Occurrence of Pharmaceutical Residues and Antibiotic-Resistant Bacteria in Water and Sediments from Major Reservoirs (Owabi and Barekese Dams) in Ghana

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The presence of pharmaceuticals in the environment is undesirable since their biological activity may impair ecosystem health of reservoirs that receive inflows from other water sources. This work determined the concentrations of analgesics and antibiotics, and the occurrence of antimicrobial resistance among microbes in water and sediment samples from Owabi and Barekese reservoirs—two main sources of pipe-borne water in the Kumasi metropolis in Ghana. The study also assessed the knowledge, attitude, and practice of inhabitants near these reservoirs regarding the disposal of unused and expired medicines. Out of nine targeted pharmaceuticals, four were detected in at least one sample. Five analytes (caffeine, ciprofloxacin, doxycycline, ibuprofen, and metronidazole) were below detection limit for all samples. The levels of pharmaceuticals were low, as expected, ranging from 0.06 to 36.51 μg/L in the water samples and 3.34–4.80 μg/kg in sediments. The highest detected concentration of any pharmaceutical in water was for diclofenac (107.87 μg/L), followed by metronidazole (22.23 μg/L), amoxicillin (1.86 μg/L), chloramphenicol (0.85 μg/L), and paracetamol (0.16 μg/L). Chloramphenicol recorded the highest concentration (10.22 μg/kg) in the sediments. Five bacteria isolates (Enterobacter, Clostridium, Pseudomonas, Acinetobacter, and Klebsiella) from the samples were resistant to all the antibiotics tested. Isolates of Corynebacterium and Listeria showed susceptibility to only doxycycline. Isolates of Bacillus were susceptible to only two antibiotics (erythromycin and doxycycline). All the 100 respondents interviewed admitted that they dispose of medications once they do not need them. Of those who disposed of unwanted medicines, 79% did so inappropriately. Disposal in household trash (67%) was the most common method used. Majority of respondents felt the need for a facility or program to collect unused medicines (77%), hence their willingness to pay to reduce pollution by pharmaceuticals in the environment. It is quite clear from the ecotoxicological risk assessment that a single pharmaceutical at very low level as those in this study and other works is likely to pose many ecological risks upon long-term exposure and therefore cannot be ignored.

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1. Introduction

Pharmaceuticals are used primarily to prevent or treat human and animal diseases. In recent times, there has been an increase in the number of reports detailing the presence of human pharmaceuticals in water in various parts of the globe [1, 2]. In a 2002 study by the United States Geological Survey, about 80% of streams sampled in 30 American states contained trace levels of various pharmaceutical products, including hormones, steroids, and analgesics [3]. Following this report, numerous others have since been published. Even at very low concentrations, the presence of pharmaceuticals in water has raised concerns among stakeholders such as drinking-water regulators, governments, water suppliers, and the public regarding the potential risks to human health due to exposure to trace levels of pharmaceuticals via drinking water [4]. Pharmaceuticals could enter the soil and aquatic environments through wastewater effluents, landfill leachates, industrial effluents, aquaculture, and animal feedlots [5–7]. They could also enter the environment through excretion and egestion of unmetabolized drugs into wastewater collection systems and improper disposal of expired and unused medicines [8].

Pharmaceutical products are generally designed to elicit specific responses from precise biological targets. Therefore, these pharmaceutical products’ effects on the environment are undesirable because their behavior in such matrices has not been studied in enough detail. The long-term effects of exposing humans, animals, plants, and other organisms in the ecosystem to pharmaceuticals remain unknown and can only be speculated to a large extent [9–11]. The presence of antibiotics in the environment could lead to an increase in the phenomenon of antimicrobial resistance. Resistance genes can develop in the environment if antibiotic residues are present; these genes can then be transferred to pathogenic bacteria. Resistance genes can be exchanged between environmental bacteria and clinical isolates [12]. Borquaye and coworkers detected amoxicillin, penicillin, and metronidazole at varying levels in leachate and soil samples from landfill sites in Kumasi, Ghana. Twenty-five bacteria belonging to nine genera were also isolated from leachates and soil samples in the study. Some isolated bacteria were resistant to benzylpenicillin, ampicillin, and amoxicillin. The results obtained from the study suggest that pharmaceutical residues are present in the environment and could have deleterious effects.

The Barekese and Owabi dams provide over 90% of potable water to domestic homes in the Ashanti region of Ghana. These reservoirs receive water inflow from various tributaries: Owabi, Sukobri, Akyeampomene, Pumpunase, and Afu for the Owabi reservoir [13] and Offin River for the Barekese reservoir [14]. Several dumpsites and landfill sites are located in and around the tributaries that feed the Owabi and Barekese reservoirs. Since the water treatment strategy for the management of these reservoirs does not necessarily remove chemical contaminants, water dispatched to various homes from these reservoirs could potentially contain pharmaceutical residues. Some water bottling companies also use water from these reservoirs as their raw materials to produce bottled and sachet water. The threat to the public could be substantial. There is, therefore, a need to ascertain the levels of pharmaceutical residues in these reservoirs that provide water to the Kumasi metropolis and examine the risk associated with their presence in the environment. In this work, the occurrence of antibiotics, analgesics, and antibiotic-resistant bacteria in water and sediment samples from the Owabi and Barekese dams was evaluated. The ecological risk associated with the presence of these pharmaceuticals in both water and soil samples was also investigated. A general survey on waste disposal patterns by residents in communities around the Owabi and Barekese reservoirs was also conducted.

2. Methods

2.1. Sample Collection. The Barekese and Owabi dams are in the Atwima district in the Ashanti region of Ghana. The Barekese dam, which was constructed between 1967 and 1972, stretches on latitude 6°44’N and longitude 142°W. The dam’s catchment area measures 909 km² with a total volume of about 35,000,000 m³ [14]. The Owabi dam is smaller than the Barekese dam and was constructed between 1928 and 1932. The reservoir surrounded by a forest reserve covers an area of about 69 km² with an approximate volume of 26,000,000 m³ [15]. A total of 28 water and sediment samples were collected from various points in the Owabi and Barekese dams. Pre-cleaned plastic bottles (1.5L) were rinsed with the water to be collected before samples were fetched into the bottles. For sediment samples, graduated hollow wooden sticks were immersed into the bottom of the reservoir and then withdrawn. Sediment samples collected were then placed in precleaned bottles. All samples were placed in an ice chest and returned to the laboratory for analyses. Maps of the study areas are shown in Figure 1.

2.2. Materials. All analgesics and antibiotics used for analytical and microbiological work were obtained as pure powders from Ernest Chemists (Accra, Ghana). HPLC-grade chemicals were used throughout the study. Ultrapure deionized water (18.2 MΩ) was used to prepare all solutions and calibration standards for HPLC analysis. All the solutions and samples were passed through 0.2–0.6 μm polypropylene filters before HPLC analysis.

2.3. Physicochemical Characterization. An aliquot (10 mL) of water sample was put into a clean beaker, and their pH, total dissolved solids (TDS), and electrical conductivity were measured using a Mettler Toledo (Switzerland) pH meter and a multiparametric probe (WTW, Weilheim, Germany). For soil samples, 10% (w/v) of dried pulverized sample to distilled water was prepared for the analysis [17, 18]. The pH meter was calibrated with buffers 4.01 and 7.01 before use.

2.4. Sample Preparation and Analysis. Water samples were filtered (0.45 μm pore size filters) to remove solid particles. Sample extraction and pre-concentration were done by solid-phase extraction (SPE). An HLB cartridge (6 mL, 200 mg, Green Mall, Jiangsu, China) was used in SPE analyses. SPE columns were conditioned using 5 mL distilled
water followed by 5 mL methanol. Water samples (1.5 L) were passed through the preconditioned 6 mL HLB SPE cartridges for the concentration of analytes. The SPE cartridge was then washed with 5 mL of distilled water. In the case of sediments, 100 g of an air-dried, pulverized sample was weighed into a beaker, and three successive extractions were done by sonication with 150 mL of acetonitrile. The supernatants were combined, filtered through 0.45 μm nylon membrane filters (Whatman, UK), and subjected to SPE as was done for water samples. To elute analgesics, 2 mL of methanol and 0.1% acetic acid mixture (1 : 1 v/v) was used. For antibiotics, 2 mL methanol and 0.05% trifluoroacetic acid mixture (1 : 4 v/v) was used. One milliliter of each eluate was then used in HPLC analyses.

2.5. HPLC Analyses

2.5.1. Antibiotics. Chromatographic separation was done on a Perkin Elmer Flexar HPLC coupled with a PDA detector. Separation of antibiotics was achieved on the Agilent Zorbax 300SB C18 column (250 × 4.6 mm, 5 μm). The mobile phase consisted of 0.05% trifluoroacetic acid (TFA, solvent A) and methanol (solvent B). Gradient elution was performed. Details of the gradient program are shown in Table S1. A flow rate of 1 mL/min was employed with an injection volume of 20 μL. Peaks for amoxicillin, chloramphenicol, ciprofloxacin, doxycycline, and metronidazole were monitored at 215 nm. The total runtime was 18 minutes, as shown in Figure S1A. HPLC analysis was done at ambient temperature.

2.5.2. Analgesics. Chromatographic separation of analgesics was done on the same Perkin Elmer Flexar HPLC coupled with a PDA detector. Separation of analgesics was achieved on a Phenomenex Luna C8 (150 × 4.6 mm, 5 μm) column. The mobile phase consisted of 0.1% acetic acid (A) and methanol (B). A gradient elution profile was utilized. Details of the gradient program are shown in Table S1. As with antibiotic analysis, a flow rate of 1 mL/min was employed with an injection volume of 20 μL. Peaks for paracetamol, caffeine, and diclofenac were acquired at 270 nm and that of ibuprofen at 220 nm (Figures S1B and S1C). The total runtime for analgesic analysis was 21 minutes, and all analyses were done at ambient temperature.

2.5.3. Standard Calibration and Method Validation. Five concentrations (2.5–200 μg/mL) of each standard were prepared and analyzed by HPLC, and a calibration curve was generated. This calibration curve was used to quantify analytes present in the samples. Linearity for all test samples was tested in the concentration range of 2.5–200 μg/mL. Limits of detection (LOD) and limits of quantification...
(LOQ) for each of the standard drug were computed from a peak signal-to-noise ratio of 1:3.

2.5.4. Recoveries and Quality Assurance. A sample blank (water and soil matrices containing none of the analytes) was prepared and spiked with known concentrations of standard drugs. After the extraction and SPE clean-up procedures (as described earlier), the sample blank was analyzed by HPLC methods described earlier. The concentration of the drugs was determined from the chromatograms obtained. Standard solutions of the analytes were injected prior to analyses and also after every 10 sample runs to ensure that the HPLC system was functioning properly. Blank samples were also injected after every 5 runs to monitor any sample interference. All sample injections were made in duplicates. Each batch of analyses was prepared to include a reagent blank to check background contamination. Table S2 shows the recovery data.

2.6. Ecological Risk Assessment. The potential effects of identified pharmaceuticals on the aquatic environment were studied through ecological risk assessment. Ecological risk assessment is generally expressed with respect to hazard quotients (HQ). The predicted no-effect concentration (PNEC: the concentration of pharmaceutical at which no adverse effect is suspected to occur) values for fish, Daphnia, and algae were determined using data from the literature on acute toxicity. The PNEC is calculated by multiplying the EC₅₀ values by an arbitrary safety factor, typically 1000 [9, 19]. The HQ was then determined by the ratio of MEC (maximum measured environmental concentration in μg/L) to PNEC, as shown in

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HQ = \frac{MEC}{PNEC}. \tag{1}
\]

The risk ranking criteria of [20] were used in this study. According to this criterion, an HQ < 0.1 implies a minimum risk to the organisms, 0.1 ≤ HQ ≤ 1 implies medium risk, and HQ ≥ 1 implies high risk [20].

2.7. Isolation and Identification of Bacteria Strains. Bacteria were isolated from water and sediment samples collected from the Barekese and Owabi reservoirs according to the method of [21]. Ten grams of each sediment sample was aseptically transferred into a sterile self-sealed bag and homogenized in 90 mL of sterile maximum recovery diluent using a pulsifier (PUL 100E; Microgen Bioproducts, UK) for 2 min. Ten-fold serial dilutions of both water and sediment samples were prepared up to 10⁻⁵. An aliquot of 1 mL of each dilution was transferred into molten plate count agar (PCA) and swirled to ensure an even distribution of cells before allowing it to solidify. Inoculated media were inverted and kept in an incubator for 24 hours. A series of sub-culturing from the primary culture resulted in pure cultures of isolates. Bacterial isolates were identified based on colony characteristics and cellular morphology [22]. Identified isolates were stored in 20% glycerol broth and at -20°C, pending further analyses.

2.8. Microbial Susceptibility Assays. The susceptibility of the isolated microbes against selected antibiotics was tested in the agar disc diffusion assays. A nutrient agar media was prepared by dissolving 28 g of nutrient agar in 1 L of distilled water. The mixture was autoclaved for 40 minutes at 121°C and then cooled. Afterward, the prepared agar was poured into Petri dishes and dried. Pure cultures of isolated microbes were suspended in saline to obtain a density of 0.5 McFarland turbidity standard using the direct colony suspension. Forty microliters of the standardized colony suspension was transferred into the Petri dish and spread evenly over the entire agar surface using sterile cotton swabs. Afterward, filter paper discs (about 6 mm in diameter) containing the standard antibiotics (ampicillin, amoxicillin, cefuroxime, ciprofloxacin, doxycycline, erythromycin, gentamycin, metronidazole, and chloramphenicol) were placed on the agar surface. The filter paper disc for each standard drug was prepared using the criteria of the European Committee on Antimicrobial Susceptibility Testing (EUCAST). After the disc application on the agar plates, the plates were incubated at 37°C for 18 hours. The inhibition of growth zones was measured in millimeters. The zone diameter of each drug was interpreted using the criteria published by the EUCAST [23].

2.9. Survey on Disposal of Pharmaceuticals. Even though manufacturer packaging usually recommends disposal of unused and expired drugs by returning to the pharmacy, studies elsewhere have revealed that disposal via the sink or toilet or in regular household waste is the practice and is a potential source of human pharmaceuticals in the environment. A survey of about 100 households in and around the dams was conducted to investigate the households' disposal routes of unused and expired pharmaceuticals as a source of pharmaceutical compounds in the environment. The survey evaluated the public's level of awareness on the presence of pharmaceutical products in rivers and drinking water and knowledge on the possible impact of the presence of these pharmaceutical products in water samples. In-depth interviews were conducted with interviewers using standardized and pretested questionnaires. Ten eligible volunteers from various households were used to pretest the instrument for readability, understanding, question design, and length. The questionnaire generated sociodemographic data, their habits regarding the disposal of expired or unused medications, and their understanding of the environmental effect of medications. The questionnaires were administered to 100 household participants. Participants could either complete the survey themselves or have their responses marked by the researchers. The data was coded and input into Microsoft Excel 2016. The completeness, consistency, and reliability of the data were all examined. When respondents did not finish a question, it was considered a missing value, and only complete answers were utilized to analyze the results. Statistical analysis was performed using IBM SPSS Statistics for Windows, version 24 (IBM Corp., Armonk, N.Y., USA).
3. Results

3.1. Physicochemical Characterization. Physicochemical data of water and sediment samples used in this study are presented in Table 1. Generally, water samples obtained from the Owabi reservoir were slightly acidic with a pH range of 6.48–6.85, whereas the sediment samples ranged from slightly acidic to slightly basic (pH range of 6.24–8.93). For the samples from Barekese, all water and sediment samples were slightly acidic (pH range of 6.32–6.83). The total dissolved solids (TDS) of water and sediment samples from Owabi ranged between 90 and 94 mg/L and 26 and 58 mg/L, respectively. For samples from the Barekese reservoir, TDS ranged from 122 to 124 mg/L and 111 to 167 mg/L for water and sediment samples, respectively. The mean TDS for water from Owabi was generally lower than that of Barekese. Electrical conductivity (EC) of sediment samples from Owabi was determined to be in the range of 8.13–59.3 μS/cm while EC for water samples from the same Owabi reservoir ranged between 98.5 and 102.2 μS/cm. For the sediment samples from Barekese, the range of EC values was 124.1–239.0 μS/cm, whereas the EC of water samples from Barekese ranged between 135.9 and 138.9 μS/cm. It was observed that the mean value of EC of water samples from Barekese was greater than the mean EC recorded for Owabi samples. A similar observation was made for sediment samples from the two reservoirs.

3.2. Occurrence of Antibiotics and Analgesics. In the HPLC analysis, method linearity was evaluated in the concentration range of 2.5–200 μg/mL for both analgesics and antibiotics. A good correlation was observed for all the antibiotics and analgesics analyzed, with $R^2$ values between 0.98 and 0.99 recorded. For all analytes, the limits of detection (LOD) and limits of quantification (LOQ) were determined to range between 0.031 and 0.116 μg/mL and 0.103 and 0.386 μg/mL, respectively. The LOD and LOQ were estimated based on signal-to-noise ratio of 1:3. The recoveries for the various analytes were high and ranged from 85.5% to 97.0% (Table 2). The elution of all analytes occurred within 19 minutes.

Of the pharmaceuticals analyzed in water samples from Barekese, chloramphenicol was the most prevalent, with a 55.56% frequency of detection. The frequency of detection for amoxicillin, diclofenac, metronidazole, and paracetamol was all 11.1% (Table 2). Caffeine, ibuprofen, ciprofloxacin, and doxycycline were not detected in any of the water samples from Barekese. The maximum concentration of chloramphenicol in water samples from Barekese was 0.85 μg/mL, whereas the maximum levels of amoxicillin, diclofenac, metronidazole, and paracetamol were 1.86, 4.36, 22.23, and 0.16 μg/mL, respectively. Caffeine, ibuprofen, doxycycline, and ciprofloxacin were not detected in water samples from Barekese. The average concentration of chloramphenicol in sediment from Barekese was 4.80 ± 3.63 μg/kg, and this was the only analyte detected in sediment samples from Barekese.

Results for the pharmaceuticals residues identified in samples from Owabi are also presented in Table 2. For the Owabi reservoir, chloramphenicol was the most frequently detected analyte, present in 77.8% of all water samples analyzed. Amoxicillin, diclofenac, and paracetamol, each at 33.3%, were detected in multiple water samples. The average concentration of amoxicillin, chloramphenicol, diclofenac, and paracetamol was 0.24 ± 0.14, 0.38 ± 0.12, 36.51 ± 68.80, and 0.06 ± 0.08 μg/mL, respectively. Interestingly, the analytes that were not detected in Barekese water samples, namely, caffeine, ibuprofen, doxycycline, and ciprofloxacin, were undetected in water samples from Owabi. Paracetamol was the only analyte detected in sediment samples from Owabi, with a 66.67% detection frequency and an average concentration of 3.32 ± 4.55 μg/kg.

3.3. Ecological Risk Assessment. Three of the most widely used organisms in monitoring the impact of pharmaceuticals on the environment were adopted for the ecological risk assessment. The ecological risk assessment was carried out by first establishing the maximum estimated concentration for the various pharmaceuticals from the study. The corresponding predicted no-effect concentration for each analyte on each organism was obtained from the literature [9, 24–26]. The hazard quotients were computed, and the risk was established using a standard criterion [20]. The results are presented in Table 3 (water samples) and Table 4 (sediment samples). Generally, the pharmaceutical residues recorded in this study posed medium to high risk to all the three organisms investigated. The ecological risk posed by diclofenac to all organisms investigated in water was classified as high risk at both Owabi and Barekese, since the RQ was greater than 1. The levels of chloramphenicol and metronidazole in water samples from Barekese also posed high risk to all the organisms. Both paracetamol and amoxicillin posed high risk to Daphnia and algae, but medium risk to fish. Amoxicillin and paracetamol, however, presented a low risk to fish in water. For water samples from Owabi, amoxicillin posed a medium risk to all organisms, whereas the levels of paracetamol and chloramphenicol posed high risks to algae but a medium risk to fish. Two analytes were detected for sediment samples from Barekese and Owabi—chloramphenicol and paracetamol. Both analytes posed high risks to all organisms investigated, with RQ’s far over 10 recorded in each case (Table 4).

3.4. Microorganism Isolation and Susceptibility Testing. A total of 30 pure bacteria isolates were obtained from water and soil samples in both reservoirs. Pure bacterial isolates were obtained by a series of subculturing from the primary cultures. Bacterial isolates were identified based on colony characteristics and cellular morphology. From the Gram staining reaction, 17 of the isolates were Gram-positive while 13 were Gram-negative. All isolates were rod-like in shape. Isolates were identified to belong to one of the 8 genera: *Bacillus*, *Corynebacterium*, *Pseudomonas*, *Klebsiella*, *Enterobacter*, *Listeria*, *Actinobacter*, and *Clostridium* (Table 5). Table 6 shows the results of the antimicrobial susceptibility testing. Antibiotic susceptibility testing (AST) was performed according to EUCAST recommendations. Bacteria isolates were tested against nine antibiotics belonging to
School Leaving Certificate levels of formal education, with most attaining either Middle Level education (29.0%). However, 23% had no formal education and have lived in the community for less than ten years. Figure 2 provides the demographic characteristics of the respondents. The majority of the respondents indicated that they disposed of both orthodox and herbal medications once they did not need them (Table S3). Antibiotic and analgesic medications, either prescription or nonprescription medicines, were the most frequently disposed of in the community. Penicillin (23%), cephalosporin (cefuroxime), macrolides (erythromycin), nitroimidazoles (metronidazole), amphenicols (chloramphenicol), and tetracycline (doxycycline), aminoglycosides (gentamicin) demonstrated susceptibility to only doxycycline. Isolates of Bacillus demonstrated susceptibility to only two antibiotics (doxycycline and erythromycin) of different classes (tetracycline and macrolides).

3.5. Survey on Disposal Routes. One hundred (100) respondents participated in this study, of which 64.0% were males and the remaining 36% were females. The participants were mostly between 21 and 50 years old (79%) and had various levels of formal education, with most attaining either Middle School Leaving Certificate (JHS) (36.0%) or a Secondary Level education (29.0%). However, 23% had no formal education and have lived in the community for less than ten years. Figure 2 provides the demographic characteristics of the respondents. The majority of the respondents indicated that they disposed of both orthodox and herbal medications once they did not need them (Table S3). Antibiotic and analgesic medications, either prescription or nonprescription medicines, were the most frequently disposed of in the community. Penicillin (23%), cephalosporin (cefuroxime), macrolides (erythromycin), nitroimidazoles (metronidazole), amphenicols (chloramphenicol), and tetracycline (doxycycline), aminoglycosides (gentamicin) demonstrated susceptibility to only doxycycline. Isolates of Bacillus demonstrated susceptibility to only two antibiotics (doxycycline and erythromycin) of different classes (tetracycline and macrolides).

### Table 1: Physicochemical parameters of water and sediment samples from the Barekese and Owabi reservoirs.

| Samples          | pH Range | Mean ± SD | EC (μS/cm) Range | Mean ± SD | TDS (mg/L) Range | Mean ± SD |
|------------------|----------|-----------|------------------|-----------|------------------|-----------|
| Barekese, water  | 6.59–6.79| 6.68 ± 0.07| 135.9–138.9      | 137.10 ± 0.93| 122–124          | 123.00 ± 0.71|
| Barekese, sediment| 6.32–6.83| 6.60 ± 0.23| 124.1–239.0      | 175.42 ± 44.81| 111–167          | 154.60 ± 39.49|
| Owabi, water     | 6.48–6.85| 6.68 ± 0.11| 98.5–102.2       | 32.48 ± 25.67| 90–94            | 91.78 ± 1.48  |
| Owabi, sediment  | 6.24–8.93| 7.60 ± 1.35| 8.13–59.3        | 99.38 ± 1.67 | 26–58            | 46.33 ± 17.67 |

### Table 2: Occurrence of pharmaceutical residues in water and sediment samples from Barekese.

| Reservoir | Sample type | Pharmaceutical | Range      | Mean ± SD | Frequency of detection (%) |
|-----------|-------------|----------------|------------|-----------|---------------------------|
| Barekese  | Water (μg/L)| Amoxicillin    | 0.08–1.86  | 1.86 ± 0.00| 11.11                     |
|           |             | Chloramphenicol| 0.10–0.85  | 0.42 ± 0.29| 55.56                     |
|           |             | Diclofenac     | 0.03–4.36  | 1.36 ± 0.00| 11.11                     |
|           |             | Metronidazole  | 0.06–22.23 | 22.23 ± 0.00| 11.11                    |
|           |             | Paracetamol    | 0.06–0.16  | 0.16 ± 0.00| 11.11                     |
|           | Sediment (μg/kg)| Chloramphenicol| 2.78–10.22| 4.80 ± 3.63 | 80.00                     |
|           |             | Amoxicillin    | 0.10–0.37  | 0.24 ± 0.13| 33.3                      |
|           |             | Chloramphenicol| 0.18–0.53  | 0.38 ± 0.12| 77.8                      |
| Owabi     | Water (μg/L)| Diclofenac     | 0.69–107.89| 36.51 ± 68.80| 33.3                     |
|           |             | Paracetamol    | 0.01–0.15  | 0.06 ± 0.08| 33.3                      |
|           | Sediment (μg/kg)| Paracetamol| 0.10–6.54  | 3.34 ± 4.55 | 66.67                     |

Frequency of detection was computed by evaluating the ratio of the number of samples in which an analyte was detected to the total number of samples analyzed.

In the survey, the respondents were also asked if they were aware that they could return their unused or expired medicines to the pharmacy for disposal. To flush medication to the pharmacy would be inconvenient. Respondents were asked whether they would be willing to pay to improve sewage treatments so that river and stream pollution by pharmaceuticals could be reduced, and 77% responded “yes” (Table S6).

4. Discussion

In the survey to gauge the disposal preferences of the inhabitants around the Owabi and Barekese reservoirs, a majority
(67%) of household respondents indicated that they disposed of expired or unused medicines by placing them into trash cans to be disposed of in landfills. Even though some respondents indicated periodic disposal of unwanted and expired medicines, 58% indicated that disposal was done whenever they did not need the medication. Prescription or nonprescription analgesics and antibiotics were commonly reported as unused and needing disposal. Pharmaceutical products constitute a significant component of waste generated from many homes. Additionally, some community pharmacies dispose of unused pharmaceuticals into municipal waste [27]. Analgesics and antibiotics have been reported to be frequently disposed of in large quantities by many households [28]. In literature, it has been noted that unwanted or expired medicines are most commonly dumped into trash cans, followed by subsequent disposal.

Table 3: Ecological risk assessment of water samples from Barekese and Owabi reservoirs.

| Pharmaceutical | MEC (μg/L) | PNEC (μg/L) | HQ | Comment |
|----------------|------------|-------------|----|---------|
| Barekese       |            |             |    |         |
| Amoxicillin    | 1.86       | Daphnia (1.1000) | 1.69 | High risk |
|                |            | Algae (1.0000)   | 1.86 | High risk |
|                |            | Fish (0.5400)    | 1.57 | High risk |
| Chloramphenicol| 0.85       | Daphnia (0.5430) | 1.57 | High risk |
|                |            | Algae (0.1850)   | 4.59 | High risk |
|                |            | Fish (0.5320)    | 8.20 | High risk |
|                |            | Daphnia (0.0220) | 198.18 | High risk |
|                |            | Algae (0.0145)   | 300.69 | High risk |
|                |            | Fish (1.0350)    | 21.48 | High risk |
| Metronidazole  | 22.23      | Daphnia (0.1000) | 222.3 | High risk |
|                |            | Algae (0.1250)   | 177.84 | High risk |
|                |            | Fish (0.3780)    | 0.42 | Medium risk |
| Paracetamol    | 0.16       | Daphnia (0.0092) | 17.39 | High risk |
|                |            | Algae (0.1340)   | 1.20 | High risk |
|                |            | Fish (2.5000)    | 0.15 | Medium risk |
| Amoxicillin    | 0.37       | Daphnia (1.1000) | 0.34 | Medium risk |
|                |            | Algae (1.0000)   | 0.37 | Medium risk |
|                |            | Fish (0.5400)    | 0.98 | Medium risk |
| Chloramphenicol| 0.53       | Daphnia (0.5430) | 0.98 | Medium risk |
|                |            | Algae (0.1850)   | 2.86 | High risk |
|                |            | Fish (0.5320)    | 202.80 | High risk |
|                |            | Daphnia (0.0220) | 4904.09 | High risk |
| Owabi          |            | Algae (0.0145)   | 7440.69 | High risk |
|                |            | Fish (0.3780)    | 0.40 | Medium risk |
|                |            | Daphnia (0.0092) | 16.30 | High risk |
|                |            | Algae (0.1340)   | 1.12 | High risk |

RQ < 0.1 implies minimum risk, 0.1 ≤ RQ ≤ 1 implies medium risk, and RQ > 1 implies high risk [20]. MEC: maximum estimated concentration; PNEC: predicted no-effect concentration; HQ: hazard quotient.

Table 4: Ecological risk assessment of sediment samples from Barekese and Owabi reservoirs.

| Pharmaceutical | MEC (μg/kg) | PNEC (μg/kg) | HQ | Comment |
|----------------|-------------|--------------|----|---------|
| Barekese       |             |              |    |         |
| Chloramphenicol| 10.22       | Daphnia (0.5430) | 18.82 | High risk |
|                |             | Algae (0.1850)  | 55.24 | High risk |
|                |             | Fish (0.3780)  | 17.30 | High risk |
| Owabi          |             | Daphnia (0.0092) | 710.87 | High risk |
| Paracetamol    | 6.54        | Daphnia (0.1340) | 48.81 | High risk |

RQ < 0.1 implies minimum risk, 0.1 ≤ RQ ≤ 1 implies medium risk, and RQ > 1 implies high risk [20]. MEC: maximum estimated concentration; PNEC: predicted no-effect concentration; HQ: hazard quotient.
into dumpsites or landfills [20, 28]. The pharmaceuticals could be washed off into nearby surface waters during rainfalls or even leach into the ground to contaminate groundwater. This is no exception in communities where open defecation and indiscriminate garbage disposal are practiced; all these waste flow directly into nearby rivers and water bodies. Some of these water bodies feed the reservoirs of water treatment plants where the water is treated and dispatched to homes and factories for various uses, including drinking. Because water treatment plants do not necessarily target the removal of specific chemicals, there is the likelihood that the chemicals may be present in treated water dispatched to homes. The threat is potentially more significant for communities that consume water directly from polluted rivers and streams. Exposure to sublethal concentrations of these pharmaceuticals could harm the ecosystem and result in long-term toxic effects. The effects could be multidrug resistance (by antibiotics), interference with a drug a person might already be taking (usually by analgesics), gender binding in fish, and decline in biodiversity [11, 29, 30]. Although this study did not assess how respondents came by unwanted or expired medication, patient noncompliance, relief of symptoms/signs, medication change, and death of patient have been reported by other studies as sources of unwanted or expired medications [31–33].

The findings of this survey clearly show that respondents do not dispose of unwanted or expired medicines through proper means. Interestingly, 50% of respondents were aware that pharmaceuticals’ pollution of rivers and streams was an environmental concern. Contrastively, respondents were more supportive towards greater care of the environment. The fact that 78% of respondents ranked health as extremely important while 48% ranked environment as slightly important suggests a lack of knowledge of the possible effects of

### Table 5: Colony morphology, cellular characteristics of isolates, and bacteria identified from water and sediment samples from Barekese and Owabi reservoirs.

| Isolates | Colony morphology | Shape | Margin | Elevation | Opacity | Cellular characteristics | Gram reaction | Shape | Microorganism |
|----------|------------------|-------|--------|-----------|---------|--------------------------|--------------|-------|--------------|
| OW1      | Cream            | Circular | Undulate | Umbonate | Opaque | Negative | Rod | Klebsiella sp. |
| OW2      | Grey             | Irregular | Entire | Convex | Opaque | Positive | Rod | Corynebacterium sp. |
| OW3      | Grey             | Irregular | Undulate | Flat | Opaque | Negative | Rod | Bacillus sp. |
| OW4      | Grey             | Circular | Entire | Raised | Translucent | Negative | Rod | Pseudomonas sp. |
| OW5      | Grey             | Circular | Entire | Flat | Translucent | Positive | Rod | Listeria sp. |
| OS1      | Grey             | Irregular | Undulate | Flat | Opaque | Positive | Rod | Bacillus sp. |
| OS2      | Unpigmented     | Circular | Mucoi | Raised | Opaque | Negative | Rod | Acinetobacter sp. |
| OS3      | Cream            | Irregular | Undulate | Flat | Translucent | Positive | Rod | Clostridium sp. |
| OS4      | Grey             | Irregular | Undulate | Flat | Opaque | Positive | Rod | Bacillus sp. |
| OS5      | Grey             | Circular | Entire | Raised | Translucent | Negative | Rod | Enterobacter sp. |
| OS6      | Grey             | Circular | Entire | Raised | Translucent | Negative | Rod | Pseudomonas sp. |
| OS7      | Grey             | Irregular | Undulate | Flat | Opaque | Positive | Rod | Bacillus sp. |
| OS8      | Cream            | Irregular | Undulate | Umbonate | Opaque | Negative | Rod | Klebsiella sp. |
| OS9      | Grey             | Irregular | Undulate | Flat | Opaque | Positive | Rod | Bacillus sp. |
| OS10     | Grey             | Circular | Entire | Flat | Translucent | Positive | Rod | Listeria sp. |
| OS11     | Grey             | Circular | Entire | Raised | Translucent | Negative | Rod | Pseudomonas sp. |
| OS12     | Cream            | Irregular | Undulate | Flat | Translucent | Positive | Rod | Clostridium sp. |
| OS13     | Grey             | Irregular | Entire | Convex | Opaque | Positive | Rod | Corynebacterium |
| BW1      | Grey             | Circular | Entire | Raised | Translucent | Negative | Rod | Enterobacter |
| BW2      | Unpigmented     | Circular | Mucoid | Raised | Opaque | Negative | Rod | Acinetobacter sp. |
| BW3      | Grey             | Irregular | Undulate | Flat | Opaque | Positive | Rod | Bacillus sp. |
| BW4      | Circular         | Undulate | Umbonate | | | | | Rod | Klebsiella sp. |
| BW5      | Grey             | Circular | Entire | Raised | Translucent | Negative | Rod | Enterobacter sp. |
| BW6      | Grey             | Circular | Entire | Flat | Translucent | Positive | Rod | Listeria sp. |
| BW7      | Grey             | Circular | Entire | Raised | Translucent | Negative | Rod | Pseudomonas sp. |
| BS1      | White            | Irregular | Entire | Convex | Opaque | Positive | Rod | Corynebacterium sp. |
| BS2      | Grey             | Circular | Entire | Raised | Translucent | Negative | Rod | Enterobacter sp. |
| BS3      | Grey             | Irregular | Undulate | Flat | Translucent | Positive | Rod | Clostridium sp. |
| BS4      | Grey             | Irregular | Undulate | Flat | Opaque | Positive | Rod | Bacillus sp. |
| BS5      | Grey             | Irregular | Undulate | Flat | Opaque | Positive | Rod | Bacillus sp. |

OW: Owabi water; OS: Owabi sediment; BW: Barekese water; BS: Barekese sediment.
environmental problems on human health and ecosystems. There was a good correlation between respondents’ awareness of the threats that pharmaceuticals pose to the environment and their willingness to pay to improve sewage treatment to minimize pollution of rivers and streams by pharmaceuticals. One strategy that has been suggested to reduce the levels of pharmaceuticals in the environment is the institution and implementation of a medicine take-back program. A search through the literature has indicated that imprudent disposal of unwanted and expired medicines is a major route through which pharmaceuticals access the environment [33–35]. There is no coordinated national outlet for the proper disposal of unused and expired drugs in Ghana. The Food and Drugs Authority (FDA) in Ghana, by the Public Health Act, 202 (Act 851), is responsible for the safe disposal of unwanted and expired drugs [36]. However, the cost and bureaucracy involved in this process are gridlocks for households looking for a safe means to dispose

Table 6: Zones of inhibition (mm) of antibiotics against isolates and corresponding susceptibility and resistance of isolates to antibiotic.

| Organisms      | AMP | AML | CXM | CIP | DOX | E  | GEN | MET | CHL |
|----------------|-----|-----|-----|-----|-----|----|-----|-----|-----|
| Bacillus sp.   | R (0)| R (0)| R (0)| R (17)| S (25)| S (25)| R (10)| R (15)| R (0) |
| Clostridium sp.| R (8)| R (12)| R (7)| R (11)| R (0)| R (18)| R (16)| R (13)| R (11) |
| Klebsiella sp. | R (0)| R (0)| R (7)| 10 (R)| R (0)| R (0)| R (15)| R (0)| R (10) |
| Corynebacterium sp. | R (0)| R (0)| R (0)| R (15)| S (24)| IP| R (0)| R (10)| R (0) |
| Enterobacter sp. | R (0)| R (0)| R (0)| R (16)| R (0)| R (0)| R (0)| R (0)| R (0) |
| Acinetobacter sp. | R (0)| R (0)| R (0)| R (9)| R (0)| R (0)| R (0)| R (0)| R (0) |
| Bacillus sp.   | R (0)| R (0)| R (0)| R (15)| S (24)| S (25)| R (0)| R (0)| R (0) |
| Listeria sp.   | R (0)| R (0)| R (0)| R (16)| S (22)| R (0)| R (0)| R (10)| R (0) |
| Pseudomonas sp.| R (0)| R (0)| R (0)| R (12)| R (0)| R (0)| IP| R (0)| R (0) |
| Clostridium sp.| R (17)| R (18)| R (0)| R (0)| R (19)| R (8)| R (8)| R (11)| R (10) |
| Enterobacter sp.| R (0)| R (0)| R (0)| R (12)| R (0)| R (0)| R (0)| R (0)| R (0) |

AMP: ampicillin; AML: amoxicillin; CXM: cefuroxime; CIP: ciprofloxacin; DOX: doxycycline; E: erythromycin; GEN: gentamicin; MET: metronidazole; CHL: chloramphenicol; S: susceptible; R: resistant; IP: in preparation (limited information in EUCAST database).

Figure 2: Demographic characteristics of respondents in household: (a) gender, (b) age, and (c) educational level of participants in the survey.
of pharmaceutical waste. In 2009, the Cocoa Clinic in Accra introduced a Disposal of Unused Medicines Program (DUMP). This comprehensive medicine take-back project urges patients and staff of its parent organization to return their unused pharmaceuticals to the clinic for safe disposal. Under the FDA’s supervision, the approved methods for controlled disposal of pharmaceutical waste are subsequently followed by the Cocoa Clinic for any drugs received [27, 37]. Even though this is an excellent program that can reduce the environmental presence of pharmaceuticals in Ghana, DUMP has a limited reach. The program serves only a small percentage of the Ghanaian population—personnel of the Ghana Cocoa Board and customers of the Cocoa Clinic. The larger section of the Ghanaian population still has unused and expired drugs in their possession, with the disposal of these drugs still uncontrolled. Establishing an effective medicine take-back program demands a lot of time, money, and human resources. Therefore, willingness to pay to minimize pollution by pharmaceuticals is a good indicator for a national roll-out of a Disposal of Unused Medicines Project (DUMP), as is done in countries such as New Zealand and Canada [35, 38]. A convenient location of the take-back program not only ensures environmental protection but provides details of unused pharmaceuticals valuable to the health community in optimizing its prescribing and dispensing practices to reduce the generation of unused or expired medicines [27]. The existence of a knowledge gap is therefore apparent about the above issues. These could form part of research questions for further studies as they were not explored in this study. It should be noted that the response from the survey may have been distorted as a result of the topic as a self-administered questionnaire collected data. For instance, questions regarding disposal practices, the environment, and payment to improve the environment may have been inflated to a socially desirable factor. Again, a response rate was not calculated in this study due to the fact that the number of participants approached for participation was not tracked. However, the findings of this study point clearly to the existence of improper medicine disposal practices in Ghana, which need to be addressed. This is particularly worrying as the disposal pattern identified as the primary route has been reported in other studies in Ghana [27, 39, 40].

In this study, paracetamol diclofenac, amoxicillin, metronidazole, and chloramphenicol were detected and quantified. Although most pharmaceuticals have high solubility in water and hence remain soluble in the aqueous phase, some drugs remain insoluble or attached to suspended solids in the wastewater. Since the liquid samples were filtered before extraction, the reported concentrations in this work represent only the water-soluble fraction of the analytes. The presence of pharmaceuticals in water and sediments suggests the widespread occurrence of the pharmaceuticals in both reservoirs. Paracetamol and chloramphenicol were present in all sample types. For sediment samples, only paracetamol was detected in samples from Owabi. On the other hand, only chloramphenicol was detected in sediment samples from Barekese. Individual pharmaceutical concentrations in untreated water were found to be generally low, with average concentrations ranging between 0.07 and 36.51 mg/L. The highest detected concentration of any pharmaceutical in untreated water was for diclofenac (107.87 μg/L), followed by metronidazole (22.23 μg/L), amoxicillin (1.86 μg/L), chloramphenicol (0.85 μg/L), and paracetamol (0.16 μg/L). The results of this research are in line with many other studies in the literature, which have observed low levels of pharmaceuticals in a variety of water sources. For instance, [41] detected five pharmaceuticals in influents of a municipal wastewater plant. Paracetamol, metformin, norfluroxetine, atenolol, and cephalaxin were the five detected pharmaceuticals at average concentrations in a range of 1–40 μg/L. In Spain, 35 pharmaceuticals were found in the raw water at a drinking water treatment facility’s intake, with levels up to 1.2 μg/L [42]. However, the levels of pharmaceuticals detected in this study were found to be higher than many other studies. In a study of 31 pharmaceuticals conducted throughout the drinking water supply system in Lisbon, the levels of pharmaceuticals detected ranged from 0.005 to 46 ng/L [43]. In Spain, pharmaceuticals were quantified in water and sediment samples from low ng/L to 168 ng/L and low ng/L to 50.3 ng/g, respectively [44]. Also, Ref. [45] quantified pharmaceuticals ranging from 0.0018 to 3 ng/L in water samples from a drinking water treatment plant. This suggests relatively higher inputs of pharmaceuticals into both reservoirs under study.

The efficiency at which pharmaceuticals are removed from water is affected by several factors, including the weather, the nature of water treatment, and its operating conditions (temperature, redox conditions, solids, and hydraulic retaining time). The physicochemical composition of most pharmaceuticals, which is acidity and high solubility in water with very low solid-liquid partition, is claimed to be the key cause. These factors often result in poor sorption of these compounds onto particles or organic matter, leaving them soluble in the aqueous phase [41]. Overall, pharmaceutical levels in the aquatic environment can be connected to specific physicochemical parameters. Physicochemical factors such as pH, temperature, conductivity, total organic carbon (TOC), and TDS can favor photodegradation and other attenuation processes [44]. The pH of all the raw water samples from both reservoirs was determined to be slightly acidic (6.32–6.89). Positively charged soil components (at pH 6) play a role in acidic pharmaceutical retention. Also, there is considerable blockage of sorption sites by humic acids at acidic pH [44]. Since the detected pharmaceuticals in this study were all acidic, the low levels of detection could be linked to the occurrence of this phenomenon. Conductivity measures the ability of the samples to conduct electricity as a result of the total effect of positive and negative ions dissolved in them. Pharmaceuticals have been found to have a stronger affinity for cationic organic matter from clay soils than anionic organic matter from sandy soils and complexation with cationic species (e.g., Ca²⁺) [46]. The high conductivity values in this study could result in one of these two given possible phenomena leading to the low quantification levels in water samples.

The concentrations of individual pharmaceuticals in the sediments were found to have an average concentration
range of 3.34–4.80 μg/kg. The highest concentrations detected in sediment samples were 10.22 μg/kg for chloramphenicol and 6.54 μg/kg for paracetamol. This study’s findings were consistent with many of the reported works in literature where low levels of pharmaceuticals have been quantified in sediments. Pharmaceuticals have been reported to be present in sediments at the outlet of a wastewater treatment plant in South Africa. Paracetamol, metronidazole, and clozapine were quantified, with concentrations as high as 18 ng/g [47]. In another study in Budapest (Hungary), naproxen and diclofenac were found in the range of 2–20 and 5–38 ng/g, respectively [48]. Although most pharmaceuticals are polar and likely to be found in the dissolved fraction, some can have specialized interactions with the solid fraction and transmitted to sediments [44]. The effects of pH on the interactions between organic matter and pH-dependent pharmaceuticals might be significant [46]. Sediment samples from Owabi were determined to be slightly acidic to basic with a pH range of 6.24–8.93. One of the five pharmaceuticals was detected in sediments from this reservoir. This is in line with the findings of [44] where pharmaceuticals were found to bond to suspended solids under basic (pH 7–8) conditions. However, only chloramphenicol was detected in sediments from the Barekese reservoir. The slightly acidic nature of these samples could be a possible explanation for the absence of the other pharmaceutical from the sediment samples.

Many pharmaceuticals can potentially pose long-term negative impacts on ecosystems and humans. The quantities of pharmaceuticals found in water and sediment samples might have considerable environmental consequences. The most obvious concern linked with the current study’s findings is that continual exposure to antibiotics even at low levels may lead to the development of antibiotic-resistant bacteria. The occurrence of antibiotics in the environment at sublethal doses can facilitate the presence of antibiotic-resistant bacteria in the ecosystem and hence the transfer of antibiotic-resistant genes to other bacteria [29, 49]. The presence of antibiotics at these sites, in the absence or presence of other resistance selective agents such as heavy metals, disinfectants, and detergents, can create a selective pressure that causes the proliferation of antibiotic resistance genes, resulting in the development or acquisition of resistance genes among microbes, rendering current treatment regimens useless [49]. Bacteria with antibiotic resistance genes have been observed in biofilms inoculated with drinking water bacteria [29]. This points to the possibility of gene transfer to the drinking water distribution network, maybe via surface and wastewater. The adverse effects of pharmaceuticals are not limited to antibiotics. In this study, diclofenac was the highest detected concentration. High levels of diclofenac in the environment could pose serious consequences. Unintentional exposure to diclofenac, for example, has been known to impact vultures, resulting in a significant decrease in their numbers a few years ago [50]. Diclofenac suppresses Gram-positive and Gram-negative bacteria growth by inhibiting DNA synthesis at high concentrations (50–100 mg/L), and alotic biofilms consisting of bacterial and algal communities lost about 70% of their overall initial biomass after 4 weeks of exposure to 100 g/L diclofenac [51]. Ibuprofen is reported to diminish the total bacterial biomass of a riverine biofilm community produced from rotating annular bioreactors and treated to a concentration of 10 g/L for 8 weeks [51]. Hormones such as estrogens are recognized as endocrine disruptors or modulators because they can negatively affect reproductive and sexual development, such as the feminization of male fishes at only ng/L concentrations and the sterilization frogs after progesterone exposure [46]. Conversely, Ref. [52] have reported that pharmaceuticals such as caffeine and salicylic acid can increase the yield of beet.

Thirty pure isolates (17 Gram-positive and 13 Gram-negative) bacteria were identified in water and sediment samples. *Bacillus, Corynebacterium, Pseudomonas, Klebsiella, Enterobacter, Listeria, Acinetobacter*, and *Clostridium* were among the bacteria that were identified. Various studies have also documented the isolation of bacteria from several of these genera from various locations across the world. [53] identified 101 bacteria belonging to 16 distinct genera from drinking water sources of Kohat, a northwestern district of Pakistan. They pointed out that these microbes are opportunistic pathogens and may constitute a considerable concern for public and occupational health. Borquaye et al. (2019b) isolated microbes from soil and leachate samples from active and abandoned landfill sites in Kumasi, Ghana. In a similar work carried out in Accra, Ghana, bacteria isolated included *Escherichia coli, Salmonella* spp., *Vibrio* spp., and *Bacillus* spp. [54]. In our study, diverse communities of microbes thriving in these sites were isolated, even though levels of antibiotics quantified were quite high in the same sites. Of the bacteria isolated from the sampling sites, *Bacillus* was the predominant genus. Five (*Enterobacter, Clostridium, Pseudomonas, Acinetobacter*, and *Klebsiella*) out of the eight isolates were resistant to all the antibiotics. Isolates of *Corynebacterium* and *Listeria* showed susceptibility to only doxycycline. Isolates of *Bacillus* demonstrated susceptibility to only two antibiotics (doxycycline and erythromycin). This is similar to the work done in Kumasi, where most of the bacteria isolated from landfill sites demonstrated resistance to antibiotics used in a susceptibility test. Tahrani et al. (2015b) studied resistance patterns of microbes isolated from industrial effluents in Tunisia, revealing six patterns of multidrug resistance among the 11 antibiotics tested. Also, in Nigeria, over 10% of the bacteria from the surface and underground water in six rural settlements were resistant to four or more antibiotics [55]. More so, Lu, Wu and Wang (2022) detected 18 types including 174 subtypes of antibiotic-resistant genes (ARGs) in water and microplastics of mariculture pond. Chloramphenicol-resistant genes were the dominant antimicrobial-resistant genes in their study. Although most *Bacillus* strains are not harmful to humans, prolonged contact with these microorganisms carrying resistance genes might spell tragedy. They might pass resistance genes on to other harmful organisms, wreaking havoc. Antimicrobial resistance in enteric bacteria, in particular, is a severe public health problem across the world. Skin, wounds, and the respiratory and gastrointestinal tracts are all places where *Acinetobacter* may invade. This microbe has also been linked to incidences of community-acquired infection [56].
The risks posed by the presence of the detected pharmaceuticals in the ecosystem were assessed in the ecological risk assessment study. The predicted no-effect concentration (PNEC: the concentration at which no adverse effect is suspected to occur) values for fish, *Daphnia*, and algae were determined using data from the literature on acute toxicity [9]. For water samples, HQ values for paracetamol indicated the medium risk to fish but a high risk to *Daphnia* and algae, respectively. The very high HQ values for diclofenac (HQ values $\gg 1$) imply quite a high risk to these species investigated especially against algae. Both paracetamol and chloramphenicol posed high risks to organisms in the sediments. Obviously, the worst hazard quotients were observed for diclofenac against *Daphnia* in the water samples and paracetamol on the same organism in sediment. In Pakistan, the HQ for pharmaceuticals from industrial effluents was determined. The maximum HQ values obtained in that study were with paracetamol (64 against *Daphnia*), naproxen (177 against fish), diclofenac (12,600 against *Oncorhynchus mykiss*), ibuprofen (167,300 against *Oryzias latipes*), ofloxacin (81,000 against *Pseudomonas putida*), and ciprofloxacin (440 against *Microcystis aeruginosa*) [20]. The high HQs recorded in this study unveil a looming danger to aquatic life which needs to be addressed urgently.

5. Conclusions

This study reveals that many households in and around the Owabi and Barekese reservoirs employ improper methods to dispose of unwanted and expired medicines, where throwing medicines into trash cans with other domestic waste are the most common disposal practice. The fact that respondents ranked health as extremely important while maintaining the environment as slightly important reveals the lack of awareness of the effects of improperly disposed pharmaceuticals on human health and ecosystems. Therefore, there is a need to create greater awareness of the problem of pharmaceuticals among the public and other sectors. However, the willingness of respondents to pay for the reduction or removal of pollutants of rivers and streams indicates concern and desire to care for the environment. This study also revealed the pollution status of the two dams with pharmaceuticals. The concentration of the pharmaceuticals ranged from 0.01 to 107.89 $\mu$g/L in the water samples and 0.10 to 10.22 $\mu$g/kg in the sediments. Although some of the targeted pharmaceuticals were below the detection limit, there was a detection of at least one pharmaceutical residue in all the samples gathered from both water and sediments, indicating potential pharmaceutical pollution of the reservoirs. Since water treatment plants do not specifically target the removal of pharmaceuticals, the effect on society could be enormous. It is important to institutionalize education programs to create awareness of the proper methods for the disposal of unused and expired medicines. Regular monitoring of the reservoirs will provide accurate information on the pollution status of the reservoirs. Finally, strategies for the removal of pharmaceutical residues during water treatment should be explored.

Data Availability

All data obtained or analyzed during this study are included in this published article.

Disclosure

The funders had no role in experimental design, data collection, or interpretation of data.

Conflicts of Interest

The authors declare that they have no known competing financial interests or personal relationships that could have influenced the work reported in this paper.

Authors’ Contributions

The study was designed by LSB, GD, FCM-R, and KM. All experiments were carried out by JNG, BAN, NAA, FF, GG, and BQ. The initial manuscript was drafted by JNG and BAN. LSB and GD supervised the work. All authors read and approved the final draft.

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Supplementary Materials

Figure S1: HPLC chromatogram for analyte standards. A: antibiotics at 215 nm; B: analgesics at 270 nm; C: analgesic at 220 nm. Peaks corresponding to specific analytes have been indicated. Table S1: HPLC flow program for the analyses of antibiotics and analgesics. Table S2: quality parameters. Table S3: frequency of disposal of prescription and nonprescription drugs. Table S4: disposal methods and medication types often disposed. Table S5: how important are personal care products in the environment: agents of subtle change? "Environmental Health Perspectives, vol. 107, supplement 6, pp. 907–938, 1999.

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