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Spatial Occurrence and Fate Assessment of Potential Emerging Contaminants in the Flowing Surface Waters

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Authors' contributions

This work was carried out in collaboration between all authors. Author MH designed the study, wrote the protocol, run experiments and wrote the first draft of this manuscript. Authors OC, NF and ME are my Ph.D. supervisors who approved this study and overall in-charge of all chemical and mathematical analyses. All authors read and approved the final manuscript.

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ABSTRACT

The current study investigated the occurrence of emerging contaminants (ECs) at the Msimbazi and Kizinga Rivers in Tanzania by using GC/MS QP2010 Ultra Shimadzu. The cross-country distribution along East Africa countries and the fate of ECs were studied for extrapolation and insight the extent of aquatic pollution in East Africa. It was revealed that all pharmaceuticals such as 0.0060 ppm of paracetamol, 0.0073 ppm of cetirizine, 0.0024 ppm of metronidazole and 0.0016 ppm of ibuprofen that were frequently quantified were also listed in the first-line medical prescriptions, they are available at over-the-counter supply and mostly used for the treatment of prominent diseases in Tanzania. It was further observed that quantified pharmaceuticals in Tanzania are certain in Kenya

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and Uganda since these countries have common diseases causing death and a similar list of essential medicines. Moreover, qualitative experiments identified the existence of norethindrone, caffeine, and polysiloxanes, which are proof of anthropogenic origin.

Keywords: Emerging contaminants; essential medicines list; surface water; solid phase extraction; Msimbazi; Kizinga.

1. INTRODUCTION

The traditional water and wastewater analysis have widely targeted the existence and fate of known and regulated environmental contaminants while ignoring emerging contaminants. The neglected contaminants were due to insufficient knowledge on their environmental occurrence, their presence below existed analytical detection limits and absence of the fear of the unknown [1]. Due to the long-term domestic and commercial uses of pharmaceuticals, personal care products, industrial solvents, disinfectants and chlorination of water, a suspicion of undesirable effects to the ecosystem existed [2,3]. With the advancement of analytical techniques over few decades, the existence of array of emerging contaminants (ECs) such as human drugs, endocrine disrupting compounds, water disinfection byproducts, algal toxins and industrial solvents have been revealed in the aquatic environment [4,5,6,7]. The knowledge of environmental occurrence and fate of ECs is limited, yet they are presumed to affect non-target organisms. Exposure to ECs is associated to hormonal interferences, feminisation, masculinisation of fishes as well as tumours and cancer to mice and rabbits [2,3]. The prevailing risks of ECs are extended to human being as they are presumed carcinogenic and induces bacterial resistance against medication as reported by Richardson et al. [1], Tahrani et al. [8].

A wide array of reports on the occurrence, transformation, and fate of ECs are frequently studied and reported in America, Asia, and Europe more than Africa [9,4,5,10,11]. These reports are themed to reflect phenomena such as the economy, technology, lifestyle and daily consumables of respective countries and therefore have limited relevant information to sub-Saharan countries rather than presumptions. In addition, few reports on the occurrence of ECs in our region such as [12] in Kenya, [13] in Tanzania, [8] in Tunisia and [14] Zambia exist. These reports haven't account for common diseases [15] and medication priorities [16,17,18] that may result in similar pharmaceutical contaminants. Thus, existing reports require knowledge broadening and wide coverage in the solicitation of sources and fate of emerging contaminants particularly in Tanzania and East Africa at large. Therefore, the current research addresses the quantitative and qualitative existence of emerging contaminants in the flowing surface waters of Tanzania, addresses the cross-country common risks of ECs in East Africa and the fate of ECs in the environment.

2. MATERIALS AND METHODS

2.1 Study Area

The Dar es Salaam city is located in the eastern coast of Indian Ocean at 6°48' South, 39°17' East of Tanzania. Before 1850s, the city was a small fishing village in the coast of East Africa. The city is among 31 Tanzania's regions with the population of 5,781,557 in the year 2017 [19].

Due to high population density, insufficient infrastructures, squatters, industrialisation, unemployment, and periodic floods some areas of the city experience excessive pollution despite of the invested cleaning efforts. Of these areas of the city, the Msimbazi and Kizinga Rivers were selected for sample collection as they were highly polluted due to wide coverage in the city, intensive habitats, industrial wastes, urban agriculture and drainage from domestic and wastewater stabilisation ponds as indicated in the Picture 1. Table 1 indicates the geographical coordinates of selected sampling points.

2.2 Sample Collection, Preparation and Analysis

Prior to sample collection, each sampling bottle was rinsed three times with the water sample to be collected. 30 samples were collected during the dry season of January 2016 in 0.5 L amber glass bottles and then placed in the dark cooled jar for transportation to avoid photo-oxidation of polyaromatic hydrocarbons. Temperature and pH values of all samples were recorded in-situ. All samples were stored at 4°C refrigerator before sample preparations and analysis.

Prior to sample preparation, all samples were subjected at room temperature before filtered through GF-C 47 mm diameter, 1.2 µm size exclusion glass fiber filter papers placed in the Buchner funnel [20]. Filtered samples allowed to pass through Hydrophilic-Lipophilic Balance (HLB) cartridges (60 µm pore size, 12 mL volume, and 500 mg) mounted on the pressurised manifold station for extraction of analytes of interest [21]. Cartridges conditioning in order to activate adsorbent attained by soaking with 5 mL of analytical grade methanol followed by 5 mL of HPLC grade water [22]. Each sample passed through independent conditioned SPE cartridges for extraction under controlled vacuum. Again, 5 mL of HPLC grade water used for washing cartridges before vacuum drying. Cartridges were dried for 30 minutes under gently flowing nitrogen gas. Target analytes were eluted from SPE cartridges by using two aliquots of 2.5 mL of analytical grade methanol at the flow rate of 3 mL/min. The resulting solvent and extracts pre-concentrated to 2 mL by using gently flowing nitrogen gas. 2 µL of the pre-concentrated sample were auto-injected in GCMS-QP2010 set under splitless mode. The injection temperature was 80ºC then raised to 250ºC while the interface temperature was 300ºC [23].

Picture 1. Kizinga (LEFT) and Msimbazi (RIGHT) rivers showing intensive pollution

Table 1. Sampling points coordinates

S/N	Sampling point	Sampling code	Coordinates	
1	Vingunguti-Segerea Bridgre	MS1	-6.8434150	39.2208633
2	Sprenko terminal	SP3	-6.8329390	39.2346550
3	MS before Luhanga River	MS6	-6.8234740	39.2395140
4	Luhanga River	TOT	-6.8217500	39.2395140
5	Kigogo (After MS&TOT junction)	MS7	-6.8236290	39.2402170
6	Kigogo Mwisho (After UB&MB junction 2)	MB5	-6.8159090	39.2624750
7	Ubungo River, MB before join MS	MB7	-6.8159090	39.2624750
8	MS before join UB, Kigogo street	MS8	-6.8176570	39.2398150
9	MS After MS&Sinza junction	MS10	-6.7985460	39.2700000
10	Inlet 1 River to Kizinga	KZ3	-6.8870767	39.2594417
11	Kizinga river before inlet 4	KZ9	-6.8561567	39.2414317
12	Control sample	C		

MS-Msimbazi River, SP-Sprenko, KZ- Kizinga River, TOT-Luhanga River

Preparation of calibration curve by using reference standards evolved preparation of stock solutions of paracetamol, cetirizine, metronidazole and ibuprofen 1000 ppm followed by serial dilutions [24]. All standards stored at 4ºC refrigerator for further procedures. These standards run in the GC/MS in order to establish the calibrations curves, limit of detection (LoD) and limit of quantification (LoQ). Reference standards were the baseline for quantitative analysis in which paracetamol found to have 115% recovery.

3. RESULTS AND DISCUSSION

The bioavailability of medicines through the oral intake is always higher and detectable in the body than rectal administration [25]. The percentage recovery of excreted free drugs is smaller than its metabolic or conjugated products. Only 1% of free and 24% of conjugated ibuprofen recovered from urine within 24 hour of intake [26]. Contrarily, the availability of cetirizine in the urine is reported 70%, indicating a limited metabolism, while 9% of paracetamol excreted in unchanged form. Thus, the probability of occurrence of large amount of pure cetirizine is higher than paracetamol that agrees with the current findings.

Environmental occurrence of similar ECs in East Africa countries might be either trans-boundary effects or merely common diseases causing death as indicated in Table 2. When looking to the list of Tanzania's first-line and essential prescribed medicine presented in Table 3 for combating general and acute diseases is almost similar across East Africa region and therefore common type of pharmaceutical pollution [16,15,27,17,28,18].

The spatial occurrence of some pharmaceuticals is presented in the Table 3 where pH and turbidity are included as they are among parameters affecting the existence of emerging contaminants in the water. The frequency of occurrence of some ECs in this study revealed paracetamol as the most frequently occurring pharmaceutical, followed by caffeine, metronidazole, norethindrone, polysiloxane, ibuprofen and cetirizine. The occurrences of selected drugs reflect quantitative daily consumption, economy of the society and root of administration and disposal as well as the possibility of cross-country contamination. Cetirizine had the largest quantitative average for its occurrence i.e. 0.0095 ppm compared to 0.0041 ppm of paracetamol, 0.0032 ppm of metronidazole and 0.0021 ppm of ibuprofen.

This might be contributed by closeness to the medical facility, limited biotransformation of cetirizine in the body where 50% is excreted as pure cetirizine and/or high prescription rate and therefore high chances of environmental availability. This implies that under similar conditions such as turbidity, flow rate, pH, vegetation coverage and other river processes cetirizine is transferable over a long distance [29] compared to other contaminants.

Table 2. Top ten total death-causing diseases from selected countries in East Africa

Diseases	Tanzania		Kenya		Uganda	
	TZ rank	WR	KN rank	WR	UG rank	WR
Influenza and Pneumonia	1	30	2	41	2	34
HIV/AIDS	2	22	3	23	1	16
Diarrhoeal diseases	3	18	1	5	5	30
Tuberculosis	4	9	7	44	8	48
Coronary Heart Disease	5	135	5	180	4	162
Road Traffic Accidents	6	9	6	16	7	6
Stroke	7	104	4	117	3	81
Malaria	8	26	9	32	10	29
Birth trauma	9	43	NL	NL	27	46
Congenital Anomalies	10	19	24	34	25	37
Cervical Cancer	22	5	10	19	12	12

WR – World Rank, NL – Not listed

Table 3. Tanzania's first-line medicines for treatment of other general diseases

Disease description	Some examples of drugs	Accessibility at dispensary
Influenza and Pneumonia	Amoxicillin and paracetamol or ibuprofen	Available
HIV/AIDS	Tenofovir, Lamivudine or Efavirenz	Available
Tuberculosis	Ethambutol	Available
Malarial	Artemether/Lumefantrine, Quinine	Available
Treatment of Acute Attack	Acetylsalicylic acid, Ibuprofen	Available
Anti-allergies	Cetirizine, Chlorpheniramine, Elixir,	Available
Antiamoebic	Metronidazole	Available
Anti-asthmatics	Cromoglycate Nasal spray	Available
Diarrhoea	Zinc tablets	Available
Antihypertensive	Bendrofluazide	Available
Ovulation Inducers	Clomiphene	Available
Oral contraceptives	Ethinyloestradiol + Norgestrel	Available
Anxiety Disorders	Diazepam	Not available
Antiarrhythmic	Atenolol	Not available
Cervical Cancer	Radiotherapy	Not available

Table 4. Spatial occurrence of selected ECs

Sampling codes	pH	Turbidity (NTU)	Paracetamol (ppm)	Cetirizine (ppm)	Metronidazole (ppm)	Ibuprofen (ppm)
MS1	7.17	948	0.022767	BDL	BDL	BDL
SP3	11.2	222	0.00836	BDL	BDL	BDL
MS6	9.60	127	0.00588	BDL	BDL	BDL
TOT	7.47	9.74	BDL	BDL	BDL	BDL
MS7	9.45	180	0.00556	BDL	BDL	0.01928
MB5	7.79	39.2	0.00724	BDL	BDL	BDL
MB7	7.83	17.5	BDL	BDL	BDL	BDL
MS8	9.28	69.6	BDL	BDL	0.01572	BDL
MS10	7.66	9.84	0.00952	0.08736	0.01348	BDL
KZ3	7.75	26.4	0.00704	BDL	BDL	BDL
KZ9	6.97	29.6	0.00588	BDL	BDL	BDL
C	5.56	1.68	BDL	BDL	BDL	BDL
Average	8.14	140.05	0.0060	0.0073	0.0024	0.0016

SP – Effluents from Sprenko wastewater stabilization ponds, MS – Msimbazi River, MB – Ubungo River, TOT – Luhanga River, KZ Kizinga River

The frequency of occurrence of paracetamol is higher and widespread at Msimbazi and Kizinga Rivers than other contaminants as shown in Fig. 1.

Paracetamol is a first line prescribed analgesic in Tanzania and therefore its abundance is probable. MS1 is a slaughtering area at Vingunguti with the highest concentration of 0.023 ppm compared to other locations. This might have sourced from the disposal of unused drug, direct urination in the river, direct discharges from intensive commercial and domestic activities. High amounts of paracetamol up to 0.043 ppm was reported from Oslo hospital [30] and 0.023.8 ppm from wastewater treatment plants in Spain [31]. The occurrence of ECs at the Msimbazi is more than Kizinga River due to population coverage, the presence of medical facility and the size of the river.

The occurrence of metronidazole at sampling points MS6 and MS7 are shown in the Fig. 2. This is an indication that metronidazole possibly not originated from residential areas, rather from industrial area. Commercially metronidazole is a medication for wide stomach disorders and therefore its presence in this area requires intensive investigation on whether it is a raw material in either of the industry or industrial workers frequently consumes it. Metronidazole is reported as a priority contaminant with high frequency of occurrence, genotoxic and not biodegradable [32].

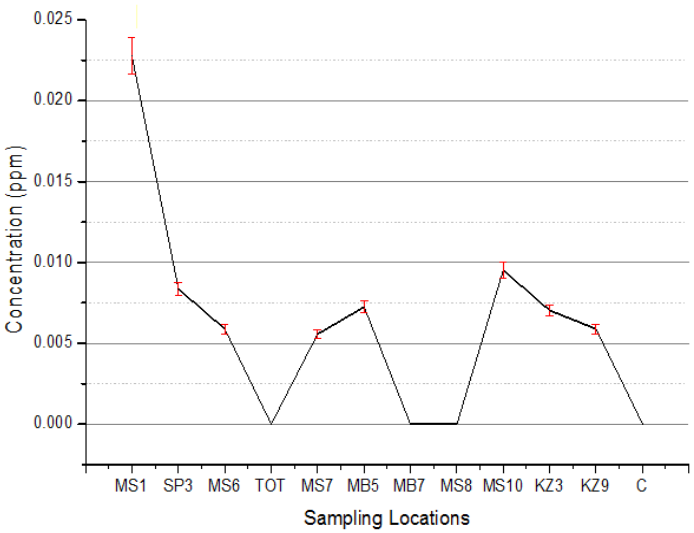

Fig. 1. Paracetamol occurrence

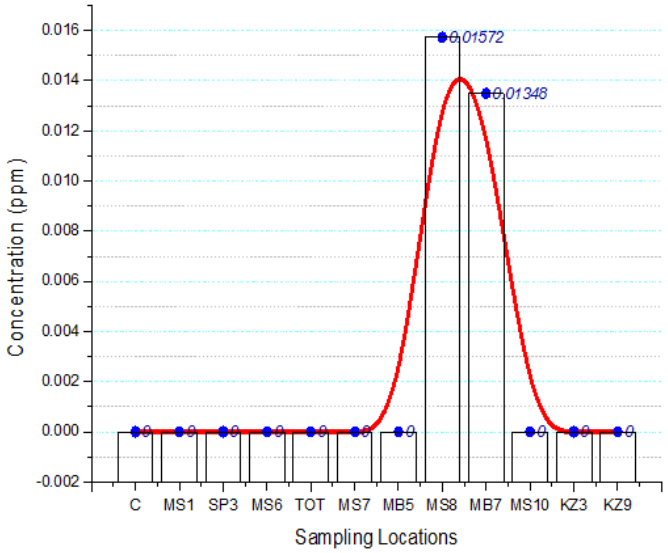

Fig. 2. Metronidazole occurrence

Cetirizine is an antihistamine medication for relief of allergic reaction such as itching, sneezing, running noses and watery eyes. It is administered either by injection or orally with 24 hours duration of action and 8.3 hours of elimination half-life. Fig. 3 shows that sampling point MB7 had significant amounts of cetirizine, contrary to its absence at sampling points MB6 and MS9. The probable source is a substantial waste dumping in the river at a couple of hundred meters before this point and non-domestic washings in this area that might. The distribution Fig. 3 predicts possibilities of contamination at sampling points MS8 and MS9, however, the flow rate at sampling point MS8 is greater than sampling point MB7 thus dilution effects, density due to turbidity and mixing capacity affects the availability of cetirizine at sampling point MS9. This process is similar but inverse to the occurrence of metronidazole at sampling points MS6 and MS7 where the dilution effects from TOT could not significantly change the concentration due to a small amount of flowing water.

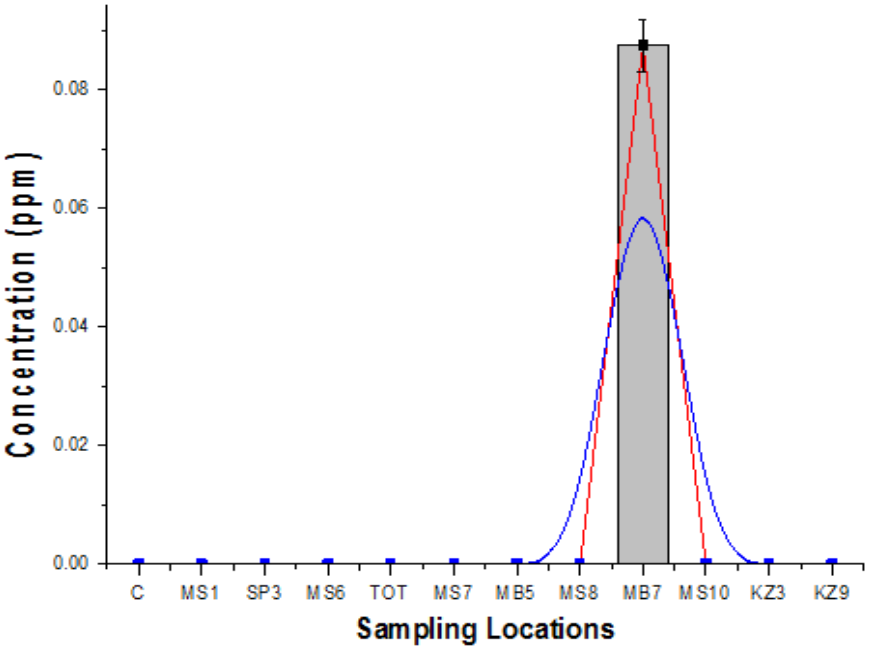

Fig. 3. Concentration of cetirizine

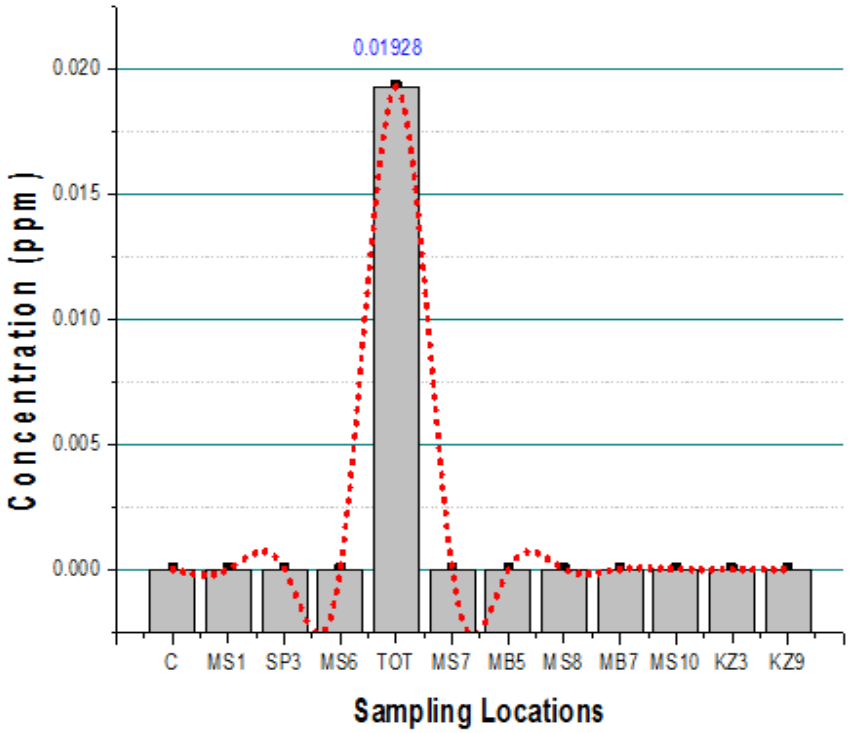

Fig. 4. Concentration of ibuprofen

A similar study in UK reports 50% of administered cetirizine is excreted via urine to the WWTPs which maximises the quantitative occurrence [9]. The occurrence of most ECs in the WWTPs' effluents is a common phenomenon since conventional methods of wastewater treatment do not remove ECs.

Table 5. Metabolism process of some selected human drugs in the body

Drugs	Metabolites	% in Urine	% in faeces	References
Ibuprofen	R and S isomers	95	5	[35]
Cetirizine		70	10	[36]
Metronidazole	Ornidazole and 2-hydroxymethyl metronidazole	75	25	[37]
Paracetamol	Acetanilide, phenacetin, O-glucuronide, N-hydroxyamide,	9	NR	[38]

NR – Not Reported

Ibuprofen is a non-steroidal anti-inflammatory medicine used for the treatment of inflammation, pain and fever. Ibuprofen is less prescribed than paracetamol, consequently; its environmental occurrence is minimal. Stasinaksi reported less frequency of occurrence of ibuprofen in the treated wastewaters [33], which is similar to observations in this study. The occurrence of ibuprofen at Luhanga River (TOT) indicated in Fig. 4 might be originating from discharges from intensive squatters, probable available industries such as NIDA textile, rat poison industry and intensive and excessive solid waste dumping. Ibuprofen was not detected at location MS6 possibly due to absence of water backflow. The same case was observed at location MS7, which can be linked to excessive water flow from location MS6 that could cause contaminants dilution to below quantification limit. While this study has quantified 0.019 ppm of ibuprofen at Msimbazi River, Jasinska reported ibuprofen ranging from 60.47 to 14.46 ng/L from the WWTP [34].

3.1 Environmental Fate of Pharmaceuticals

Some medicines that are quickly accessible at the dispensary levels and over-the-counter supply represent a group of drugs frequently consumed and therefore high probability of their environmental occurrence and distribution as similarly observed and reflected in our current findings. The environmental occurrence of these drugs is a proof of the presence of various diseases, easy accessibility of these drugs, improper disposal of unused or expired drugs, direct domestic discharges to the river and direct urination in the rivers. Some medicines that are expensive, rarely used and absence from over-the-counter supply are less common in the environment probably due to less consumption. Injectable drugs are also rare in the environment due to their mode of administration [27,17] compare to tablets and capsules. Table 5 indicates drug metabolites and significant root of body excretion. Urine proved to have high percentage of consumed drugs excretes [39] and therefore a probable potential domestic and commercial source of pharmaceuticals in the water bodies.

4. CONCLUSION

Cross-contamination of emerging contaminants along the East Africa region is a probable cause since some antibiotics such as sulfamethoxazole, ciprofloxacin, trimethoprim, and antiviral such as zidovudine, lamivudine, and nevirapine reported in Nairobi River basin in Kenya are similar essential pre-scribed drugs in Tanzania and Uganda therefore high possibility of their occurrence in East Africa. ECs are characterised by fluxing, bioaccumulation and bio-concentration in organisms thus through food chain they get to human body leading to health effects. There is a need for further investigation on the existence of other classes of emerging contaminants such as norethindrone, polysiloxane, and caffeine, which were qualitatively determined in this study. It is important to note that the absence of some ECs in some location is an indication of less consumption and dispose of such contaminants and effective adaptation of available environmental conservation regulations.

COMPETING INTERESTS

Authors have declared that no competing interests exist.

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