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MODELLING SEDIMENT-ASSOICATED *Escherichia coli* IN A NATURAL RVIER: COMPARISON OF THE REVERSIBLE AND IRREVERSIBLE ADSORPTION

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Impairment of water quality caused by faecal pollution has a deleterious impact on human health and quality of life. The majority of known indicator bacteria in aquatic systems are associated with sediment and this association influences survival and transportability of the bacteria. However, how sediment processes affect the transport of indicator bacteria is unclear. In this study, a combination of steady-flow field experiments and mathematical modelling based on advection dispersion equation were used to better understand the processes controlling the fate of *Escherichia coli* (*E. coli*) in a stream, incorporating reversible and irreversible adsorption between sediment and *E. coli*. The simulation indicated that the reversible adsorption process produced a better result than the irreversible process when the concentration ratio of *E. coli* to total suspended solids was lower than 1×10^7 (CFU/g). The results also showed that calculated *E.coli* fluxes of reversible adsorption at each sampling points were closer to measured values than of irreversible adsorption.

Key Words: *water quality modelling, faecal pollution, bacteria, sediment, adsorption*

1. INTRODUCTION

A wealth of literature indicates the majority of faecal indicator bacteria in aquatic systems are associated with sediment and this association influences the survival and transport of bacteria. These processes, however, remain ill-defined and are key to model the fate of bacteria and control diffused pollution[1)-3)]. Two types of bacterial adsorption have been previously identified[4),5)]. The first is strong adsorption due to cellular appendages or extracellular polymers excreted from the cell[6)]. The second is weak adsorption mediated by van der Waals forces which collectively exceed repulsive forces. Bacteria tend to adsorb weakly to soil particles; they are not directly attached to the soil surface but are closely associated with it[7)]. Weak adsorption is considered to be a reversible process, whereas strong bonding mechanisms are thought to be irreversible. Reversible sorption processes are typically classified as kinetic adsorption and equilibrium adsorption. If the rate of bacterial adsorption to sediment is comparable to the flow speed, the sorption process should be treated as a kinetic process[8),9)]. On the other hand, if the time scale for sorptive interaction is smaller than that of advection, it should be treated as an equilibrium process [2)]. However, little report was concerned with different results from models that regard *E. coli* adsorption to sediment being reversible or irreversible. In the present study, we employed a combination of field experiments, lab experiments and numerical calculation to better understand the processes controlling the fate and movement of indicator bacteria in streams, and assessed the influence on the model simulation results when *E. coli* adsorption was considered to be reversible or irreversible.

2. MATERIALS AND METHODS

(1) In-situ field experiment

An experimental procedure was developed to simulate sediment-associated bacteria in steady-flow conditions. *E. coli* was used as a model bacterium. Bed sediments were collected from the studied river and indigenous *E.coli* was cultured. The experiment was conducted twice in a relatively straight section in the upstream portion of the Mizugaki Experimental Watershed, Japan[10]. A mixture of stream water, sieved sediment (mesh size < 1mm) and inoculum of cultured *E. coli* was pulse-loaded into the stream. The subsequent transport of the sediment and *E. coli* was monitored 10, 20 and 30 m downstream of the injecting point. Channel characteristics included cross-sectional area using the velocity-area method [11], longitudinal profile along the thalweg, and the water surface slope. Cross-sectional surveys were performed every 10 m; the slope of the water surface in this region was 0.8% of this stream.

Prior to the experiment, the mixture was prepared in a clean container and shaken for an equilibration period to allow for adsorption of the bacteria to the sediment[4]. A background sample collected at each sampling location was analysed for total suspended solids (TSS) and *E. coli*. After injecting the mixture, water samples were collected at time intervals ranging from 0.25 to 0.5 min at each location. Following each experiment, water and sediment samples were carried into the laboratory at cool temperature. *E. coli* analyses were performed on the day of the experiment using the membrane filtration (MF) technique[12]. We measured turbidity using an automatic turbidity meter, Compact-CLW (JFE ALEC). TSS concentrations were calculated by the turbidity-TSS relationship, which was obtained from water quality monitoring during base flow and storm flow periods in the study site. Particle size distribution was analysed using a Multisizer™ 3 electronic particle counter; the median particle diameter (D50) used in the field experiment was 20 μm.

(2) Adsorption experiment

An adsorption experiment was performed in the laboratory to determine the characteristics of *E. coli* adsorption to sediment. We prepared mixtures of pre-filtered river water, sieved sediment and cultured *E. coli* collected from the studied site, shook for 60 min at 200 rpm, and measured concentrations of sediment-associated *E. coli* by modifying the method[4),13] previously reported. Ten millilitres of the mixture was sampled and filtered by a membrane with 8 μm pore size. The filter was gently washed twice with sterile water then placed in a jar containing 100 mL of sterile water. Two or three drops of a non-toxic surfactant, Tween80™, were added to remove sediment and bacteria from the filter. The concentrations of *E. coli* in the filtrate and the resulting rinse water were determined by the MF technique. The adsorption was determined under two conditions. In the first, a mixture had similar *E. coli* and TSS concentrations as the peak concentrations of the in-situ field experiment at three sampling locations. In the second, the TSS concentration was maintained similar to the peak concentration at 10m, while the *E. coli* concentration was varied to 1.0, 0.5 and 0.1 times of the peak concentration at the 10 m.

3. MODEL DEVELOPMENT

The transport of *E. coli* and suspended sediments in river water was modelled using the 1D advection–dispersion equation (ADE) modified based on the framework proposed by Jamieson[4],

$$\frac{\partial C_{EC}}{\partial t} = D_{EC}\frac{\partial^2 C_{EC}}{\partial x^2} - v\frac{\partial C_{EC}}{\partial x} - k_1 C_{EC} \quad (1)$$

$$\frac{\partial C_{SS}}{\partial t} = D_{EC}\frac{\partial^2 C_{SS}}{\partial x^2} - v\frac{\partial C_{SS}}{\partial x} - k_2 C_{SS} + R_H \quad (2)$$

where C_{EC} and C_{SS} are the concentration of free-floating *E. coli* (CFU/m^3) and suspended sediment above background levels (g/m^3) respectively, D_{EC} and D_{SS} are dispersion coefficient for free-floating *E. coli* and suspended sediment (m^2/s), R_H is resuspension rate (g/ m^{2}s), x is the longitudinal distance along the channel (m), v is the velocity (m/s), k_1 is the *E. coli* inactivation constant (/s), k_2 is the net suspended sediment removal constant due to settling(/s). The value of the net suspended sediment removal constant, k_2, is computed as[4],

$$k_2 = \frac{V_s}{d} \quad (3)$$

where V_s is the sediment net settling velocity (m/s) and d is the water depth (m).

The two governing equations were solved using a mixed implicit finite-difference method. Both of the above ADEs require two boundary conditions and one initial condition for solution. The initial conditions are decided by the background concentration of TSS concentration and *E. coli* in this stream. The *E. coli* and TSS concentrations observed at 10 m were used for the boundary condition upstream. The boundary condition downstream was assumed to be a *Neuman*

Table 1 Parameters related to irreversible and reversible adsorption of *E.coli* on sediment.

Equation (5)	F (-)	0.3[a] (0.02[b])
Equation (8)	Q_{max} (CFU/g)	1.43×10^7
	k_L (ml/CFU)	0.0008
	R^2	0.965
Equation (9)	Q_{max} (CFU/g)	1.11×10^7
	k_L (ml/CFU)	0.00114
	R^2	0.983
Equation (10)	n (-)	0.632
	k_F (ml/g)	1.85×10^3
	R^2	0.971

[a] Nov.16th, [b] Nov.10th

type[4]. The concentration of sediment-associated *E.coli* within each segment at each time step was calculated as,

$$C_{SEC} = C_{ECsed} \times C_{SS} \quad (4)$$

where C_{SEC} is the concentration of sediment-associated *E. coli* (CFU/100 ml) and C_{ECsed} is the concentration of *E. coli* on sediment (CFU/g). C_{ECsed} was determined by considering irreversible [4] or reversible adsorption. For the irreversible adsorption, the fraction coefficients (F) did not vary. The concentration of *E. coli* on the injected sediment was computed as

$$C_{ECsed} = \frac{EC_{in} \times F}{SS_{in}} \quad (5)$$

where EC_{in} is the number of viable *E. coli* in the inoculums (expressed as CFU), F is the fraction of *E. coli* associated with sediment at injects point, as shown in **Table 1**, and SS_{in} is the mass of suspended sediment in the inoculum (g). As regards the reversible adsorption, two types of equation were adopted by fitting with the experimental results.

4. RESULTS AND DISCUSSION

(1) Adsorption experiment results

In order to describe equilibrium adsorption, the Langmuir and Freundlich models are usually used[2].

$$\text{Langmuir model: } C_{ECsed} = \frac{Q_{\max} k_L C_{EC}}{1 + k_L C_{EC}} \quad (6)$$

$$\text{Freundlich model: } C_{ECsed} = k_F C_{EC}{}^n \quad (7)$$

where $Q_{\max}$ is the maximum adsorption quantity of *E. coli* on sediment (CFU/g); k_L (ml/CFU) and k_F (ml/g) are equilibrium adsorption coefficients; *n* is the index factor. The results of adsorption experiment were analysed as Langmuir and Freundlich models using the linear regression method. Two linear regression methods were used for the Langmuir model, which yielded the following equations:

$$\frac{C_{EC}}{C_{ECsed}} = \frac{1}{Q_{\max}} \cdot C_{EC} + \frac{1}{Q_{\max} \cdot k_L} \quad (8)$$

$$\frac{1}{C_{ECsed}} = \frac{1}{Q_{\max} \cdot k_L} \cdot \frac{1}{C_{EC}} + \frac{1}{Q_{\max}} \quad (9)$$

Coefficients of k_L and $Q_{\max}$ can be deduced from the rate of slope and intercept of trend lines, which were taken from the graph between C_{EC}/C_{ECsed} and C_{EC} or between $1/C_{ECsed}$ and $1/C_{EC}$ by the method of least squares. The Freundlich model was converted to the linear style by use of a logarithm,

$$\log C_{ECsed} = n \log C_{EC} + \log k_F \quad (10)$$

where k_F and n were determined by plotting $\log C_{ECsed}$ and $\log C_{EC}$. The correlation coefficients (R) of the models are also listed in **Table 1**. The fitting results of adsorption experiments are depicted in **Fig. 1**.

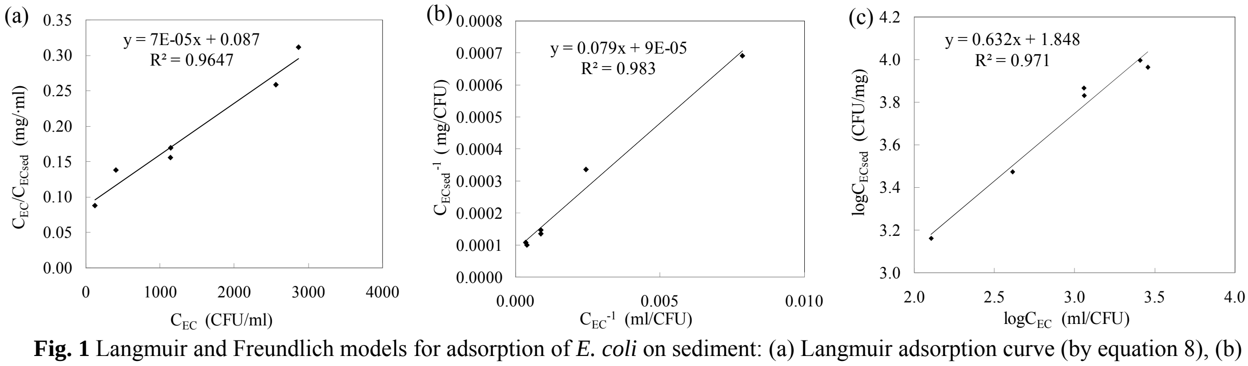

Fig. 1 Langmuir and Freundlich models for adsorption of *E. coli* on sediment: (a) Langmuir adsorption curve (by equation 8), (b) Langmuir adsorption curve (by equation 9) and (c) Freundlich adsorption curve (by equation 10).

Table 2 Conditions of the field experiment.

Items	Nov.16th	Nov.10th
d: Average depth (m)	0.11	0.10
v: Average velocity m/s)	0.12	0.11
River flow (m^3/s)	0.010	0.011
EC_{in}: *E. coli* input (CFU)	1.29×10^9	1.09×10^{11}
SS_{in}: Tss input(Kg)	0.1	0.2
Maximum particle diamete(μm)	150	150
Median particle diameter (μm)	20	20
Stream water input(m^3)	0.01	0.01
Background TSS (mg/L)	3.2	4.2
Background *E. coli* (CFU/100ml)	12	10

Table 3 Calibrated parameters of the model.

Parameters	TSS	*E. coli*
D_{EC}: Dispersion coefficient (m^2/s)	0.08	0.1
V_s: Set velocity (cm/s)	0.012	-
R_H: Resuspension rate (g/ m^2s)	0.0035	-

Table 4 Factors of total *E.coli* transport in experiment and modelling.

Point	Type of data		Peak value (CFU/ 100ml)	Time to peak (sec)	RMSE (CFU/ 100ml)	Cumulative flux (CFU)
20 m	Experiment		75655	143	-	1.07×10^9
	Modelling	Irreversible	75200	145	7500	1.21×10^9
		Reversible	74050	145	6700	1.11×10^9
30 m	Experiment		46883	255	-	9.45×10^9
	Modelling	Irreversible	57300	230	10300	1.19×10^9
		Reversible	48900	240	4200	1.05×10^9

From the regression analysis, it was evident that *E. coli* adsorption to the sediment was compliant with the Langmuir and Freundlich models. However, equation 9 was most significantly compliant. So, the coefficients produced from equation 9 were chosen for incorporation into the ADE model to compare the results of reversible adsorption. The values of Q_{max} and K_L in the Langmuir adsorption model were 1.11×10^7 CFU/g and 0.0011 ml/CFU, respectively, similar to previous data [14].

(2) Model simulation

a) Calibration

The field experiment data obtained on November 16 was used for the model calibration. Depths, *E. coli* input and sediment input were listed in **Table 2**. The F value was 0.3 as shown in **Table 1**. The value of die-off coefficient used in the calibration was 0.02 h^{-1} which is typical for *E. coli* in a freshwater environment[13),14)]. *E. coli* inactivation had, however, a negligible impact on the results due to the short duration of each field experiment (0.5 h). Several parameters (dispersion coefficients, net settling velocities and resuspension rate) were varied to obtain model outputs that best predicted experimental observations at 20 m and 30 m. The calibrated parameter values are presented in **Table 3.**

The following calibration procedure was used to determine values of model parameters that were not directly measured. First, the dispersion coefficient, set velocity and resuspension rate were varied to obtain model outputs, which closely agreed with observed TSS concentrations at the second and third sampling locations, TSS calibration results are shown in **Fig. 2**. Second, D_{EC} was varied to obtain model outputs that closely agreed with observed *E. coli* concentrations at downstream sampling locations considering the irreversible and reversible adsorption, as shown in **Fig. 3** and **Fig. 4** respectively. The predicted concentrations of sediment-associated *E. coli* have also been included in the figures.

The timing and spread of the TSS were simulated reasonably using the calibrated parameter values listed in **Table 3**. The D_{SS} , 0.08 m^2/s, was agreed with previous results[4)] with similar particle sizes and hydrodynamic conditions. The net settling velocity, Vs, 0.012 cm/s, represented the same order value as counting by Stokes fall velocity[11)] which was 0.034 cm/s. The difference was supposedly caused by particle densities, shape factors or effective diameter.

The adequacy of model simulations was determined qualitatively by visual comparison in Fig4, peak value and time to peak, quantitatively by computing root mean square error (RMSE) between observed and calculated concentrations and total *E.coli* flux (**Table 4**). The results were better at the recession limbs than rising limbs with both models. However, RMSE of reversible adsorption was smaller than that of irreversible adsorption at both sampling locations. Comparing with results of two models at 30 m, a better simulation yielded for peak as well as rising and recession limbs of total *E. coli* when reversible adsorption was considered. The calculated total *E.coli* flux of reversible adsorption at each sampling location was closer to the measured flux. The better results in reversible adsorption simulation at rising limb were constantly deduced by increase of C_{ECsed} , however, the value did not vary in irreversible adsorption. Increasing C_{ECsed} means adsorbed quantity of *E.coli* to TSS increased, more *E.coli* consequently settled down to the river bed with TSS during rising limb.

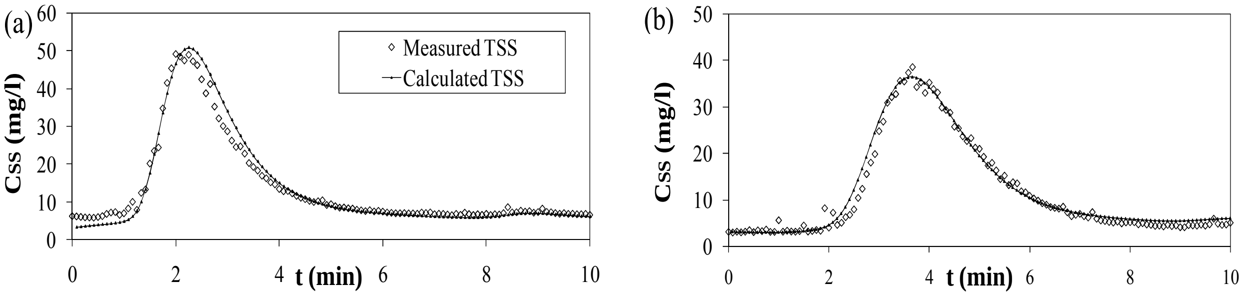

Fig. 2 Simulation of TSS at (a) 20 m and (b) 30 m downstream.

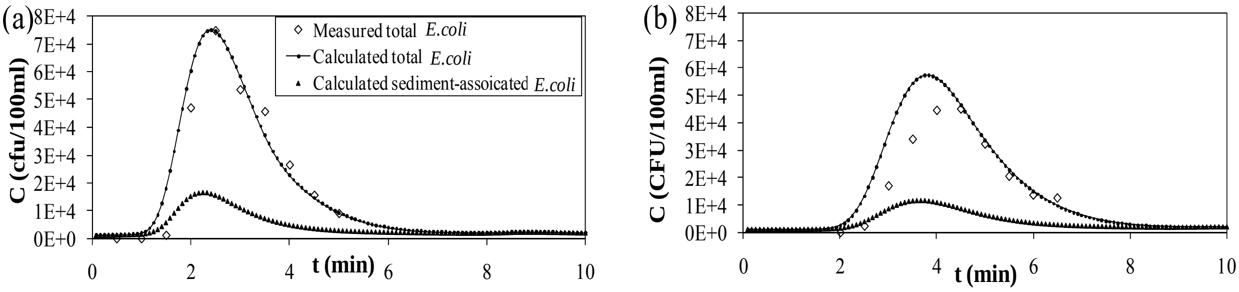

Fig. 3 Irreversible adsorption simulation results of *E. coli* at (a) 20 m, and (b) 30 m downstream.

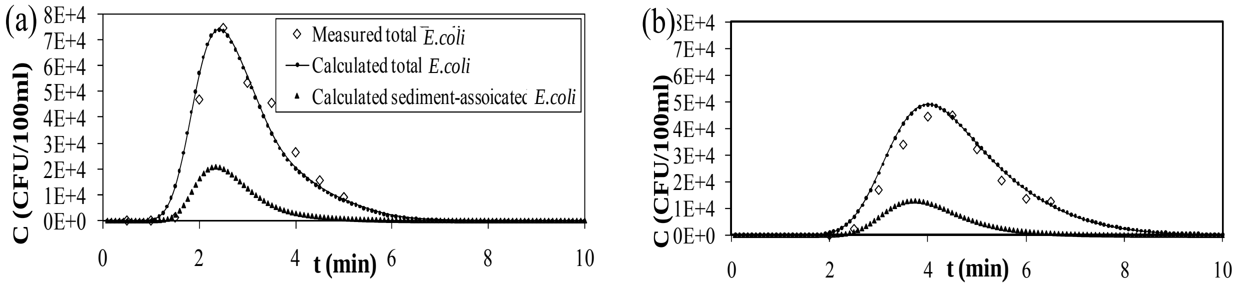

Fig. 4 Reversible adsorption simulation results of *E. coli* at (a) 20 m and (b) 30 m downstream.

b) Validation

The experimental data obtained on November 10 was used for model validation. Depths, partition fractions, *E. coli* input and sediment input were considered. Values of these parameters for the experiment are listed in **Table 2**. Validation results did not show significant difference between two models, except for a slightly better result in reversible adsorption during recession period. The ratio of *E. coli* concentration to TSS concentration was probably very high within a 30 m reach, even higher than the maximum adsorption quantity ($Q_{\max}$) in **Table 1** obtained from the adsorption experiment, thus little amount of *E.coli* was able to desorb from the sediment. This result indicates the importance of indentifying the parameter such as $Q_{\max}$ which is crucial to determine whether the adsorption process is reversible or not.

c) Sensitivity analysis

The parameters related to adsorption in the model were determined by the experiment. Sensitivity analysis was performed to assess the influence of the parameters (F , $Q_{\max}$ and k_L) on the simulation results. If sensitive parameters are not identifiable, there will be a high level uncertainty of parameter values concerned and a consequent high level uncertainty of output. The analysis indicated that Q_{max} and k_L were sensitive to the concentration of total *E.coli* within a certain range. The increase of $Q_{\max}$, in the range of 10^6 ~10^8 (CFU/g), resulted in 20% decrease of peak value. The change in k_L varied 10% of peak concentration of total *E.coli* when k_L varied in the range of 10^{-5} ~10^{-1} (CFU/g). These two parameters can be calibrated through the model in this study. On the other hand, F was not sensitive to the concentration of total *E.coli* through the analysis, and hence it should be determined by experimental method to simulate more realistic process of bacterial transport in river system.

5. CONCLUSIONS

In this research, the transport of sediment-associated *E. coli* and the adsorption characteristics of bacteria on sediment were investigated through a combination of field experiments, lab experiments and mathematical modelling. Fundamental model parameters that control the fate of bacteria within a river water column of were determined. The major conclusions were as follows:

(1) The 1D advection-dispersion equation including the processes of adsorption, deposition and resuspension provided reasonable simulation of *E. coli* transport under steady-flow conditions.

(2) The characteristics of *E. coli* adsorption to sediment were compliant with both Langmuir and Freundlich model, however, regression result was better with Langmuir type. Parameters of the Langmuir model such as Q_{max} and K_L were determined.

(3) The reversible adsorption model simulated better results for the concentrations and cumulative flux of total *E.coli*.

(4) When the ratio of concentration of *E. coli* to TSS concentration is higher than the maximum adsorption quantity, the reversible adsorption had no affect on *E.coli* transport.

(5) The maximum adsorption quantity and adsorption coefficient were sensitive to the peak value of total *E.coli* within a certain range and calibrated through the model.

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