPCPs occurred in lower amounts, e.g., UV filters (benzophenone and 3-(4-methylbenzylidene)camphor, whose concentrations reached up to 16,640 ng/L) and metabolites (4-*n*-octylphenol and 4-*n*-nonylphenol). Concentrations of diethylstilboestrol, 17β-oestradiol, oestrone and triclosan were at a moderate level. Trace amounts of carbamazepine from the antiepileptics group were also expected [49,50]. Interestingly, another study (conducted in north-eastern Poland) found bisphenol A in leachate in considerable amounts, reaching 2,202,000 ng/L [69].

5.4. Surface Water

Among natural water sources in Poland, surface water bodies have been relatively well researched, and almost all the considered PPCP groups (36) have been investigated. This especially applies in the case of flowing streams, with information on PPCP concentrations found for 39 rivers, including the longest ones, such as the Vistula, Odra and Warta. In the case of standing water bodies, six lakes and artificial reservoirs were studied. In total, the results were presented in 29 publications. The studies related to over 220 compounds, the majority of which were antibiotics. The performed analyses often indicated a lack or trace amounts of PPCPs in surface water (<LOD or <LOQ). Concentrations of some PPCPs were considerably higher, which pointed to a link between sewage discharge from WWTPs and the level of water pollution in surrounding areas. Their quality and the amount of microcontaminants influence other water bodies, for instance, groundwater through surface-water infiltration and aquifer recharge, and seawater, through river inflow to the sea, along with PPCP loads. Based on this, studies of rivers and streams may reflect, to some extent, the overall state of the entire water environment in terms of PPCP contamination.

With regard to antibiotics, 73 substances were analysed in all the considered research, and this formed the most common PPCP group. In most cases, concentrations did not exceed detection or quantification limits. Some studies revealed presence of antibiotics

up to several dozens of ng/L, e.g., lincomycin, sulfadimethoxine, ofloxacin, sulfapyridine, amoxicillin, carbadox and sulfasalazine [71,74,80,97,98]. Some substances occurred in concentrations of several hundreds of ng/L, e.g., sulfathiazole, metronidazole, clindamycin, sulfamethoxazole, azithromycin and ciprofloxacin [72,74,99]. The highest concentration of antibiotics exceeded 1000 ng/L but only in the case of the Vistula River [74]. This applied to antibiotics such as sulfamethoxazole (with the highest concentration, reaching 1770 ng/L), azithromycin, clarithromycin, ceftazidime and erythromycin. According to this review, the main river in Poland seems to be more contaminated with antibiotics compared with other, smaller streams, also tested for the presence of PPCPs [81,97,100,101].

Another group often analysed in surface water has been that of NSAIDs, with 15 substances having been tested to date. Contrary to antibiotics, NSAIDs were found to occur not only in major Polish rivers but also in smaller streams and standing water bodies. For instance, up to 3730.1 ng/L of ibuprofen was found (one of the most popular drugs in this group) [71,86]. NSAIDs in concentrations above 1000 ng/L also included diclofenac and naproxen [44,71,91,102]. The remaining non-steroidal anti-inflammatory drugs were detected in lower concentrations, e.g., acetylsalicylic acid, diflunisal, fenoprofen, ketorolac and indomethacin [43,63,71,75,80,82,87,101–105].

Other PPCP groups were studied less often, and lower concentrations of compounds were observed. However, quantities of several substances in surface water were relatively high, even comparable with treated wastewater; for example, beta-blockers, i.e., bisoprolol, sotalol and metoprolol, were found in concentrations of up to 2190 ng/L [70]. Similar results were reported for painkillers, such as tramadol and metamizole [75,98]. The most popular painkiller, paracetamol, occurred in lower quantities (up to 337.7 ng/L) [86]. This group also included opiates, e.g., codeine and morphine. Interestingly, they were found in smaller streams, but their concentrations were lower compared with other painkillers [71]. In longer rivers, such as the Vistula, other PPCP groups were abundant, e.g., antihypertensives (especially irbesartan, with concentrations of up to 6558.8 ng/L) and diuretics [70,71]. Among anti-epileptics, carbamazepine (794 ng/L) was dominant [81,98]. In the case of antidepressants, desvenlafaxine and venlafaxine stood out from the compounds within this group.

Hormones were detected frequently (9 compounds) [51,71,80,87,105]. Their levels in surface water did not exceed 100 ng/L and ranged from <LOD to 84.15 ng/L (diethylstilbestrol). Similar results were reported for stimulants and fibrates, with the exception of caffeine (found at a high concentration of 29,995.5 ng/L), nicotine and clofibric acid [55,70,71,80]. Regarding alkylphenols, contrast agents and corrosion inhibitors, among these groups, bisphenol A occurred in the highest concentration (3113 ng/L) [63,81,90,99,106]. Among personal care product residues (such as cosmetic and pharmaceutical preservatives, UV filters and repellents), interesting results were found for methylparaben, ethylparaben, propylparaben and DEET [71,80]. Within the remaining PPCP groups, one or a few substances were examined. Their concentrations ranged from <LOD or <LOQ to several hundreds of ng/L, and they were mostly detected in the main Polish rivers, i.e., the Vistula and Warta. These involved groups such as antiarrhythmics, antiretroviral and antidiabetic drugs, hypnotics, PDE5 inhibitors, lipid-modifying agents, sugar substitutes, antiparasitic, antibacterial and antifungal agents, antihistamine and histamine receptor modulators, and immunosuppressive drugs. Examples of contaminants from the above-mentioned groups are fluconazole, darunavir, mycophenolic acid, triclosan and saccharin [74,81]. In smaller streams, substances detected in higher concentrations included fexofenadine and temazepam [71,72,77,80,95,100].

Moreover, in the Polish research we studied, 20 metabolites were taken into account. Most of these were not detected (<LOD) or were found at a low level, except for several compounds, e.g., paraxanthine (caffeine metabolite, with the highest concentration in this PPCP group, reaching 90,665 ng/L), oxazepam (diazepam metabolite), cotinine (nicotine metabolite) and oxypurinol (allopurinol metabolite) [71,81]. For several PPCP groups, the

pharmaceuticals were determined in some Polish rivers. However, all the results were below the detection or quantification limits [70,92,94,98,100,105].

5.5. Seawater

In this review, seawater was analysed separately from other surface water resources due to its specificity and distinct conditions, different from freshwater. Poland has access to the Baltic Sea, and thus all the results concern only this water body. For seawater, low PPCP concentrations are generally assumed because of their dilution in large amounts of water. However, inflowing polluted streams and tourist activity by the seaside in summer may result in the presence of some contaminants in seawater.

Research on seawater confirmed the presence of substances from five PPCP groups (antibiotics, antiepileptics, NSAIDs, painkillers and stimulants). These included 23 compounds many of which were found in concentrations higher than the limit of detection. Research results published in 5 scientific articles referred to determination of PPCPs in the southern Baltic Sea along the Polish border, and the remaining ones concerned only the Gulf of Gdańsk and Bay of Puck in the Gdańsk region. The PPCPs qualified as antibiotics (13 compounds) and NSAIDs (8 compounds) had the greatest representation among the PPCP groups [102,107]. Among these groups, oxolinic acid and ketoprofen dominated (up to 1026 ng/L and 616 ng/L, respectively) [83,108]. Detected substances with lower concentrations from other PPCP groups were paracetamol, carbamazepine and caffeine [83,101].

5.6. Groundwater

Groundwater bodies in Poland were studied less frequently compared with surface water in terms of PPCP occurrence. Apparently, these are associated with a lower risk of significant pollution and a lower possibility of high concentrations of microcontaminants since wastewater is directed mainly into streams and does not usually affect aquifers directly. Even in riverbank filtration sites, along the flow path and where there is migration to shallow aquifers, pollutants undergo various natural processes that lead to self-attenuation. Therefore, levels of PPCPs in groundwater are generally lower than on the surface. Nevertheless, aquifers are an essential source of drinking water in Poland, and thus great emphasis is placed on the protection of groundwater reservoirs. Locally, aquifers may be more vulnerable to pollution, which can be observed in the vicinity of municipal or industrial landfills. In such places, PPCP concentrations may be considerably higher due to constant and large loads discharged to aquifers and specific conditions occurring in the groundwater environment, which often influence the low degradation rate of some contaminants. The problem of groundwater contamination was noticed, as (in 2017) complex screening studies of PPCPs in groundwater were performed for the whole Poland (carried out for 93 sampling points, by the Polish Geological Institute National Research Institute) [28]. Apart from this, the rest of the studies were incidental and limited to a specific location.

The occurrence of PPCPs in Polish groundwater bodies has been investigated in 14 scientific papers to date. Most of the results have been published only recently, which suggests a growing interest in the problem of groundwater contamination with compounds qualified as PPCPs, and more studies may be expected in the future. In the research considered in this review, 122 compounds from 33 PPCP groups were analysed in total, although the vast majority were found at levels below the detection or quantification limits.

So far, antibiotics constitute the most studied PPCP group in Polish research (with 19 compounds). The highest concentration was reported for sulfapyridine (177.1 ng/L) and sulfamethoxazole (66 ng/L) [28,101,109]. The remaining antibiotics occurred in lower quantities [28,73,109]. Most studies on PPCP performed, published in 11 articles, were about NSAIDs. Pharmaceuticals included in these analyses were, among others, diclofenac (with the highest concentrations, reaching 2770 ng/L), ibuprofen, ketoprofen and naproxen [28,50,63,73,99,105,109,110]. NSAID concentrations were higher com-

pared with other medicines used for similar purposes, i.e., painkillers such as paracetamol (up to 113.1 ng/L) [73,81]. Another interesting compound is carbamazepine, found in concentrations of up to 869 ng/L, and this was the most abundant drug among anti-epileptics [28,81,109]. A similar maximum concentration was reported for caffeine (873.3 ng/L) [101].

Hormones were also taken into consideration in research into groundwater quality. Among eight studied substances, the highest concentrations were detected for estrone (up to 309 ng/L), and 17α-ethynyloestradiol (61 ng/L) [28,109,111]. For the other hormones, the results were mostly below or very close to the quantification limit [28,49,73,105,109,110]. Another fairly frequently analysed group is that of beta-blockers but only sotalol and atenolol concentrations exceeded the quantification limits [73,81]. Similar outcomes were revealed for antidepressants, among which only fluvoxamine was detected [28,73,81,95,109,110].

Some personal care products (i.e., cosmetic and pharmaceutical preservatives, UV filters and repellents) were quantified only in north-eastern Poland, in the aquifer extending within municipal landfills [49,50,111]. This was found to mainly apply to DEET, whose concentrations reached 17,280 ng/L. In this region, large quantities were also reported for bisphenol A (an alkylphenol), 3-(4-methylbenzylidene)camphor and methylparaben [49,50,106,111]. The other PPCPs detected in Polish groundwater belong to such groups as antibacterial, antifungal and chemotherapeutic agents, antidiabetic drugs, antihistamines and histamine receptor modulators, antihypertensives, antiischemic agents, contrast agents, corrosion inhibitors, diuretics, sugar substitutes and vasodilators [50,63,73,81]. In the case of compounds qualified as alpha-1 blockers, beta2-adrenergic antagonists, antiarrhythmic drugs, antidementia drugs, antiparasitics, antithrombotics, fibrates and lipid-modifying agents, these were analysed in Polish groundwater, but all the results were below the detection or quantification limits [28,73,81,105,109,110]. Furthermore, PPCP metabolites were taken into account only in several studies. Interestingly, oxypurinol definitely stood out from these substances, as it appeared in concentrations of up to 1350 ng/L [81].

5.7. Drinking Water

In Poland, drinking water is derived from either groundwater or surface water supplies. In the case of both water sources, Polish research confirmed the presence of some PPCPs. Undoubtedly, prior to distribution by waterworks, water intended for consumption is treated at water treatment plants (WTPs) using various treatment technologies. Nonetheless, even the most advanced methods do not always remove PPCP contamination. Hence, some slowly degradable substances may occasionally be found in tap water, although their concentrations are generally at trace levels. In this review, all results for drinking water related to samples taken and analysed after treatment in WTPs.

National research indicated that 27 PPCP groups were analysed in drinking water. These included 95 compounds, some of which had concentrations higher than the limit of detection. The presence of PPCP groups in drinking water in large Polish cities was reported in 10 articles [70,73,74,77,82,95,102,105,106]. The published results mainly referred to research in Warsaw, Gdańsk and Poznań. The most common were antibiotics (27 compounds). Fewer substances represented other PPCP groups: antihypertensives (10 compounds), NSAIDs and beta-blockers (8 compounds each) as well as fibrates and hormones (5 compounds each). The remaining PPCP groups included mostly one or two substances. The maximum concentrations in drinking water investigated in Poland were observed in the case of ibuprofen (up to 224 ng/L), azithromycin (193 ng/L), paracetamol (173 ng/L) and caffeine (159 ng/L). Concentrations of one order of magnitude lower were reported for erythromycin, clarithromycin, telmisartan, valsartan, furosemide, hydrochlorothiazide, levonorgestrel and bisphenol A. In drinking water, there were only trace amounts in the case of compounds from PPCP groups such as beta2-adrenergic agonists, antidepressants, lipid-modifying agents, antiparasitics, chemotherapeutic agents and immunosuppressive drugs.

6. Discussion

This review has shown that PPCP determination in water samples in Poland is performed more and more frequently. Yet, the number of analyses still seems to be relatively low, particularly in the case of landfill leachate and seawater, which makes a results comparison unfeasible. Similarly, it was not possible to assess wastewater or drinking water treatment efficiencies based on the published data as the results tended to depict one-shot and non-time-shifted sampling, and hence a comparison of these results would have been inaccurate. The collected data have only facilitated a general summary of PPCP occurrence (and variations in concentrations) in different types of water samples.

Undoubtedly, the most studied PPCP groups were antibiotics and NSAIDs. Antibiotics appeared in raw wastewater in concentrations reaching several thousands of ng/L. Apart from a few exceptions (e.g., azithromycin and clindamycin), antibiotics were usually found in smaller quantities in wastewater after treatment processes, decreasing mostly by one or two orders of magnitude (Figure 4). A similar pattern can be seen in the case of triclosan, whose antibacterial properties are used in cosmetics and detergents. As mentioned earlier, this cannot be compared in terms of treatment efficacy, but the results revealed that some compounds might be more resistant, so the quantities found in treated wastewater are comparable to those in samples before treatment. However, it should be noted that the efficacy depends on both compound properties and the treatment methods used.

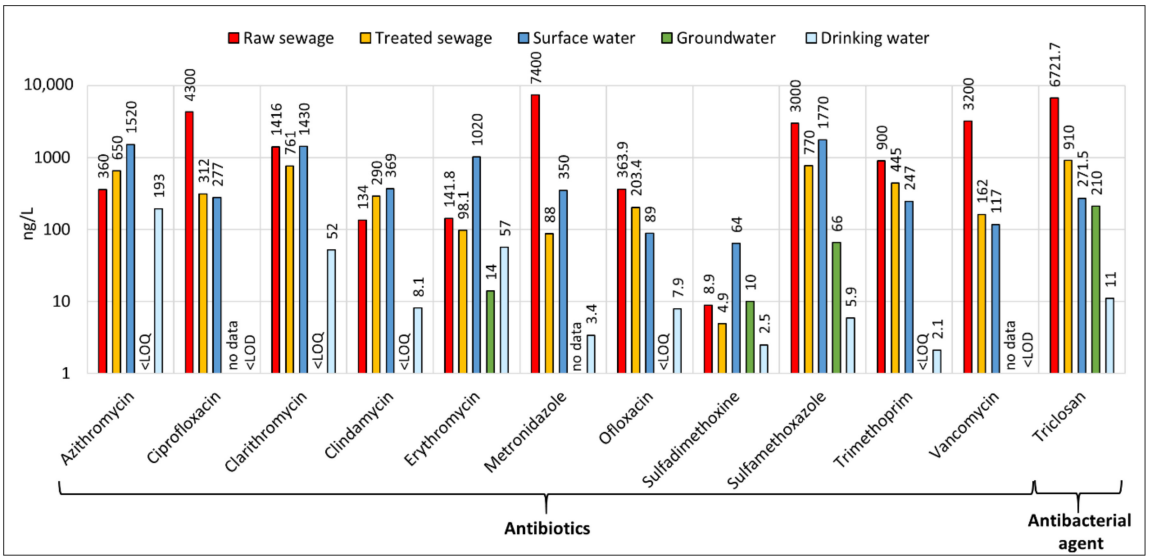

Figure 4. Variations in maximum concentrations of selected antibiotics and one of the antibacterial agents in different water sample types.

Antibiotics were often present in rivers, and their concentrations did not differ much from those in treated wastewater (Figure 4). In some cases, the concentration values were higher than 1000 ng/L and exceeded the highest levels reported for wastewater (e.g., erythromycin and clarithromycin). This may suggest that the problem with antibiotics in water and sewage is even more common, and more studies are required in order to ascertain the extent of contamination from this pharmaceutical group. Such studies could also clarify migration of this group to aquifers, as previous studies of groundwater were only conducted for several locations. According to the gathered data, antibiotics rarely occur in groundwater, and their concentrations are low. These results might be slightly misleading, however, as the presence of antibiotics was confirmed in a few studies of drinking water derived from groundwater supplies. Concentrations reaching dozens or even hundreds of ng/L (e.g., azithromycin, clarithromycin and erythromycin) would suggest that even higher amounts are present in source water.

NSAIDs are not as relevant as antibiotics. However, as representative pharmaceuticals are over-the-counter drugs, they are the medications taken most often. They are very common in the water environment, at high concentrations especially in raw wastewater, sometimes reaching hundreds of thousands of ng/L (e.g., naproxen and ketoprofen). The reported concentrations in wastewater after treatment were significantly lower (Figure 5), with the exception of ibuprofen (the most popular NSAID in Poland), whose maximum concentration in treated wastewater was above 20,000 ng/L. High concentrations of ibuprofen were also observed in surface water, which confirms its widespread use. In contrast, paracetamol, one of the painkillers used for similar purposes (and prescribed as often as ibuprofen), occurred in smaller amounts in both treated wastewater and rivers. This, in turn, may suggest lower resistance to applied treatment methods and processes taking place under natural conditions. NSAIDs and paracetamol are present in groundwater and drinking water, in some cases reaching levels of hundreds of ng/L (Figure 5).

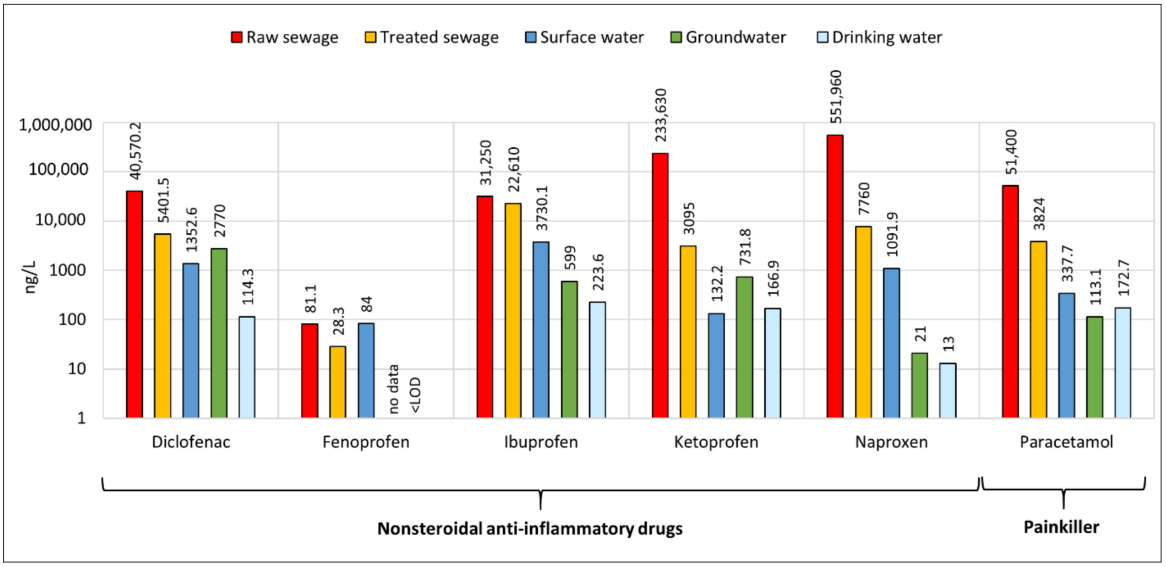

Figure 5. Variations in maximum concentrations of selected NSAIDs and one of the painkillers in different water sample types.

To date, it is clear that less attention has been paid to other PPCP groups. Some of them are important in terms of water contamination due to their dominance among prescription drugs or proven adverse effects on aquatic organisms. One of the most frequently prescribed drug types is that of pharmaceuticals used in the treatment of cardiovascular diseases, e.g., antihypertensives, beta-blockers and fibrates. Within these groups, antihypertensives prevailed, and valsartan was found to be a dominating compound, with concentrations close to 100,000 ng/L in raw wastewater (Figure 6). Valsartan also prevailed in the drinking water samples studied. This has not been found in groundwater so far, but it should be emphasized that only one study focused on this compound in an aquifer within one region. Given its abundance in other water samples, valsartan is also likely to be present in groundwater. For this type of water body, beta-blockers were determined more frequently, but in most cases, their concentrations were very low or below the detection limit. Nevertheless, PPCPs qualified as belonging to the described groups were still present in treated wastewater and surface water. As with antibiotics, the maximum concentration values for some PPCPs in rivers were close to or above their level in wastewater (e.g., irbesartan and metoprolol). This may suggest that the problem associated with their appearance in the water environment is more acute than previously thought.

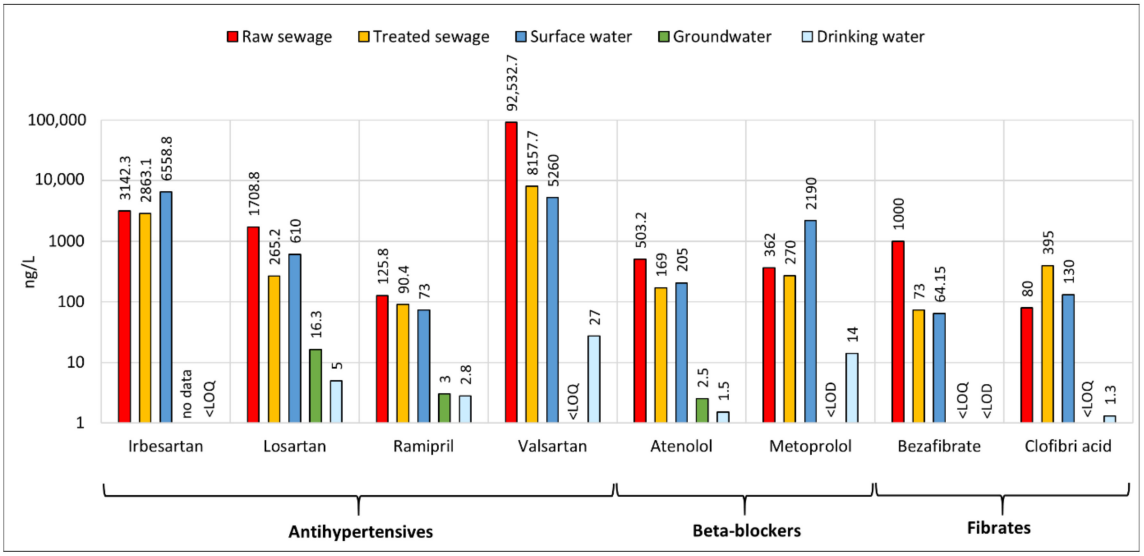

Figure 6. Variations in maximum concentrations of selected antihypertensives, beta-blockers and fibrates in different water sample types.

As mentioned before, hormones and metabolites are practically the only groups whose presence in the environment may be associated with natural processes and reactions. However, the concentrations of compounds representing these two groups vary significantly. Hormones are regarded as endocrine-disrupting chemicals, negatively affecting living organisms. This group was generally observed in low concentrations, reaching 1000 ng/L (except for 17β-oestradiol in one study) (Figure 7). There are scarce data on hormones in drinking water in Poland, but one Polish study confirmed the occurrence of levonorgestrel and progesterone in tap water. Since compounds from this group were observed in drinking water supplies such as surface water and groundwater, it may be assumed that future studies will reveal their presence in tap water taken from other areas in Poland.

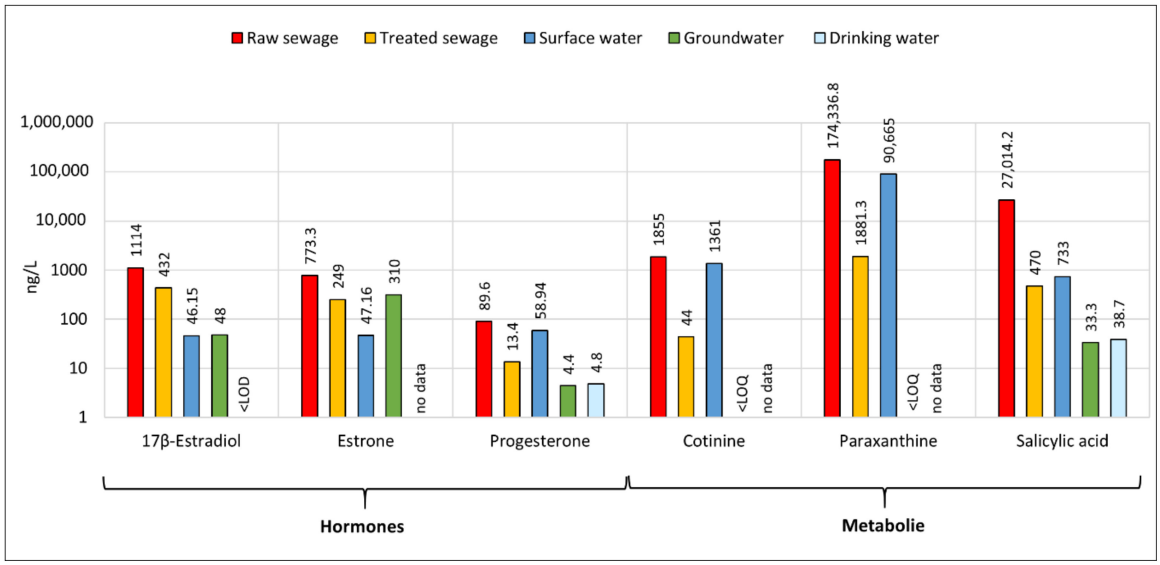

Figure 7. Variations in maximum concentrations of selected hormones and metabolites in different water sample types.

There are potentially countless PPCP metabolite substances. To date, studies have shown that the principal metabolites are those originating from stimulants and OTCs, e.g., metabolites of caffeine (paraxanthine) and nicotine (cotinine) and acetylsalicylic acid,

widely known as aspirin. Apart from caffeine, the parent compounds have rarely been taken into account in the reviewed publications, while their derivatives have been found to reach dozens or hundreds of ng/L (Figure 7). The collected data allow us to assume that relatively high levels of these substances are removed from wastewater. Nonetheless, they seem to be very common in rivers and streams. The probable reason may be that they are derived from products in widespread use (coffee, tea, cigarettes, OTCs, etc.) and are thus released more frequently into water. As with hormones, metabolites were the subject of only a few studies, and their significance currently seems to be underrecognized.

The other PPCPs groups have not been studied as often in Poland. Yet, some individual compounds have been detected in water samples in significant amounts. Examples of such PPCPs are presented in Figure 8. Among the remaining groups, caffeine stands out due to its abundance in all types of water samples. Like its metabolite, paraxanthine, the highest concentrations were observed in raw wastewater and rivers, and lower values were found in treated wastewater. Its prevalence in the environment has been confirmed, although the compound seems to be less resistant to treatment processes in WWTPs. Moreover, caffeine was also detected in relatively high concentrations in groundwater and drinking water samples (Figure 8). Other interesting PPCPs found in the review were bisphenol A, methylparaben and DEET. These stood out in terms of their concentrations in groundwater, exceeding 1000 ng/L. However, it should be emphasized that these results were obtained for aquifers next to municipal landfills. Therefore, the concentrations of these substances in groundwater away from landfills may be lower. Nevertheless, it is interesting to note that bisphenol A, regarded as an EDC, was found in drinking water samples. In turn, no information was found in terms of determination of methylparaben and DEET in drinking water. As these compounds have been found to be present in all other water sample types, migration of these substances to drinking water supplies cannot be excluded. Furthermore, there is still little information on EDCs included in the proposal for the Drinking Water Directive, i.e., bisphenol A, nonylphenol and 17β-oestradiol. In Poland, these have not yet been analysed in drinking water at all, except for one study, which looked at 17β-oestradiol. Thus, more attention should be paid to these compounds in future research.

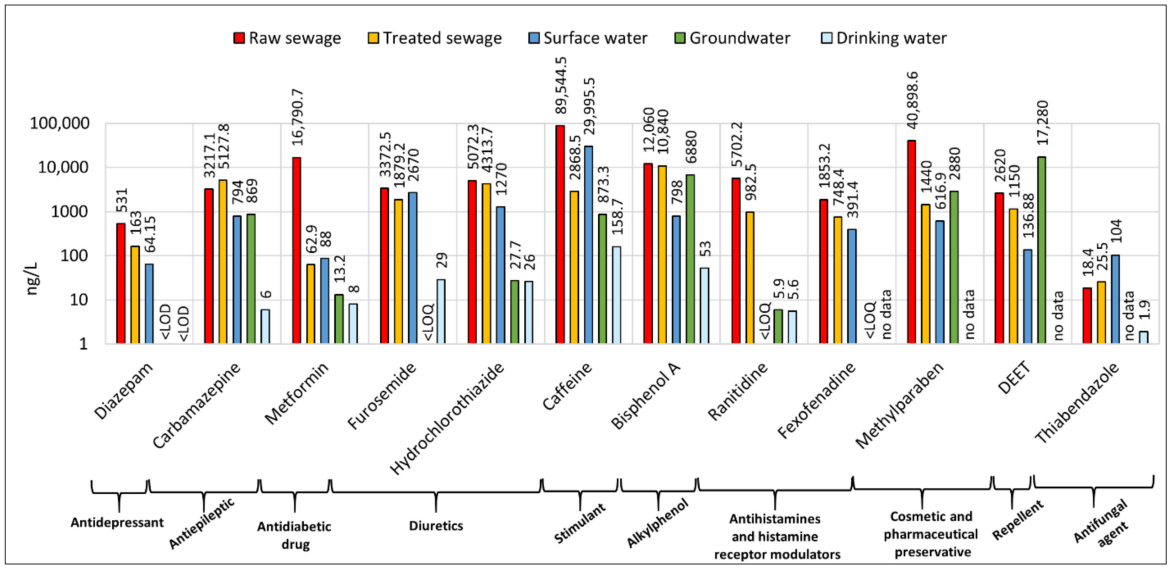

Figure 8. Variations in maximum concentrations of selected PPCPs in different water sample types.

From the other PPCP groups, carbamazepine (an anti-epileptic), one of the most often studied PPCPs in all water sample types, was found in significant concentrations (e.g., in groundwater). Another interesting compound was metformin (an anti-diabetic drug), which was analysed in a few studies, almost all of which found its concentrations to be higher than quantification limits. Finally, two diuretics, furosemide and hydrochloroth-

iazide, were reported in drinking water at concentrations above 20 ng/L, while their quantities in wastewater were moderate compared with other PPCPs (Figure 8).

Due to limited research performed in Poland to date, the collected data enable only a general comparison of approximate concentrations in various water environments. Landfill leachate and seawater have been the subject of fewest studies. Studies have only been conducted within north-eastern Poland (municipal landfills) and along the Polish coastline (the Baltic Sea). Only single studies have focused on these types of water samples, and information on possible PPCP occurrence is minimal (not taken into consideration in the above-described analysis). However, these results may form the basis for further studies.

In landfill leachate, concentrations of studied PPCPs were found to be very high, in some cases even greater than in wastewater. The studies focused mainly on personal care product residues rather than pharmaceuticals. The highest concentrations (above 100,000 ng/L) were reported for DEET, bisphenol A and diclofenac. An interesting fact is that aquifers close to landfills were impacted significantly, manifested in groundwater highly contaminated with certain PPCPs, with concentrations even reaching several thousands of ng/L. This was found for both unsealed and sealed landfills. The review has shown that landfills may constitute a severe threat to groundwater quality in terms of PPCPs. For this reason, more attention should be paid to both landfill leachate and groundwater in the vicinity of dumps in order to more fully understand the possible migration of PPCPs to aquifers.

Interpretation of data on seawater presented even more of a challenge. Due to specific conditions, this was studied separately from other surface water resources in the review. To date, seawater research has focused on the most common PPCP groups, i.e., antibiotics, NSAIDs, and several other pharmaceuticals. The results are very diversified, and PPCP contamination in seawater seems to be a complex issue, with numerous factors influencing the level of microcontaminants concentrations, including dilution, sampling depth, seawater-groundwater exchange, additional PPCP loads from smaller streams, direct sewage discharges to the sea and even tourist activities during the summer season.

Threshold values have yet to be set for PPCPs in drinking water and in water bodies, and it is not possible to compare PPCP concentrations with existing regulations. However, in the recent proposal for a Directive of the European Parliament and of the Council on the quality of water intended for human consumption, the WHO suggested precautionary benchmark values for 17β-oestradiol, nonylphenol and bisphenol A, as aquatic life is sensitive to the effects of estrogenic EDCs. For these substances, the proposed values are 0.001 μg/L (i.e., 1 ng/L), 0.3 μg/L (i.e., 300 ng/L) and 0.01 μg/L (i.e., 10 ng/L), respectively. To date, there is little information about these substances in drinking water in Poland. Nonylphenol was not analysed in either drinking water or source water (surface water and groundwater). Only one study measured 17β-oestradiol in drinking water, and the hormone was not detected. As 17β-oestradiol, bisphenol A was analysed in only one study, which revealed that the benchmark value for this compound was occasionally exceeded. Interestingly, 17β-oestradiol and bisphenol A were found in surface water and groundwater, which may be used for drinking water supplies. In several cases, the concentrations of these substances greatly exceeded the proposed benchmark values. For example, 17β-oestradiol was observed in concentrations of up to 46.15 ng/L in surface water and up to 48 ng/L in groundwater. Concentrations of bisphenol A were even higher, reaching 3113 ng/L and 6880 ng/L in surface water and groundwater, respectively. This confirms the importance of developing of advanced treatment technologies effective against organic microcontaminants in drinking water.

Regardless of the PPCP or water sample type, most studies on the occurrence of pharmaceuticals were conducted only once. Therefore, there is little available knowledge on PPCP variations in water in relation to seasons and existing conditions. These kinds of substances have not been monitored on a regular basis in Poland. Based on the gathered information, it was difficult to identify the trend in terms of water pollution from PPCPs. Extremely high or low concentrations may result from local conditions or temporary

actions. Thus, scarce data can be misleading, and moreover, longitudinal studies should be considered in the future.

7. Conclusions

This review of the current situation in Poland has examined the levels of contamination from pharmaceuticals and personal care products in the water environment, considering different types of water samples: raw and treated wastewater, landfill leachate, surface water, seawater, groundwater and drinking water. All these water environments were studied in terms of the presence of PPCPs, but the scope of analyses in the research studied varied significantly depending on the sample type. Several of the analysed results were single values, providing the only source of information about PPCP concentrations in given types of water samples.

We confirmed that studies have predominantly focused on the most common PPCP groups (antibiotics and NSAIDs). Pharmaceuticals often consumed by humans were found in all types of water samples considered. Although NSAIDs, antibiotics and several other popular compounds dominated in water samples, the presence of less common substances in water cannot be excluded. Further research could confirm the PPCPs prevailing in different parts of the water environment. In particular, more attention should be paid to landfill leachate and seawater. Although few studies focused on samples from these sources, they have confirmed that PPCPs might sometimes occur in significant amounts.

According to the review, the highest amounts of PPCPs were reported for wastewater samples and landfill leachate. The highest concentrations were observed for naproxen in raw wastewater (551,960 ng/L), and bisphenol A in treated wastewater and landfill leachate (10,840 ng/L and 2,202,000 ng/L, respectively). Apart from several exceptions, the quantities of studied substances in natural water (surface water and groundwater) were considerably lower. Maximum concentrations reached 29,995.5 ng/L for caffeine in surface water and 17,280 ng/L for DEET in groundwater. However, in many cases, the reported results did not exceed quantification or detection limits. In seawater, oxolinic acid dominated (up to 1026 ng/L). Some compounds were also analysed and detected in drinking water (e.g., widely used substances such as ibuprofen, paracetamol and caffeine up to 223.6 ng/L, 172.2 ng/L and 158.7 ng/L, respectively). With several exceptions, EDCs were mostly found to be at levels below the benchmark values suggested for three included in the proposal for the EU Drinking Water Directive. It is worth noting that there was no evidence of risks to health posed by PPCPs in drinking water, and these parameters were included in the directive on the basis of the precautionary principle. Nevertheless, as consumption of medications is increasing worldwide, higher concentrations of PPCPs in water might be expected in the future. Development of advanced technologies in both wastewater and drinking water treatment would minimize the risk of natural water being contaminated with PPCPs and would enable future environmental quality standards to be established (and met) for protection of the aquatic environment.

The presence of PPCPs in water is an issue of emerging concern in Poland. The amount of Polish research on PPCPs is limited, and most studies have only been published in the last few years. In general, studies on PPCPs in Poland have been single and incidental. The lack of regular studies and obligatory monitoring prevents us from ascertaining the full scale of the problem. In the case of wastewater and drinking water, limited data on PPCP concentrations at successive stages of treatment preclude accurate assessment of removal efficiencies and thus selection of appropriate technologies. Similarly, it is not feasible to make predictions or draw detailed conclusions about PPCP behaviour in water. Instead of random sampling, future research should be conducted repeatedly to ensure proper understanding of PPCP occurrence in the water environment of Poland.

As currently available data only enable a broad outline to be drawn of the overall degree of water contamination, future research should be carried out systematically, instead of ceasing after one screening. So far, only one complex screening of PPCPs has been conducted covering the whole of Poland (carried out in groundwater by the Polish

Geological Institute National Research Institute). Unfortunately, this did not turn into systematic monitoring. Implementation of the legislation related to EU-wide monitoring and inclusion of pharmaceutical substances would definitely contribute to much-needed development in this field.

Supplementary Materials: The following are available online at https://www.mdpi.com/article/10.3390/w13162283/s1, Table S1: List of PPCPs Determined in Poland and Their Concentrations in Environmental Samples.

Author Contributions: Coordination of the research team, K.Ś. and A.J.W.; resources, K.Ś., S.J.-K. and J.R.; investigation, K.Ś., S.J.-K. and J.R.; visualisation, K.Ś.; writing—original draft preparation, K.Ś., S.J.-K. and J.R.; writing—review and editing, S.J.-K. and A.J.W. All authors have read and agreed to the published version of the manuscript.

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Data Availability Statement: The data presented in this study are available within this article and the Supplementary Materials (Table S1).

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