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1-D Transport Behaviour of Emerging Contaminants in the Unidirectional Flowing Surface Waters of the Msimbazi River

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Abstract

The primary objectives of this investigation of environmental pollution were to assess spatio-temporal concentrations and predictions of transport behaviour of pollutants in aqueous media. In order to attain these goals for the Msimbazi river, the analytical and numerical solutions to the 1-D advection-dispersion equation (ADE) implemented the first-type inlet condition. The MATLAB software was used to solve the analytical equation resulting from the ADE equation. The solution to the ADE model enabled effective prediction of contaminants'spatial distribution within a distance of 200 m from a point source in the direction of flowing water. Among the first and third-types inlet conditions, the first-type had the best exponential concentration prediction with descending concentration gradients between –100 ng/L.m and –0.4 ng/L.m, while beyond it the concentration gradient was merely constant. These findings are the first of this type in our region to the best of our knowledge, and hence a milestone for understanding the transport behaviour of emerging contaminants, and enable estimation of probable pollution-free zones. This study gave results that can form a basis for analytical and public interventions.

Keywords: Modelling emerging contaminants; Advection-Dispersion Equation; Contaminants transport; First-type inlet condition; Concentration gradient.

Introduction

Emerging contaminants (ECs) constitute chemicals and microbial contaminants originating from clinical, industrial, water treatment plants, municipal wastewaters and agricultural runoffs. Although the first report on the occurrence of ECs in the aquatic systems of Sweden was in the 1970s (Darnerud et al. 2001), mitigation approaches were documented from the year 2000s (Philips et al. 2005). The delayed mitigation approaches were not an accident, it was due to inadequate information and analytical techniques were yet in place. The pressure for high motives on investigation of ECs during the 21st century was due to their alleged ecological threats (Daughton 2004, Shane 2014), the advancements in analytical technology (Richardson 2006, Richardson 2007) and due to the failure of conventional methods for water treatment on removing ECs (Benitez et al. 2013).

Most ECs, particularly organic ones, are insoluble in the water and thus are transported as free contaminants or adsorbed on the surfaces of suspended matters or

sediments in the direction of moving water. This type of transport is a longitudinal transport which is referred to as an advection process. The advection process is, therefore, a concentration-based condition, in this case, considered as the first-type boundary condition. The lateral and vertical transportation of contaminants in the fluid is a dispersion process, in this case, regarded as the third-type or flux-based boundary conditions. 1-D modelling of ECs transport accounts for longitudinal movement and time, while the 2-D modelling accounts for both lateral and vertical transport (Leij and Dane 1990). During the transport of ECs in the flowing surface waters, ECs undergo sorption, dispersion, decay, and desorption, which affect their spatial and temporal occurrence (Genuchten 1981, Lee et al. 2014). The interaction between ECs with air and heat may lead to either their natural oxidation or degradation. Thus, the use of a model can answer the fundamental question of the spatial and temporal transport behaviour of ECs in the flowing surface waters (Yadav et al. 2010). Modelling simplifies the understanding of a complex system and enables future predictions of contaminants' transport behaviour (Spangenberg 2007). Although integrating monitoring and modelling data provides more reliable information than a single approach, a good model can take care of both process and numerical data problems (Loucks and Beek 2019).

Most researches in this area have focussed on the occurrence and remediation of ECs (Grober et al. 1998, Agüera et al. 2005, Lopez et al. 2015, Česen et al. 2019) with few researches concentrating on the use of modelling (Genuchten 1981, Leij and Dane 1990, Genuchten et al. 2013). Yet, few existing researches that have used the ADE model equations (Genuchten et al. 2013) cover theoretical aspects than practical applications. Therefore, the current study details the applications and implementation of ADE relative to the flowing surface water in the modelling of ECs transport, as a fundamental basis to explain their transportation behaviour.

Materials and Methods

The numerical and analytical models are among the existing mathematical computations for evaluation of transport behaviour of contaminants in the surface water. A numerical model is useful under complex environments, and when more details are needed, while the analytical model relies on uniform properties and regular geometry (Chen et al. 2012). Guerrero reported that the analytical model is numerically stable because it allows fixation of confounding variables, quick to use, valuable as a screening tool and applicable in all dimensions (Guerrero et al. 2013). The advection-dispersion equation (ADE) which is an analytical-based model, was adopted in this study.

Advection-Dispersion Equation (ADE)

The general advection-dispersion equation (ADE) presented in Equation 1A (Jaiswal et al. 2011) stands as a critical equation for unidirectional movements of contaminants in the flowing surface waters (Genuchten et al. 2013).

$$\frac{\partial C}{\partial t} = D_x \frac{\partial^2 C}{\partial x^2} - u \frac{\partial C}{\partial x} \qquad (1A)$$

Where: C is the contaminant concentration at a specified sampling point (M/L^3), and t is time (T), u is the longitudinal fluid flow velocity (L/T), Dx is the longitudinal dispersion coefficient (L^2T^{-1}) and x is longitudinal direction (L).

However, the intra-molecular assumptions including biodegradation, inactivation, production and contaminant decay may affect contaminant concentrations during linear transport. Thus, Equation 1B is a compliment equation that accounts for the named assumptions.

$$\frac{\partial C}{\partial t} = D_x \frac{\partial^2 C}{\partial x^2} - u \frac{\partial C}{\partial x} - \mu C + \gamma \qquad (1B)$$

Where: μ is a first-order decay rate (T^{-1}), and γ is a zero-order contaminants production ($ML^{-3}T^{-1}$), with the assumption that the μ and $\gamma \geq 0$.

Conditions for advection-dispersion equation

To solve the ADE, we need one initial condition and two boundary conditions. The initial condition takes the form, as presented in Equation 2. It accounts for the concentrations of contaminants as a function of distance at a constant contaminant release.

$$C(x,0) = f(x) \quad (2)$$

Equations 3 and 4 present boundary conditions whereby Equation 3A is an upstream boundary condition depicting that any concentration must be continuous across the medium all over the time.

$$C(0,t) = g(t), t > 0 \quad (3A)$$

The Equation 3B is a downstream boundary condition, assuming the system to be finite or semi-infinite.

$$\left.\frac{\partial C}{\partial x}\right|_{x=\infty} = g(t), t > 0 \quad (3B)$$

The Equation 4A is an upstream boundary condition introducing mass balance conservation by assuming the position of the system to be positive.

$$uC(0,t) - \omega D_x \cdot \left.\frac{\partial C}{\partial x}\right|_{x\geq 0^+} = ug(t) \quad (4A)$$

Where 0^+ indicates a position just inside the system being considered.

The Equation 4B is a downstream boundary condition accounting for the concentrations against distance gradient to be zero as the distance from the source of contamination approaches infinite.

$$\frac{\partial C}{\partial x} = 0, x \to \infty \quad (4B)$$

There are some molecular factors neglected from 1-D contaminants transport cases because of their insignificant contributions and for simplification of formulation and application of the ADE model. The neglected inter-molecular factors include the extraction and production of contaminants, accumulation, and depletion induced by injection or pumping. The neglected intra-molecular factors include biodegradation, inactivation, production and contaminant decay that affect contaminant concentrations during linear transport.

1-D solution for advection-dispersion equation

Advection-dispersion equations can be solved either by Laplace transformation or by the transformation of variables (Jaiswal et al. 2011). Transformation helps reduce the equation to a simpler form and therefore easier to solve.

The analytical solution is preferred in modelling transport of suspended and dissolved contaminants than numerical since it makes the use of variables transformation via mathematical principles. The analytical solution is not only for simplicity; instead, it is suitable with most flowing waters because they are more likely to be infinite in the longitudinal direction. It is also suitable for non-uniform established initial concentrations with continuous application of background concentrations.

By solving the Equation 1B under the stated boundary conditions, 2 to 4B, where the values of omega are zero for the first-type inlet condition and one for the third-type inlet condition gives out the analytical solutions. Equation 5A represents the analytical solution for the first-type inlet condition which is adopted in this study, while Equation 5B shows the analytical solution for the third-type inlet condition. These equations are useful at the semi-infinite domain with non-uniform initial concentration and continuous application of background concentration 'C_b'.

$$C(x,0) = E(x) = \frac{\gamma}{\mu} + \left(C_b - \frac{\gamma}{\mu}\right)\exp\left(\frac{x(u-\varepsilon)}{4D_x}\right) \quad (5A)$$

$$C(x,0) = E(x) = \frac{\gamma}{\mu} + \left(C_b - \frac{\gamma}{\mu}\right) * \left(\frac{2u}{u-\varepsilon}\right)\exp\left(\frac{x(u-\varepsilon)}{4D_x}\right) \quad (5B)$$

Zeng reported the computation of dispersion coefficient (Dx) with the best assumption compared to others such as Fischer (1967), Liu (1977), Kashefipour (2002) as indicated in Equation 6 (Zeng and Huai 2014).

$$D_x = 5.4\left(\frac{B}{H}\right)^{0.7} * \left(\frac{u}{U^*}\right)^{0.13} * Hu \quad (6)$$

Whereby *B* is the average width of the studied river (2.544 m), *H* is the mean depth of the river (0.2542 m) and U* is the shear velocity.

While Zeng and Huai (2014) were silent about the shear velocity, Katul reported how shear velocity (U*) could be computed based on Equation 7 (Katul 2002).

$$U^{*} = \frac{u}{h^{1/6}} * n\sqrt{g} \qquad (7)$$

Where: u is the average velocity measured in the field (0.482 m/s), h is the average depth of the studied river (0.2542 m), n is the Manning's roughness (0.1-0.035 sm^{-3}) (Oregon 2014) and g is the gravitational force of the earth (9.81 ms^{-2}). The calculation of μ value previously reported by Godfrey and Frederick (1970) and later reported by Rajeev is as presented from Equation 8 (Rajeev 2013).

$$\mu = 0.18\left(\frac{U^{*}}{u}\right)^{1.5} \qquad (8)$$

The computation of epsilon (ε) indicated in Equation 9 is adopted from Genuchten et al. (2013).

$$\varepsilon = u\sqrt{1+\frac{4\mu D_x}{u^2}} \qquad (9)$$

Sample collection, preparation and analysis

Prior to sample collection, each sampling bottle was rinsed three times with the water sample to be collected. In order to reflect spatial aspect in this study, sampling points were random systematically selected to account for sampling point accessibility, presence of multiple industries and wastewater stabilization ponds (WWSP), as well as open areas allowing human access for activities like dumping, washing and sand excavation. As presented in the sampling area map (Figure 1), samples were collected over the whole main river as well as from each tributary.

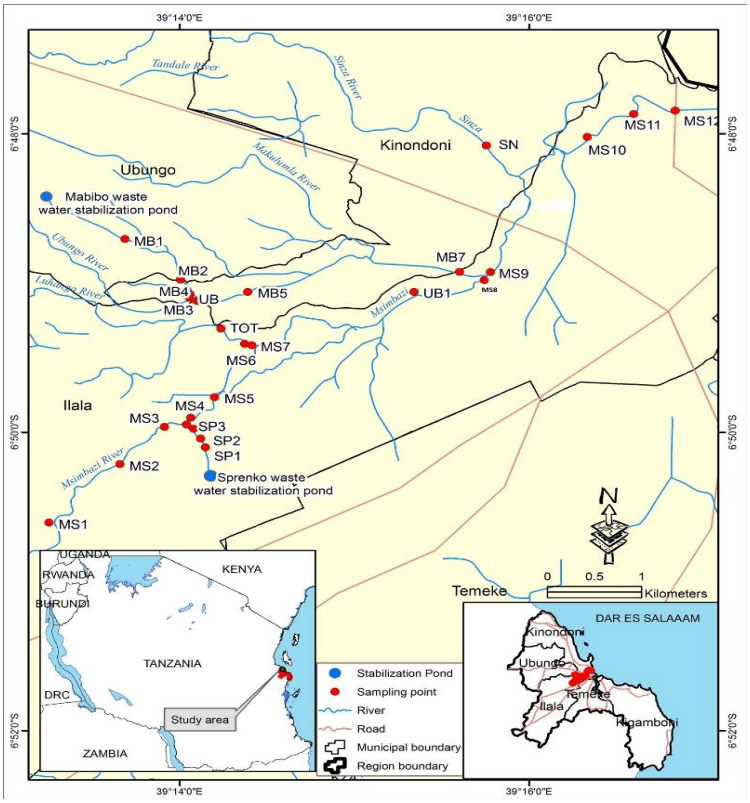

Figure 1: Map showing sampling points.

Sampling points in the main Msimbazi river covered from MS1 (Vingunguti ward) to MS12 (Selander bridge, 50 m before Indian ocean) which is about 8.4 km apart. Sampling points from wastewater stabilization pond tributary included SP1 (at the spout of Vingunguti WWSP) to SP3 (5 m before joining main Msimbazi river), which is about 0.7 km apart. The second tributary was Luhanga river at TOT where the first sample was collected at a distance of 1 km before, and the second sample was collected at a distance of 5 m before the main Msimbazi river. The third tributary was Mabibo WWSP from MB1 (Mabibo mwisho) to MB7 (Jangwani) which is nearly 4.7 km apart. The fourth tributary was Sinza river SN where a single sample was collected about 1.2 km before the main Msimbazi river as there was no any other closer accessible point.

Since each tributary contributes to the pollution of the main river, in order to account for its effects, three samples were collected at each intersection too, i.e., 5 m before and after each intersection (SP3, MS3 and MS4), (MS6, TOT and MS7), (MS8, MB7 and MS9), (MS9, SN and MS10).

In order to reflect temporal aspects, samples and data collection, as well as analysis involved both dry and wet/rainy periods. Based on climate of Tanzania, short rains occur between October and December, while more intense long rains occur from March to May of each year. The period between January and February is normally dry with hot weather. However, sometimes slight climatic changes occur. For example, the 2016/2017 short rains were observed from 27th January 2017 instead of between October and December 2016, while the 2019 short rains commenced from December 2019 to nearly end of February 2020. Thus, in order to reflect the dry season, sample collection was conducted on 17th January 2017. By that time, the short rainy period of 2016/2017 was yet to begin as it was late until 27th January 2017 where it commenced. The collected samples and data were for establishment of basic analylical and transport behaviour information of the river in the dry season. Towards the acquisition of water samples in the wet period, sampling was conducted in the short rain period. In that period, the experienced extended short rains commenced from December 2019 to nearly end of February 2020. In our case, sample collection conducted on 4th January 2020 to account for the wet/rainy season.

Thirty (30) samples were collected 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 on-site. All samples were stored at 4 °C refrigerator before sample preparations and analysis. During wet season, the water was very turbid and had high flow rates thus was highly risky and unsafe for sample collection and gave highly diluted samples. In this case, mathematical data, i.e., the flow rate obtained during the rainy season were useful in acquisition of important information for computation of transport behaviour of Msimbazi river for the rainy season.

Prior to sample preparation, all samples were subjected to room temperature before being filtered through GF-C 47 mm diameter, 1.2 µm size exclusion glass fiber filter papers placed in the Büchner funnel. Filtered samples were 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. Cartridges conditioning in order to activate the adsorbent was attained by soaking with 5 mL of analytical grade methanol followed by 5 mL of HPLC grade water. Each sample was passed through independent conditioned SPE cartridges for extraction under controlled vacuum. Again, HPLC grade water (5 mL) was 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 were pre-concentrated to 2 mL using gently flowing nitrogen gas.

A 2 µL volume of the preconcentrated sample was auto-injected into a GC-MS-QP2010 set under splitless mode. The injection temperature was 80 °C then raised to 250 °C, while the interface temperature was 300 °C (Miraji et al. 2018).

Preparation of calibration curves by using reference standards involved preparation of 1000 ppm stock solutions of paracetamol, cetirizine, metronidazole and ibuprofen followed by serial dilutions. All standards solutions were stored at 4 °C in a refrigerator pending further procedures. These standards were analysed on the GC-MS in order to establish the calibrations curves, limits of detection (LoD) and limits of quantification (LoQ). Reference standards were the baseline for quantitative analysis wherein paracetamol was found to have a 115% recovery.

Results and Discussion

The analysis revealed the presence of some emerging contaminants particularly pharmaceuticals such as paracetamol (0.0066 mg/L), cetirizine (0.0097 mg/L), metronidazole (0.0032 mg/L) and ibuprofen (0.0021 mg/L), where cetirizine had the highest average concentration as indicated in Table 2. Figure 2 displays the measured flow rates of water in the Msimbazi river at different selected points, with the average flow rate of 389.2 L/s in the dry season and 11,613.5 L/s in the wet season, four days after heavy rains. Thus, with a constant background concentration of 0.0041 mg/L about 1.6 mg of paracetamol flows per second, which is equivalent to 5.7 g per hour in the dry season. The actual wet season concentrations of ECs were difficult to establish due to inaccessible sampling points, high dilutions that resulted to lower concentrations below the detection limits of the method and also high turbidity that resulted to excessive clogging of glass fiber filter papers beyond expected performance.

The fact that about 85–95% of the therapeutical dose of paracetamol is excreted in the urine within 24 hours (Forrest et al. 1982, Cooper et al. 2008) supports this observation. Thus, wastewater stabilisation ponds and commercial/public areas can be potential sources of paracetamol.

Witte (2012) reported a water flow rate of 492 L/s, which was obtained from dry season of July to August 2011 at the Msimbazi river (Witte 2012), contrary to the previous flow rate of the Msimbazi river in 1984 which had a range of 0.07 m^3/s to 0.17 m^3/s as reported (Ak'habuhaya and Lodenius 1988, De Wolf et al. 2001). During field survey, it was noted that these variations were due to the variations of water volume during the rainy and dry seasons, rates of industrial discharges since each industry has a specific cleansing day, soil erosion which reduces the depth of the river, and amounts of suspended materials and vegetation coverage which returded the speed of water and the slope of the sampling point which determines the speed of water.

A mathematical prediction of the distance in which ECs can be transported in the flowing surface water of Msimbazi river was obtained from the solved ADE as previously presented in the Equations 5A and 5B. The analytical and numerical computations of these equations were done using Matlab software version R2013b: 8.2.0.701 (Table 1). The solution obtained using the first-type inlet condition for paracetamol is shown in Figure 3. It is a smooth decay curve indicating a continuous decrease in the concentrations as one moves from the source of contamination downstream. The concentration gradient of paracetamol shows that the predictions are compelling within a range of 200 m along the direction of flowing water. As moving far beyond 200 m, the variations in concentrations were insignificant. For example, looking at point MS1, with concentration of 0.0228 ppm, paracetamol could not be observed at point MS2 which was about 375 m apart unless there is another source in between. The absence of paracetamol from SP1 and SP2 despite their distance being less than 200 m from Vingunguti WWSP were not significant sources of paracetamol at point SP3, rather there were other sources of this paracetamol, possibly disposal of unused drugs. The

0.00556 ppm of paracetamol at point MS7 might have originated from MS6 because of close distance of about 146 m and the slight decrease in the concentrations may be associated with dilution effects. The distance between MS7 and MS8 is about 2.8 km thus, there no possibility of transfer of paracetamol along such a distance. Also, there are very limited points of accessibility to the river between MS7 and MS8; therefore reduces the possibilities of dumping in this area. MS10 is very close to Muhimbili national hospital and therefore it is at risk of clinical waste contamination.

The flow rate is still a very significant factor for determination of lethal dosage for aquatic organisms resulting from bioaccumulation and bioconcentration. The up-stream concentration gradient presented in Figure 4 shows that between the distances of 0 to 75 meters, concentration gradients are higher than the preceding distance ranges. This observation is a reflection of excessive pollution at upstream and gradual contaminant dilution as moving down stream. After 75 m, the gradient is not as sharp as it happens down-stream where deposition is typical than transportation. During wet season, similar data were difficult to be generated due to erlier presented challenges.

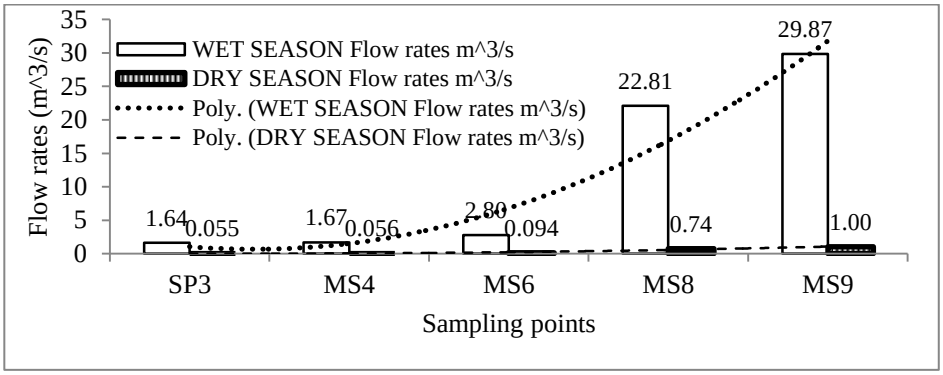

Figure 2: Dry and wet season (temporal) flow rates of the Msimbazi River.

Table 1: Matlab coding for ADE analytical solutions

Matlab command	Descriptions
γ=a=1;	Γ zero-order production term, (must be zero or positive) ($ML^{-3}T^{-1}$)
μ=m=0.045;	μ-first-order decay rate, must be zero or positive (s^{-1})
cb=C=6600;	Variable 1 (concentration, *100 ng/L)
x=0:0.5:200;	Variable 2 (distance, m)
u=0.482;	Mean velocity of water (m/s)
Dx= D=3.75;	Longitudinal dispersion coefficient (L^2T^{-1})
ε=E=0.953;	Transformation factor, ε=E
c=(a/m)+(C-(a/m))*exp(x*(u-E)/(4*D));	ADE first-type analytical solution,
plot(x,c,'k')	Command for graph sketching
xlabel('Distance [m]');	Horizontal axis label
ylabel('Concentration [ng/L]');	Vertical axis label
title('Analytical Solutions for First-type Inlet Condition');	Title of the chart

Table 2: Levels of of observed ECs in water during dry season **(**ppm)

Sampling codes	Paracetamol	Cetirizine	Metronidazole	Ibuprofen
MS1	0.022767	BDL	BDL	BDL
SP3	0.00836	BDL	BDL	BDL
MS6	0.00588	BDL	BDL	BDL
TOT01	BDL	BDL	BDL	0.01928
MS7	0.00556	BDL	BDL	BDL
MB5	0.00724	BDL	BDL	BDL
MS8	BDL	BDL	0.01572	BDL
MB7	BDL	0.08736	0.01348	BDL
MS10	0.00952	BDL	BDL	BDL
Average	**0.0066**	**0.0097**	**0.0032**	**0.0021**

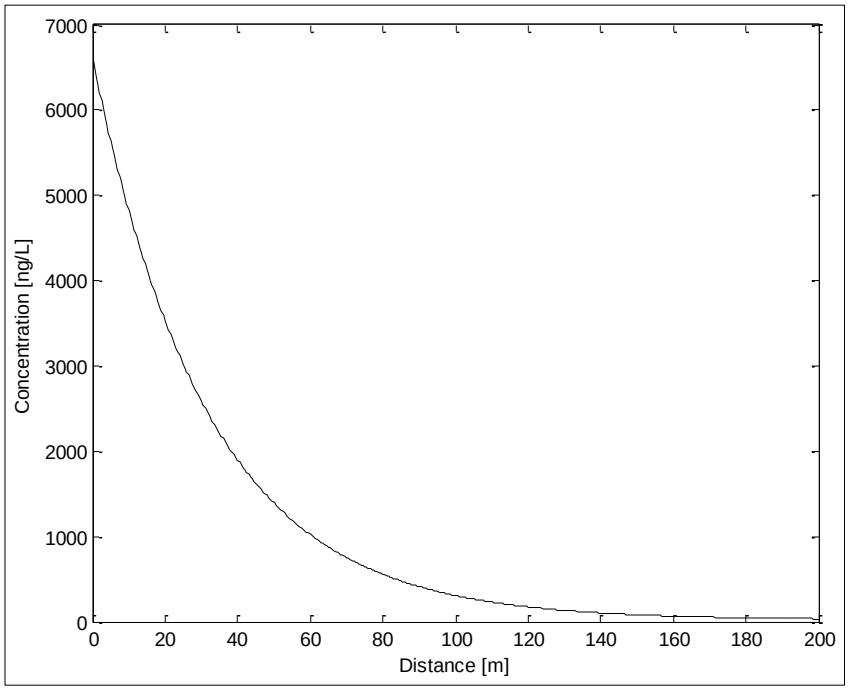

Figure 3: Upper limits analytical solution for first-type inlet condition for paracetamol.

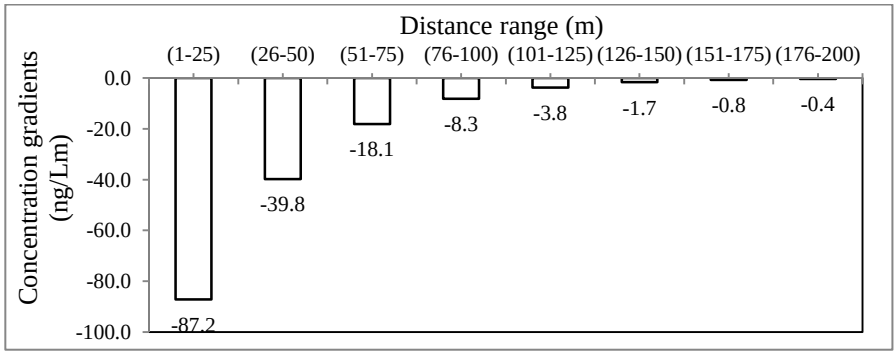

Figure 4: Spatial concentration gradient of paracetamol during dry season.

Figures 3, 4 and 5 show significant factors affecting the concentration gradient such as initial concentration, dilution, pumping off the water, irrigation and adsorption, which are noticed within 200 m once contamination occurs at a point source on the Msimbazi river. However, beyond 200 m, there is a slightly low concentration gradient change of about –0.00004 mg/L per meter. This concentration is too low as the technique used could not detect it, contrary to the model reported by Yadav et al. (2011). This observation is an average whereby river inlets, outlets and morphology are assumed not to affect the determination. The first-type inlet condition is preferred as it reflects the actual environmental impact and therefore, clearly portrays the probable ecological risks upon contamination. It shows a continuous predictable decrease of contaminant concentrations per unit distance transported. A decay curve is the transport behaviour of all the contaminants implying concentrations that are decreasing per unit distance (Guerrero et al. 2013). Time-based monitoring of injected contaminants in the flowing water generates a different concentration gradient curve, yet the levels decreased as time lapsed (Genuchten et al. 2013).

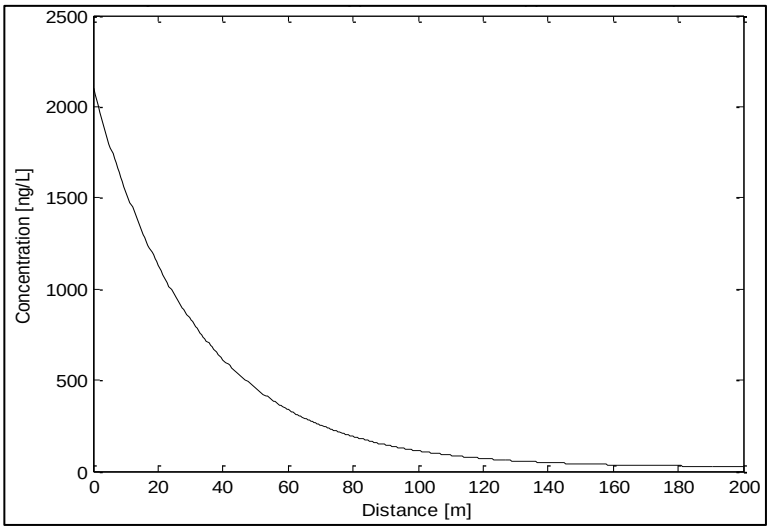

Figure 5: Analytical solution for first-type inlet condition for ibuprofen.

The implemented ADE model through the applications of mathematical principles enabled integration of mathematics in acquisition of analytical knowledge and prediction of transport behaviour of the contaminant in the Msimbazi river. These findings predicted the probable distance at which ECs are confidently quantifiable. A significant advantage of this model is the ability to predict minute concentrations of ECs that could have taken much time and resources in quantifying them in the laboratory. It is important to note that, once essential variables are taken care of, the model is useful in predicting the performance of a system. Outputs of this model forecasted the future of the Msimbazi river in terms of contaminants transport behaviour, particularly in predicting contaminant free zones along the river, identification of contaminants risky areas and prediction of possible harmless time for exposure which in this study are controlled by dilution effects. No previous report on un-expected phenomena such as contaminants transport behaviour at upstream and downstream have been reported for Msimbazi. Figure 3 shows clearly that the downstream water are much safer compared to upstream due to dilution effects.

In these findings, the transport behaviour of ECs deduced here include, but

not limited to; there being a noticeable concentration gradient when moving from uphill to downhill, no uniformity upon the location of occurrence of each emerging contaminant and the chance for occurrence of an emerging contaminant is independent of other ECs. There is high possibility of synergism among ECs since this was also observed when several ECs standards were mixed. While human activities such as sand excavation affect the spatial distribution of ECs, many sources of ECs are located uphill, while the lower lands are basically depositional areas. Moreover, the flow rate of the river determines the extent of exposure as it governs the amounts of contaminants transported per unit time. It was further observed that, in the dry season more ECs were detected and were transported due to pre-concentration caused by evaporation, less dilution and dehydration causing highly concentrated human excretes.

Sample collection during rainy season is obviously risky and unsafe, thus presents significant challenges in data collection. While all the sampling points were accessible during the dry season except sampling point MS12 due to presence of mud depositions, only point MS4 was accessible in the rainy season. And even this point had some challenges including motion sickness, fear of the associated risks, unpleasant smell of the water, and multiple reflection of sunlight on small water waves, unpredicted trenches, high flow rates and mass movements of bed-sand. Other points were not accessible because of high depth of the river, high flow rates, mass flow of bed-sand that increased the depth of the river, flooding of lower lands, and also that no casual labourers were ready to risk crossing the river in order to measure the surface area of the river at that point.

Conclusion and Recommendations

The investigation of 1-D transport behaviour of emerging contaminants along the Msimbazi river attained by using first-type inlet conditions is reported. The findings show that a continuous exponential gradient of contaminant transport between 0 to 200 m was predictable, while beyond it the gradient was changing at a negligible rate possibly due to infinite dilution of contaminants and concentration being below detection limits or due to the nature of the model. The preference of the first-type inlet condition is due to its ability to reflect the actual environmental conditions of the sample. These findings are useful in prediction of contaminants safe zones where human activities can continue with minimum exposure as well as least risk-free areas. By knowing either concentration or the distance from the point of contamination, this model can predict dynamic properties of Msimbazi river before engaging in furthering possible approaches such as remediation. In the future, 2-D and 3-D studies of emerging contaminants transport behaviour along this river will need similar analytical attention too.

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