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High Concentrations of Pharmaceuticals in a Nigerian River Catchment

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Abstract: Pharmaceutical contamination of the environment is recognized as a global problem although most research has focused on Europe and North America to date, and there remains a dearth of information for developing countries, including those in Africa. To address this data gap, the occurrence of 37 pharmaceuticals belonging to 19 therapeutic classes was monitored in surface water and effluents in Lagos State, Southwest Nigeria. Samples were collected quarterly between April 2017 and March 2018 from 22 sites, and 26 compounds were detected at least once, many in the μg/L range. Maximum concentrations for those compounds detected ranged from 75 to 129 μg L^{-1}, and even mean concentrations for 13 compounds were in the order of μg L^{-1}. These values are among the highest ever measured globally. Sewage effluent was more important than drug manufacturing waste in polluting rivers, although there are likely to be numerous unregulated sources of effluent being discharged to rivers that require further study, including urban waste collection areas and vacuum trucks that collect effluent. Seasonal trends in the data were complex, with some compounds being found at higher concentrations in the dry season and, conversely, others being greater during the wet period; this variation potentially relates to the variety of pollution sources in the catchment. Pharmaceuticals are indispensable to human health, although their usage and discharge into the aquatic environment may lead to ecological problems and antibiotic resistance. The data we present indicate that pharmaceutical pollution of freshwaters is a serious issue in Nigeria, and management efforts are needed to improve this problem. *Environ Toxicol Chem* 2022;41:551–558. © 2020 The Authors. *Environmental Toxicology and Chemistry* published by Wiley Periodicals LLC on behalf of SETAC.

Keywords: Pharmaceuticals; Emerging contaminants; Rivers; Water quality; Pollution; Sewage

INTRODUCTION

Pharmaceuticals were first detected in the environment in the 1970s (Tabak and Bunch 1970; Norpoth et al. 1973), and since then numerous studies have quantified their occurrence in aquatic systems. The majority of these studies have been undertaken in Europe and North America (Hughes et al. 2013), with far fewer performed in Africa, South America, and the Middle East (Hughes et al. 2013). The small number of studies that have been done in Africa have detected high frequencies of pharmaceuticals (60–100%; Ngumba et al. 2016), with concentrations typically greater than those measured in the West (Fekadu et al. 2019). In Kenya, concentrations of analytes up to 167 μg L^{-1} were found in sewage effluent and surface waters (K'Oreje et al. 2012, 2016, 2018), and in South Africa pharmaceuticals have also been found to be ubiquitous in effluent and freshwaters at concentrations ranging from ng L^{-1} to μg L^{-1} (Agunbiade and Moodley 2014, 2016; Gumbi et al. 2017). Such high concentrations may be due to a number of factors including high drug usage with poor regulation, the presence of numerous pharmaceutical manufacturing plants, and poorly developed sewage treatment facilities (Fekadu et al. 2019). In Nigeria, analgesics, antibiotics, antacids, antihistamines, anticonvulsants, steroids, antimalarials, and antihypertensives are among the most consumed classes of drugs and are routinely

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purchased without a prescription (Odunsanya 2005). However, the statistics available on the usage of pharmaceuticals are not reliable because of the activities of unregistered pharmacies in some cities such as Lagos (Odunsanya 2005; Nwolisa et al. 2006; Akande and Ologe 2007; Oshikoya and Ojo 2007). The present study aims to add to the limited occurrence data for pharmaceuticals in African effluent and surface water and thus improve our understanding of the global importance of pharamceutical pollution.

We report the results of a monitoring campaign to determine the occurrence of 37 pharmaceuticals in the Odo-Iya Alaro River catchment in Lagos, Nigeria, a country where few pharmaceutical monitoring data are available. The main objectives were: 1) to determine the extent to which drugs belonging to different therapeutic classes are found in effluents and surface water, 2) to quantify spatial and temporal patterns of pharmaceutical contamination, and 3) to highlight particular compounds of environmental concern.

MATERIALS AND METHODS

Substances monitored

The pharmaceuticals monitored (Table 1) were chosen based on their expected presence in surface waters (Burns et al. 2018) because they are high-use drugs that have been found previously in rivers around the world (Hughes et al. 2013), thus enabling us to compare our new information from Lagos with studies undertaken worldwide.

Study catchment

The Odo-Iya Alaro River (Figure 1) forms a subcatchment of the Ogudu River, which discharges into the Lagos lagoon. The river is 15.8 km in length and flows through Ogba, Ikeja, and Maryland, which have a combined population of 2.5 million people. The catchment contains a sewage treatment plant (STP), 2 major pharmaceutical manufacturing plants, and many smaller such plants located in the industrial estates of Ogba and Ikeja; these plants discharge their effluents through drainage pipes and canals into the river. Some of these canals pass through densely populated urban areas that discharge untreated domestic waste to them. Along the river are located mechanical workshops, illegal buildings, and shanty structures with domestic waste discharged untreated into the river; in places like this the river flow is slow. Raw sewage may also enter the river due to emptying of the vacuum trucks that collect untreated effluent in urban areas (Ogunbanwo and Faleti 2018).

Twenty-two sampling stations were selected along the river based on accessibility and the potential to sample both receiving waters and effluents being discharged to them (Supplemental Data, Table S1). Alausa STP is one of the 4 STPs in Lagos State, which has a population of 22 million people.

The treatment plant aerates the wastewater influent by stirring, after which it undergoes sedimentation and chlorination before the final effluent is discharged into the receiving water. The treatment plant was designed to serve a population of 255 000, but there are indications that the plant is handling far more than its installed capacity. The plant has an inflow rate of 1000 $m^3 d^{-1}$, a hydraulic retention time of 18 h, and a sludge retention time of 20 d; both domestic and municipal wastewater are treated at Alausa STP.

Sample collection

Effluent and surface water samples were collected on a quarterly basis to incorporate both the wet (April and July 2017) and dry seasons (October 2017 and January 2018). Amber glass sampling vessels were rinsed with 100% methanol once and deionized water 3 times to remove potential contamination before sampling. Samples were collected at the same time of day and location, checked using a Global Positioning System. At each sampling site, 3 50-mL water samples were collected and then homogenized into a single 150-mL composite sample. A 10-mL aliquot of each composite sample was then filtered on site through a Whatman GFF (0.7-μm pore size) glass microfiber syringe filter into a 20-mL amber glass vial with a Teflon-lined screw cap (Fisher Scientific). Samples were frozen immediately with dry ice before shipping within 24 h to the York Centre of Excellence in Mass Spectrometry, University of York, United Kingdom, for analysis. On arrival (3 d), samples were thawed immediately and analyzed.

Chemical analysis

Quantification of pharmaceutical concentrations was achieved using high-performance liquid chromatography–tandem mass spectrometry (HPLC–MS/MS) with a Thermo Scientific TSQ Endura Mass spectrometer coupled with an UltiMate 3000 liquid chromatograph. The method used was adapted from Furlong et al. (2014) and further validated (Burns et al. 2018). Briefly, prior to starting the quantitative analysis, 500 μL of each water sample was diluted with 495 μL of HPLC-grade water and spiked with 5 μL of a mixture of internal standards (each at a concentration of 80 μg L^{-1}) in glass autosampler vials. The 50% dilution was done to clean the samples and bring analyte concentrations to within the calibrated range. When concentrations were found to still exceed the calibrated range, further dilution and reanalysis was carried out. A random number generator was used to randomize the order in which samples were injected onto the HPLC–MS/MS. Analysis was conducted by direct injection of 100 μL of each sample into a Phenomenex Eclipse Plus C18 chromatography column using a Phenomenex C18 (ODS, Octadecyl) 4- × 3-mm ID guard column. Mobile phase A was HPLC-grade water with 0.01 M formic acid and 0.01 M ammonium formate, and mobile phase B was 100% HPLC-grade methanol. A flow rate of 0.45 mL min^{-1} was used with a gradient starting at 10% B, which then increased to 40% at 5 min, 60% at 10 min, and 100% at 15 min, remained at 100% B until 23 min, and then dropped to 10% at 23 min prior to re-equilibration. The autosampler temperature was kept at 4 °C and the HPLC column compartment at 40 °C. The collision gas was argon at a

TABLE 1: Physicochemical properties of the pharmaceutical compounds monitored in the Odo-Iya Alaro River, Lagos, Nigeria

Therapeutic group	Compound	CAS no.	Log K_{OW}	pKa	Molecular weight ($g\ mol^{-1}$)	Molecular formula	Solubility ($mg\ L^{-1}$)
Analgesic and anti-inflammatory	Codeine	76-57-3	1.19	10.6	299.37	$C_{18}H_{21}NO_3$	9000
	Hydrocodone	125-29-1	2.16	8.23	299.37	$C_{18}H_{21}NO_3$	2.49×10^2
	Paracetamol	103-90-2	0.46	9.38	151.16	$C_8H_9NO_2$	14 000
	Tramadol	27203-92-5	3.01	9.23–13.08	263.37	$C_{16}H_{25}NO_2$	1151
Antacid	Cimetidine	51481-61-9	0.4	6.8	252.34	$C_{10}H_{16}N_6S$	9380
	Ranitidine	66357-35-5	0.27	8.08	314.41	$C_{13}H_{22}N_4O_3S$	24 700
Anti-allergic	Loratadine	79794-75-5	5.2	5	382.89	$C_{22}H_{23}ClN_2O_2$	0.011
Antibiotic	Erythromycin	114-07-8	3.06	8.9	733.94	$C_{37}H_{67}NO_{13}$	4.2
	Sulfamethoxazole	723-46-6	0.89	1.60–5.70	253.28	$C_{10}H_{11}N_3O_3S$	610
	Trimethoprim	738-70-5	0.91	7.12	290.32	$C_{14}H_{18}N_4O_3$	400
Anticonvulsant	Carbamazepine	298-46-4	2.45	13.9	236.27	$C_{15}H_{12}N_2O$	18
	Gabapentin	60142-96-3	–1.1	3.68–10.70	171.24	$C_9H_{17}NO_2$	4.49×10^3
Antidepressant	Amitriptyline	50-48-6	4.92	9.76	277.41	$C_{20}H_{23}N$	9.71
	Desvenlafaxine	93413-62-8	2.72	9.45–10.66	263.37	$C_{16}H_{25}NO_2$	3.7×10^3
	Diltiazem	42399-41-7	2.7	8.06	414.52	$C_{22}H_{26}N_2O_4S$	465
	Oxazepam	604-75-1	2.24	1.55–10.90	286.71	$C_{15}H_{11}ClN_2O_2$	179
	Venlafaxine	93413-69-5	3.2	10.09	277.41	$C_{17}H_{27}NO_2$	267
Antihistamine	Diphenhydramine	58-73-1	3.27	8.98	255.35	$C_{17}H_{21}NO$	3.06×10^3
	Fexofenadine	83799-24-0	2.81	4.28–8.76	501.7	$C_{32}H_{39}NO_4$	2.4×10^{-2}
	Ketotifen	34580-13-7	3.85	8.43	309.43	$C_{19}H_{19}NOS$	15.3
	Cetirizine	83881-51-0	1.7	2.70–3.57	388.89	$C_{21}H_{25}ClN_2O_3$	6.96×10^4
Antidiabetic	Metformin	657-24-9	–2.64	12.4	129.1	$C_4H_{11}N_5$	1.06×10^6
	Sitagliptin	486460-32-6	1.39	8.78	407.31	$C_{16}H_{15}F_6N_5O$	179.2
Antipsychotic	Diazepam	439-14-5	2.82	3.4	284.74	$C_{16}H_{13}ClN_2O$	50
	Temazepam	846-50-4	–0.82 to –0.66	10.68	300.75	$C_{16}H_{13}ClN_2O_2$	164
Antimalaria	Artemisinin	63968-64-9	2.9	4.6	282.33	$C_{15}H_{22}O_5$	49.7
Antiarrhythmic	Lidocaine	137-58-6	2.26	7.86	234.34	$C_{14}H_{22}N_2O$	4.10×10^3
Antiretroviral	Lamivudine	134678-17-4	–9.54	–14.45	229.26	$C_8H_{11}N_3O_3S$	7.0×10^4
Antiviral	Oseltamivir	196618-13-0	0.95	7.7	312.41	$C_{16}H_{28}N_2O_4$	1.6×10^3
Contraceptive	Norethisterone	68-22-4	2.97	–19.29	298.43	$C_{20}H_{26}O_2$	7.04
Beta blocker	Atenolol	29122-68-7	0.16	9.6	266.34	$C_{14}H_{22}N_2O_3$	13.3×10^3
	Propranolol	525-66-6	–0.45	9.42	259.34	$C_{16}H_{21}NO_2$	61.7
SERM	Raloxifene	84449-90-1	6.09	7.99–9.92	473.6	$C_{28}H_{27}NO_4S$	0.56
Diuretic	Triamterene	396-01-0	0.98	3.11–15.88	253.27	$C_{12}H_{11}N_7$	48.2
Calcium channel blocker	Verapamil	52-53-9	2.15–3.79	8.92	454.6	$C_{27}H_{38}N_2O_4$	4.47
SSRI	Sertraline	79617-96-2	4.3	9.47	306.23	$C_{17}H_{17}Cl_2N$	3.5
	Citalopram	59729-33-8	3.74	9.78	324.4	$C_{20}H_{21}FN_2O$	31.09

[a]Sources: Wishart Research Group (2006); National Library of Medicine (2004); and Drugs.com (2001).
Log K_{OW} = octanol/water partition coefficient; pKa = dissociation constant; SERM = selective estrogen receptor modulator; SSRI = selective serotonin reuptake inhibitor.

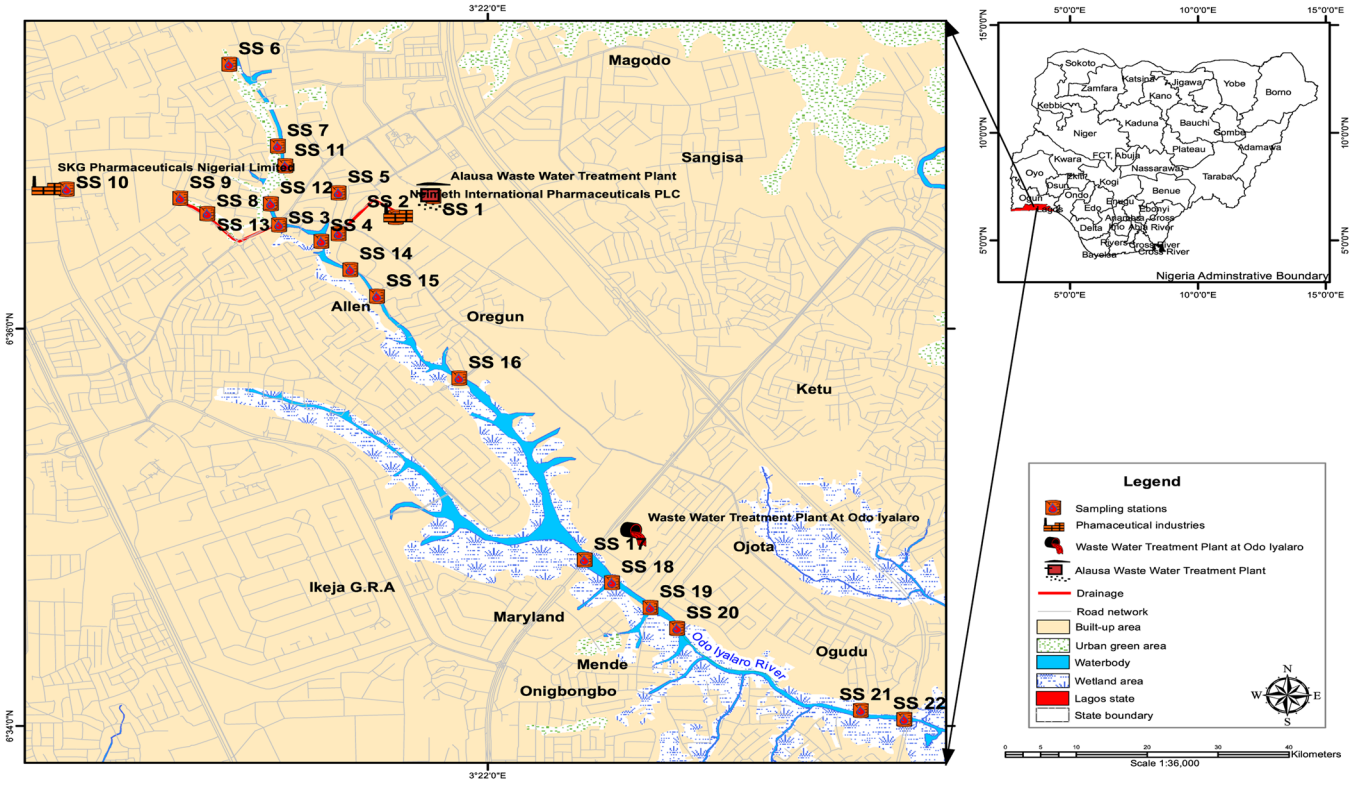

FIGURE 1: The Odo Iya Alaro River in Lagos State, Southwest Nigeria, and the 22 sampling stations used in the present study.

pressure of 2 mTorr. Quantification was done with a 16-point calibration using deuterated internal standards (Burns et al. 2018) ranging from 1 to 32 000 ng L^{-1}, and calibration r^2 values were consistently >0.95.

Analytical limits of detection were calculated as described by Burns et al. (2018) and ranged from 0.9 ng L^{-1} (carbamazepine) to 12.4 ng L^{-1} (gabapentin; Supplemental Data, Table S2). Quality control measures were used throughout the analysis; briefly, method blanks ($n = 6$) were made with an identical collection procedure as the environmental samples although HPLC-grade water was used. Concentrations of the target pharmaceuticals were consistently below levels of analytical quantification in the method blanks. In addition, quality controls consisting of all target pharmaceuticals at a concentration of 80 ng L^{-1} were injected after every 4 samples followed by an instrumental blank consisting of pure HPLC-grade water. Analytical tolerance was consistently within ±15%, and the instrumental blanks did not contain detectable residues of the target analytes.

Data analysis

Data were organized using Excel, and residuals of the data were checked for normal distribution using the Shapiro–Wilk normality test; homogeneity of variance was checked using the Bartlett test of homogeneity of variances. The R program (R Development Core Team 2008) was used to analyze the data; the general linear model (GLM) and Chi-square were used to determine whether there were differences between the sampling sites. Seasonal variations were analyzed using one-way analysis of variance where assumptions of normality and homogeneity were met followed by Tukey's post hoc tests to detect whether there was any variation in concentrations between the wet and dry seasons.

RESULTS

Detection frequencies

All the study compounds were detected, although the frequency of detection varied greatly for different substances (Figure 2). Some, including carbamazepine, fexofenadine, and paracetamol, were present in sewage effluent and surface waters most of the time, whereas others, such as diltiazem, propranolol, and venlafaxine, were rarely detected. Detection frequencies in pharmaceutical manufacturing effluent were significantly lower than in sewage effluent and river water (GLM: χ^2 (3) = 883.3, $p < 0.001$).

Mean and maximum concentrations

Peak pharmaceutical concentrations were in the range of hundreds of µg/L, whereas mean concentrations were several orders of magnitude lower (Figure 3). The antibiotic sulfamethoxazole was detected at the highest concentration, 129 µg L^{-1}, and paracetamol was measured at 111 µg L^{-1}. Paracetamol and sulfamethoxazole also had the highest mean concentrations of 18 and 11 µg L^{-1}, respectively. Cimetidine had the third highest maximum concentration (96 µg L^{-1}), and a

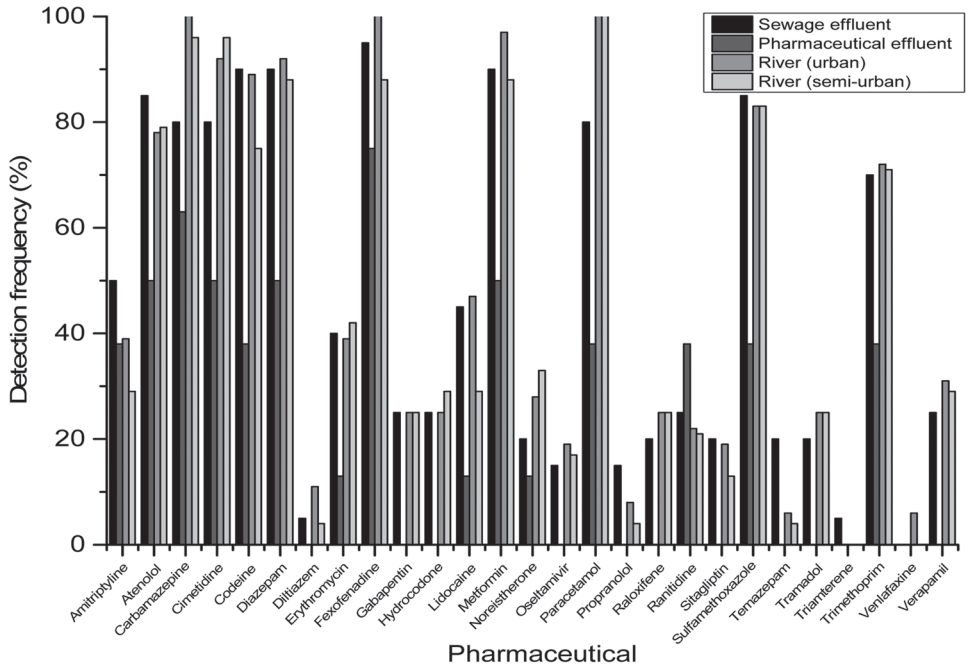

FIGURE 2: Detection frequencies for pharmaceuticals measured in sewage effluent, pharmaceutical manufacturing effluents, and rivers (urban and semi-urban).

mean concentration of 11 μg L^{-1}. The maximum concentration of a further 7 analytes (fexofenadine, carbamazepine, metformin, diazepam, atenolol, trimethoprim, and codeine) also exceeded 39 μg L^{-1}. Mean concentrations for these substances were; metformin (10 μg L^{-1}), fexofenadine (8 μg L^{-1}), carbamazepine (8 μg L^{-1}), atenolol (3 μg L^{-1}), diazepam (3 μg L^{-1}), trimethoprim (2 μg L^{-1}), and codeine (2 μg L^{-1}; Supplemental Data, Tables S3 and S5).

There was a significant difference in pharmaceutical concentrations in the different matrices sampled (GLM: $\chi^2(3) = 883.32$, $p < 0.001$).

When pharmaceuticals were detected in manufacturing effluent, concentrations were higher than in sewage effluent and river water. Drugs were diluted after sewage effluent had entered urban rivers, and concentrations were lower still in semi-urban reaches. Although detection frequencies were higher in the wet season, concentrations were higher in the dry season (Figure 4).

DISCUSSION

Pharmaceuticals are biologically active and pseudo-persistent in the environment due to the continual input of wastewater effluent to rivers (Hughes et al. 2013; Kay et al. 2017; Burns et al. 2018), and they therefore pose a potential toxicological risk to nontarget organisms (Boxall et al. 2002; Huang et al. 2012). Monitoring has mainly taken place in

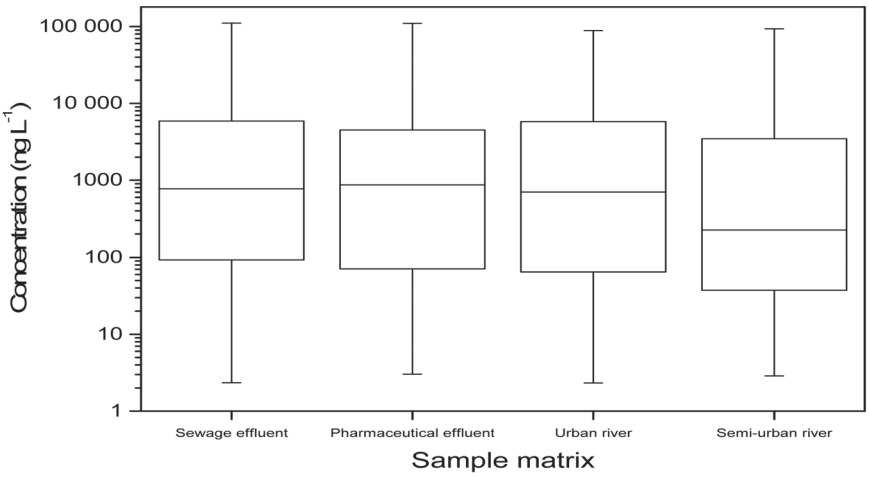

FIGURE 3: Pharmaceutical concentrations measured in the Odo Iya Alaro river catchment, Lagos, Nigeria. Boxes represent median and 25th and 75th percentiles, and whiskers show minimum and maximum values.

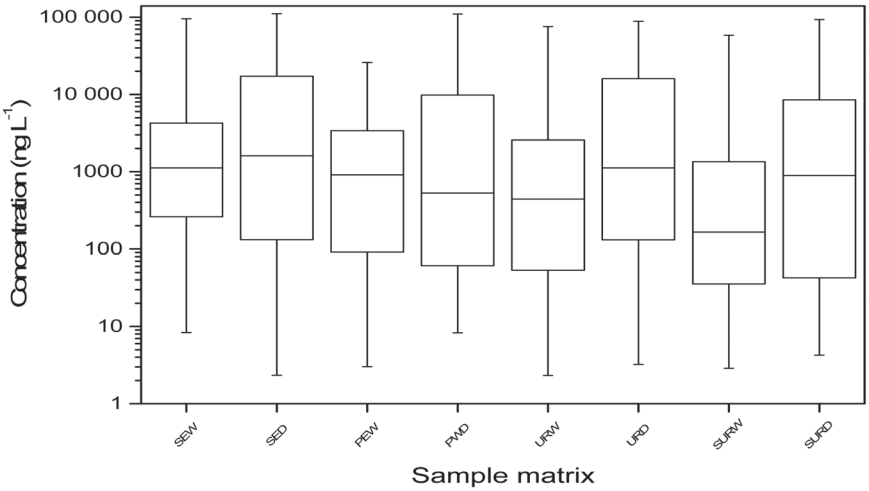

FIGURE 4: Comparison of pharmaceutical concentrations measured in the wet and dry seasons in different matrices in the Odo Iya Alaro river, Lagos, Nigeria. SEW = sewage effluent in wet season; SED = sewage effluent in dry season; PEW = pharmaceutical manufacturing effluent in wet season; PWD = pharmaceutical manufacturing effluent in dry season; URW = urban river in wet season; URD = urban river in dry season; SURW = suburban river in wet season; SURD = suburban river in dry season. Boxes represent median and 25th and 75th percentiles, and whiskers show minimum and maximum values.

Europe and North America, and data from Africa are sparse. The present study addresses this important research gap by presenting new information about the presence of pharmaceuticals in pharmaceutical manufacturing effluent, sewage effluent, and surface water in Lagos, Nigeria, a country that has been studied very little.

The detection of 26 pharmaceuticals in the Odo-Iya Alaro river confirmed the presence of these substances in Nigeria watercourses including some that have not previously been observed in African rivers. These were from a wide range of therapeutic classes including anti-inflammatories (codeine), antidepressants (amitriptyline), and antihistamines (fexofenadine), and were present at relatively high concentrations compared with some other drugs (propranolol and venlafaxine) that were measured. Pseudo-persistence was observed, presumably due to continuous discharge of effluents to the river, similar to that found in other studies.

One of the key findings of the present study is that pharmaceutical concentrations in the environment are often 2 to 3 orders of magnitude higher than typically reported in Europe and the United States, where most monitoring has been undertaken (Verlicchi et al. 2012; Hughes et al. 2013; aus der Beek et al. 2016; Madikizela et al. 2017; Burns et al. 2018). We report some of the highest pharmaceutical concentrations ever found in rivers globally (Supplemental Data, Table S4), with measured levels not uncommonly in the 10s of μg/L, ranging up to 129 $\mu g\,L^{-1}$. This is in line with findings in Kenya (K'Oreje et al. 2012, 2016, 2018). Particular compounds of concern in the Nigerian environment appear to be carbamazepine, cimetidine, fexofenadine, metformin, paracetamol, and sulfamethoxazole. The presence of a range of substances at such high concentrations may be attributed to a number of factors including over-the-counter sales, differences in health issues, poorer removal efficiencies at wastewater treatment plants (WWTPs), unregulated discharges by pharmaceutical manufacturing companies, and illegal disposal of sewage by vacuum trucks. Other studies have proposed that these sources are likely to be important in Africa (Fekadu et al. 2019), and further study is needed to disentangle the inputs from these various sources.

We observed few spatial trends in pharmaceutical pollution, which appears to be ubiquitous, with the absence of many compounds in pharmaceutical production effluent suggesting that sewage effluent is the main source of pollution. Indeed, the occurrence and concentrations of pharmaceuticals in WWTP effluent and surface water were very similar. This highlights the fact that receiving waters have little capacity to dilute effluent but also that further unregulated and unmonitored sources of effluent may be discharging to the river, such as vacuum trucks collecting effluent in urban areas. However, a study from India has proposed that pharmaceutical production facilities are a key source of pharmaceutical pollution in developing countries (Balakrishna et al. 2017).

Season had an impact on pharmaceutical pollution. Some compounds were found at extremely high concentrations in the dry season, and concentrations of other compounds were relatively high during the wet season. Previous studies have proposed a range of reasons for variation across the year, including seasonal usage and changes in environmental conditions (e.g., temperature and river flow; Tewari et al. 2013; Kolpin et al. 2014; Fekadu et al. 2019). It may be that the multiple sources of pharmaceuticals in the catchment result in the complexity of spatial patterns found in the present study, with some continuous effluent discharges being diluted in the wet season, but other sources (e.g., urban waste sites) mobilizing pollutants in periods of rainfall.

CONCLUSIONS

Our study is the most detailed to date on pharmaceutical pollution in African river catchments and it has highlighted some of the highest concentrations ever found globally

(Supplemental Data, Table S4). Concentrations in Nigeria rivers appear to be several orders of magnitude higher than those reported for Europe and the United States and, in some cases, even higher than the few existing concentration values produced for other developing countries. Sewage effluent appears to be the key source of pollution, although further investigation of unregulated sources is needed. It also appears that many compounds are not discharged from drug manufacturing plants. We propose that the complexity of temporal patterns across seasons is due to a greater range of sources contributing to pharmaceutical loads than in many existing studies, which poses a particular research challenge for understanding and managing pharmaceutical pollution in African rivers. It is very likely that the scenario we present is the same in other major African cities as well as megacities in other developing nations globally, where pharmaceuticals are available over the counter and where wastewater discharges to rivers proceed without regulation. A key implication for the global research agenda on pharmaceutical occurrence, fate, and effects is that studies should focus more on developing countries where contamination of water is likely to be most significant because of inadequate facilities.

Supplemental Data—The Supplemental Data are available on the Wiley Online Library at https://doi.org/10.1002/etc.4879.

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Data Availability Statement—Data, associated metadata, and calculation tools are available from the corresponding author (ogunbanwo.o@mylaspotech.edu.ng).

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