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Modeling the release of *Escherichia coli* from soil into overland flow under raindrop impact

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ABSTRACT

Pathogen transport through the environment is complicated, involving a variety of physical, chemical, and biological processes. This study considered the transfer of microorganisms from soil into overland flow under rain-splash conditions. Although microorganisms are colloidal particles, they are commonly quantified as colony-forming units (CFUs) per volume rather than as a mass or number of particles per volume, which poses a modeling challenge. However, for very small particles that essentially remain suspended after being ejected into ponded water and for which diffusion can be neglected, the Gao model, originally derived for solute transfer from soil, describes particle transfer into suspension and is identical to the Hairsine–Rose particle erosion model for this special application. Small-scale rainfall experiments were conducted in which an *Escherichia coli* (*E. coli*) suspension was mixed with a simple soil (9:1 sand-to-clay mass ratio). The model fit the experimental *E. coli* data. Although re-conceptualizing the Gao solute model as a particle suspension model was convenient for accommodating the unfortunate units of CFU ml⁻¹, the Hairsine–Rose model is insensitive to assumptions about *E. coli* per CFU as long as the assumed initial mass concentration of *E. coli* is very small compared to that of the soil particle classes. Although they undoubtedly actively interact with their environment, this study shows that transport of microorganisms from soil into overland storm flows can be reasonably modeled using the same principles that have been applied to small mineral particles in previous studies.

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1. Introduction

One of the major themes in Dr. Sposito's work has been the role of colloids in the environment (e.g., Sposito 1983; Sposito and Schindler 1987; Sposito 1989; Heil and Sposito 1993a, 1993b, 1995; Sposito 1993; Chang and Sposito 1994; Chorover and Sposito 1995; Sposito and Malengreau 1998, among many more). Here we add to those pioneering studies by looking at the interactions between bio-colloids (bacteria) and raindrops to better understand the transport processes facilitating the exchange of colloids from soil and into overland flow.

The World Health Organization (WHO, 2011) considers water borne pathogens to be the most important water quality risk to address in the foreseeable future. Indeed, pathogens impair more kilometers of US rivers and streams than any other pollutant for US water bodies officially listed as impaired or failing to meet

water quality standards for their designated uses (US EPA, 2010). Storm water is a primary transport pathway for many pathogens and increased concentrations are often correlated with large precipitation and snowmelt events (Falbo et al., 2013; Jamieson et al., 2003; Kistemann et al., 2002; Pettibone and Irvine, 1996; Simon and Makarewicz, 2009a; Simon and Makarewicz, 2009b; Traister and Anisfeld, 2006). An improved understanding of the transport mechanisms of microorganisms, like *Escherichia coli* (*E. coli*), will help in developing strategies for mitigating pathogen loads between the landscape and surface waters.

Previous studies of *E. coli* transport primarily focused on the attachment of *E. coli* to solid particles (Guber et al., 2005a; Guber et al., 2005b; Muirhead et al., 2006a; Muirhead et al., 2006b; Oliver et al., 2007), the transport of *E. coli* through soil columns (Smith et al., 1985; Barton and Ford, 1995; Barton and Ford, 1997; Schäfer et al., 1998; Powelson and Mills, 2001; Sherwood et al., 2003; Olson et al. 2005; Jiang et al., 2007; Truhlar et al. 2015), or the transport of *E. coli* in runoff over bare soil or through vegetated buffers (Muirhead et al., 2006a; Ferguson et al., 2007).

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Several studies used rainfall experiments to investigate rain-driven erosion of manure slurry amended soil or cowpats and the associated transfer of *E. coli* into runoff (Zyman and Sorber, 1988; Roodsari et al., 2005; Muirhead et al., 2005; 2006c; Ferguson et al., 2007; Kouznetsov et al., 2007). The experiments of Ferguson et al. (2007) included bare soil treatment, first as a control and then receiving *E. coli* enriched cowpats. Their objective was to determine how vegetation and microbe size influence microbial transport in runoff through statistical comparisons, i.e., the physical processes were inferred rather than explicitly explored.

We hypothesize that microorganism transfer between soil and overland flow during rainfall can be explained by the same underpinning processes that have been used to explain the transfer of other small particles (e.g., clay). We combine laboratory experiments and mechanistic models to test this hypothesis. This process has not been well described in previous research although it is important because it initiates nonpoint source pathogen pollution. We designed a simple experiment to investigate the rain-splash erosion of *E. coli* from soil into overland flow and applied two mechanistic models.

E. coli (roughly a 1 μm diameter and 2 μm long rod - Neidhardt et al., 1990) is a colloidal particle, comparable in size to kaolinite clay particles (ranging from 0.1 μm to 2 μm - Mackinnon et al., 1993). However, it is difficult and expensive to quantify *E. coli* as either a number-of-cells or a mass-of-cells per volume of sample, in part due to possible or even likely aggregation. The simplest way to measure *E. coli* (and other bacteria) is to culture samples on agar plates and enumerate the concentration as colony-forming units (CFUs) per volume of sample. While this is a fairly standard and reasonably repeatable measurement technique, it is difficult to relate CFUs to bacteria numbers or masses. This is unfortunate, because the well-known Hairsine–Rose (1991) model is a good representation of small particle transfer from soil into overland runoff (e.g., Heilig et al., 2001; Gao et al., 2003; 2005), but it requires the initial mass ratios for all particle classes and, if we consider bacteria a particle class, this information is not known. Gao et al. (2004; 2005) adapted the Hairsine–Rose rain-impact concept to derive a solute transfer model. Because both models are predicated on the same fundamental principles, i.e., rain-impact ejects material from the soil surface into the overland flow and the depth of impact is discrete (described as either shield - e.g., Heilig et al. 2001 - or exchange layer - e.g., Gao et al. 2004), we suggest that they are identical when applied to colloidal transfer between soil and overland runoff.

2. Colloid suspension models

Bacteria are often classified as bio-colloids, which are somewhat different from the definition of particles used in erosion modeling, in part because they have near-neutral buoyancy and do not settle out of suspension very rapidly. However, they are obviously not solutes either. However, we think that with the appropriate caveats, the Hairsine–Rose erosion model and the Gao solute model are equivalent when subjected to the conditions required to model colloids, e.g., settling velocity and diffusion are negligible (See Tables 1 and 2 for all variables and parameters). Here we describe the two models as adapted to colloids (assuming constant ponding depth and rain rate), and then show that they are identical for our application.

2.1. Hairsine–Rose model

Following Hairsine and Rose (1991), soil can be characterized by equivalent mass-particle classes. Including bacteria as a mass-particle class introduces an almost certain unknown, so we propose normalizing all classes to the bacteria (*E. coli*) class, $i = 1$. For

the sake of this derivation and, indeed, our experimental design as described later, let us arbitrarily assume n mass-classes of clay and f mass-classes of sand (other particle classes could be included, but we will assume a simple, *E. coli*-clay-sand soil here). So the total particle mass-classes, $I = 1 + (1 + f)n$. We use a coefficient, M_{cfu} , to convert CFUs to mass of *E. coli*; this also establishes how many classes of clay and sand there will be.

The Heilig et al. (2001) version of Hairsine–Rose model adopts the assumption that once ejected by the raindrops, particles with low settling velocity, e.g., *E. coli* and clay, do not settle out of the ponded water (overland flow). Particles with high settling velocities, like sand, settle out of the overland flow quickly and deposit on the soil surface to form a shield layer which protects the soil underneath it from further erosion.

With the above assumptions, for the *E. coli* and clay classes (settling velocities ≈ 0), the suspension concentrations in overland flow are non-zero:

$$d_w \frac{dc_i(t)}{dt} = \frac{ap}{I} (1 - H(t)) - pc_i(t) \quad (1)$$

where $c_i(t)$ (g/ml) is the concentration of *E. coli* or of each class of clay in the suspension at time t , d_w (cm) is ponding water depth, a (g/ml) is the soil detachability, p (cm/min) is rainfall intensity, I is the total number of equal-mass particle classes, and $H(t)$ is the fraction of soil protected by shield layer at time t , with no erosion when $H = 1$. The first term in the square brackets represents the rate at which i -class particles are ejected from the soil and the second term represents the rate at which they wash away in the overland flow. Because the rainfall-runoff is at steady state, the runoff rate is equal to the rainfall intensity, p .

The mass of the deposited sediment is the sum of the deposited mass of all the sand classes,

$$M_d = \sum_{i=n+2}^{(1+f)n+1} M_{di} \quad (2)$$

Following Hairsine and Rose (1991), the rate of accumulation of the deposited sediment, $\frac{dM_d(t)}{dt}$, is proportional to the rate at which clay and *E. coli* are ejected from the soil (Eq. 1). Therefore, it can be expressed as:

$$\frac{dM_d(t)}{dt} = \frac{fn}{I} ap [1 - H(t)] \quad (3)$$

The $H(t)$ term in Eqs. 1 and 3 is given by Sander et al. (1996) as:

$$H(t) = \frac{M_d(t)}{M_d^*} \quad (4)$$

where M_d^* (g/cm²) is the mass of shield layer (sand) per unit area at complete shielding, i.e., when raindrop impact is prevented by the shield layer from ejecting underlying particles, i.e., $H = 1$.

We define N_e^* (g/cm²) and M_c^* (g/cm²) to be the total ejected CFUs of *E. coli* and mass of clay per unit area, respectively, when $H = 1$:

$$N_e^* = \int_0^T pc_{cfu}(t) dt \quad (5)$$

$$M_c^* = \int_0^T pc_c(t) dt \quad (6)$$

where $c_{cfu}(t)$ and $c_c(t)$ are the concentration of *E. coli* CFUs and concentration of clay at time t in the ponded water, respectively, and T is the time at which all the erodible clay has washed out of the experiment.

From the nonselective detachment, we obtain the following ratios:

$$N_e^*(t)M_{cfu} : M_c^* : M_d^* = 1 : n : fn \quad (7)$$

Table 1
Summary of modeling parameters and the ways they were determined.

Notation	Definition (Unit)	Value				
		run 1	run 2	run 3	run 4	run 5
a	Soil detachability ^c (g/ml)	4.500	0.350	0.800	0.450	0.450
C_0	Initial concentration of <i>E. coli</i> in the suspension added to the soil ^c ($\times 10^6$ CFU/ml)	2.29	7.05	13.4	3.20	3.17
d_e	Exchange layer depth ^c (cm)	0.294	0.175	0.085	0.180	0.126
d_w	Ponding water depth ^a (cm)	0.825	0.800	0.900	0.950	0.950
I	Total number of normalized classes (when 1 colony = 1 cell)	2,450,891	798,451	420,811	1,757,421	1,764,711
I	Total number of normalized classes (when 1 colony = 1000 cell)	2451	801	421	1761	1761
K_p	Partition coefficient for dissolved and adsorbed <i>E. coli</i> (ml/g)	0	0	0	0	0
M_e^*	Eroded clay per unit area ^b (g/cm ²)	0.0453	0.0270	0.0131	0.0277	0.0194
n	Number of classes representing clay (when 1 colony = 1 cell)	245,089	79,845	42,081	175,742	176,471
n	Number of classes representing clay (when 1 colony = 1000 cell)	245	80	42	176	176
N_e^*	Eroded <i>E. coli</i> per unit area ^b ($\times 10^5$ CFU/cm ²)	1.947	3.563	3.277	1.660	1.160
p	Rainfall intensity ^a (cm/min)	0.28	0.276	0.26	0.26	0.24
θ	Soil water content by volume at saturation ^a	0.288	0.288	0.288	0.288	0.288
ρ_b	Bulk density of the soil ^a (g/cm ³)	1.543	1.543	1.543	1.543	1.543

^a directly measured, see Section 2 for details

^b calculated from directly measured values, explained in Section 3

^c calculated or calibrated, elaborated in Section 4

Table 2
Summary of variables and complex parameters considered in Hairsine–Rose erosion and Gao solute models.

Notation	Definition (Unit)
$c_c(t)$	Estimated concentration of clay in the suspension at time t (g/ml)
$c_e(t)$	Estimated concentration of <i>E. coli</i> in overland flow at time t (g/ml)
C_e	Gao model estimated concentration of <i>E. coli</i> in exchange layer pore water (CFU/ml)
$c_{efu}(t)$	Measured concentration of <i>E. coli</i> in overland flow at time t (CFU/ml)
$c_i(t)$	Estimated concentration of <i>E. coli</i> or each class of clay in the suspension at time t (g/ml)
C_w	Gao model estimated concentration of <i>E. coli</i> in overland flow (CFU/ml)
e_r	Shown in Eq. 15
$H(t)$	Fraction of soil protected by shield layer at time t
M_{cfu}	Conversion factor from CFU to g (g/CFU)
$M_d(t)$	Cumulative masses of ejected sand per unit area at time t , also equal to the mass of sand deposited on the surface per unit area at time t
M_d^*	Mass deposited sand (shield layer) at which no additional underlying soil can be eroded (g/cm ²)
t	Time (min)
T	The time at which all the erodible clay has washed out of the experiment (min)
z	Vertical axis, as shown in Fig. 1
α	Shown in Eq. 15

Inserting Eq. 4 into Eq. 3 and solving with the initial condition $M_d(0) = 0$ yields:

$$H(t) = 1 - \exp\left(-\frac{fna}{IM_d^*}pt\right) \quad (8)$$

Inserting Eq. 8 into Eq. 1 and solving it with the initial condition $c_i(0) = 0$ yields the concentration of *E. coli* or concentration of each clay class:

$$c_i(t) = \frac{\frac{a}{Id_w}}{\frac{1}{d_w} - \frac{fna}{IM_d^*}} \exp\left(-\frac{p}{d_w}t\right) \left\{ \exp\left[\left(\frac{1}{d_w} - \frac{fna}{IM_d^*}\right)pt\right] - 1 \right\} \quad (9)$$

Applying Eq. 7 to Eq. 9 we can obtain the concentrations of clay, $c_c(t)$, and *E. coli*, $c_e(t)$:

$$c_c(t) = n \frac{\frac{a}{Id_w}}{\frac{1}{d_w} - \frac{fna}{IM_d^*}} \exp\left(-\frac{p}{d_w}t\right) \left\{ \exp\left[\left(\frac{1}{d_w} - \frac{fna}{IM_d^*}\right)pt\right] - 1 \right\} \quad (10)$$

$$c_e(t) = n \frac{N_e^* \times M_{cfu}}{M_c^*} \frac{\frac{a}{Id_w}}{\frac{1}{d_w} - \frac{fna}{IM_d^*}} \exp\left(-\frac{p}{d_w}t\right) \times \left\{ \exp\left[\left(\frac{1}{d_w} - \frac{fna}{IM_d^*}\right)pt\right] - 1 \right\} \quad (11)$$

Note that diffusion and adsorption are not considered in this particle model. We discuss elsewhere possible *E. coli*-clay aggregation.

2.2. Gao model

The Gao solute model (Gao et al., 2004; 2005; Walter et al. 2007) is predicated on the same underpinning concepts as the Hairsine–Rose model, although the shield layer is re-conceptualized as an exchange layer where soil solutes are ejected into the overland flow. However, the exchange layer is conceptually different than the shield layer. The depth of shield layer increases from zero to its final depth as the rainfall continues (Heilig et al., 2001). And the final depth of the shield layer conveniently indicates the depth of the exchange layer. In converting the Gao model into a particle/colloid suspension model, we assume negligible diffusion of colloids into the exchange layer from the underlying soil. The Gao model is conceptually similar to the Hairsine–Rose model but instead of characterizing the system as a set of equal masses-classes of particles this model performs a “mass” balance of the solutes ejected from the mixing layer into the ponded water (the layers of the Gao model are labeled in Fig. 1). This minor difference allows modelers to adopt units of CFU ml⁻¹ without having to define a more specific, physically-based definition of CFU, e.g., a mass or number of bacteria cells associated with a CFU. Accepting the vaguely-defined concentrations of CFU ml⁻¹, we can re-write the Gao model in terms of non-diffusing colloids:

$$\text{Ponded water: } d_w \frac{dC_w}{dt} = e_r C_e - p C_w \quad (12)$$

$$\text{Exchange layer: } \alpha d_e \frac{dC_e}{dt} = -e_r C_e \quad (13)$$

Fig. 1. Experiment set-up, adapted from Gao et al. (2004, 2005). The ponded water (d_w) and exchange layer (d_e) as defined in the Gao model are labeled next to the column.

$$\text{where } e_r = \frac{ap\theta}{\rho_b}, \alpha = \rho_b K_p + \theta, \quad (14)$$

$$\text{Initial conditions: } C_e = \frac{\theta}{\alpha} C_o, C_w = 0 \quad (15)$$

where C_e and C_w are concentrations of *E. coli* in exchange layer pore water and overland flow (a.k.a., ponded water), respectively; C_o is the initial concentration of *E. coli* in the suspension added to the soil; d_e (cm) is exchange layer depth; t (min) is time; p (cm/min) rainfall intensity; d_w (cm) is ponded water depth; a (g/cm³) soil detachability; θ is volumetric soil water content at saturation; ρ_b (g/cm³) is bulk density of the soil; and K_p (ml/g) is the soil-water partition coefficient for *E. coli* adsorbed to soil. We assume all pore water ejected from the exchange layer is replaced with clean rainwater (see Gao et al. 2004 for a full analysis and justification of this assumption). Here we assume $K_p = 0$ and we revisit this assumption in our discussion.

The above equations can be solved analytically for C_e and C_w (details are shown in Appendix A) so that the Gao colloid suspension model takes the form of:

$$C_e = C_o \exp\left(-\frac{ap}{\rho_b d_e} t\right) \quad (16)$$

$$C_w = C_o \frac{ap\theta}{\rho_b d_w \left(\frac{p}{d_w} - \frac{ap}{\rho_b d_e}\right)} \left\{ \exp\left[\left(\frac{p}{d_w} - \frac{ap}{\rho_b d_e}\right) t\right] - 1 \right\} \exp\left(-\frac{p}{d_w} t\right) \quad (17)$$

2.3. Relation between the models

We contend that these two models, when applied to the special case of non-settling, non-diffusing colloid transfer between soil and ponded water are identical. The fundamental, conceptual difference is that the Hairsine–Rose model simulates the development of the shield layer, which grows as $1 - \exp(-pt)$ (Eq. 8), while the Gao model simulates the flushing of the exchange layer, which is exponentially depleted as a function of pt (Eq. 16). We can show the equivalence of these two concepts mathematically. Note that

$\frac{M_d(t)}{M_d^*} = 1 - \frac{C_e}{C_o}$, so Eq. 3 of the Hairsine–Rose model can be expressed as:

$$\frac{dM_d^* \left(1 - \frac{C_e}{C_o}\right)}{dt} = \frac{fn}{I} ap \cdot \frac{C_e}{C_o} \quad (18)$$

which can be rearranged:

$$\frac{dC_e}{dt} = -\frac{fn}{IM_d^*} ap \cdot C_e \quad (19)$$

Note that $\rho_b d_e$ in the Gao model is the mass of dry soil (n parts clay and fn parts sand) in exchange layer per unit area and M_d^* is the mass of sand in exchange layer per unit area after all the fine particles have been flushed out, so $\frac{IM_d^*}{fn}$ is also the mass of dry

soil in exchange layer per unit area. Substituting $\frac{IM_d^*}{fn}$ with $\rho_b d_e$, Eq. 19 results in Eq. 13 from the Gao model. Similarly, recognizing that $c_i(t) = M_{cfu} C_w$, we can rewrite Eq. 1 from the Hairsine–Rose model as Eq. 12 of the Gao model. Also, initial conditions are equivalent, i.e., $M_d(0) = 0$ used to solve Eq. 3 is equivalent to $C_e(0) = C_o$ and the $c_i(0) = 0$ used to solve Eq. 1 is equivalent to $C_w(0) = 0$ in the Gao model. As mentioned above, the Gao exchange layer has the same depth as the fully developed Hairsine–Rose shield layer, therefore, the boundary condition that no substances in the soil underneath the exchange layer/shield layer are transported into or through the exchange layer/shield layer is the same for the two models as well.

So, the two models used in this study are identical (with the equivalent governing equations, and equivalent initial and boundary conditions) for non-settling and non-diffusing suspended substances. However, we will apply both to our experimental data as an additional verification.

3. Experimental design

The experiment set-up (Fig. 1) was similar to that utilized by Gao et al. (2004, 2005) and Wang et al. (2013). Four holes in the side of our soil column kept the water level constant. The oscillating rain simulator generated uniform rainfall over the soil column. This design is consistent with the assumptions we used in

our model descriptions above, i.e., the ponding water depth and rain intensity are constant throughout the experiment. The soil is a simple 9:1 mixture of dark sand and kaolinite clay ($f=9$), equivalent to loamy sand soil; we assume that sand settles out of the ponding water very rapidly and the clay effectively remains in suspension once ejected from the soil. We also assume *E. coli* remains in suspension once ejected from the soil.

The soil mixture consisted of 225 g sand (University Sand & Gravel, Brooktondale, NY) and 25 g clay (Kaolinite, Englehard Corp, NJ) in a 7.6 cm diameter cylindrical acrylic (a.k.a. Plexiglas) column (Fig. 1). This soil mixture is similar to that used by Gao et al. (2004, 2005) with the exception of a slightly larger sand particle size used in this experiment (250–300 μm versus 198–212 μm).

E. coli ATCC 25922, a nonpathogenic surrogate of pathogenic *E. coli* O157: H7 (Muirhead et al., 2006b; Salleh-Mack and Roberts, 2007; Sauer and Moraru, 2009), was grown in Tryptic Soy Broth (TSB; Becton, Dickinson and Company, Sparks, MD) for 18 h at 37 °C on a shaking table. Two milliliters of this culture was then mixed with 60 ml 1.08% potassium chloride (KCl) solution. The resulting suspension and the soil mixture were homogenized, and poured into a column.

Note that we used 1.08% KCl solution (ionic strength = 0.145 M) as a substitute of physiological saline (0.85% NaCl (ionic strength = 0.145 M) or 0.9% NaCl (ionic strength = 0.154 M)) to serve as a blank to keep the osmotic pressure outside the cell balanced with that inside the cell, as NaCl would disperse the clay. The 1.08% KCl is equivalent to 5.14 g Cl^-/L , which has much smaller ionic strength than the solution Gao et al. (2004, 2005) used (29.82 g Cl^-/L) in both of their studies, and both of which were not affected by clay aggregation. Moreover, in Sections 4 and 6 we will show that our models work well whether there were *E. coli*-clay micro-aggregates or not. Briefly, for the Gao model, each cell or micro-aggregate is regarded as one CFU, and we used CFU from the beginning to the end; while for the Hairsine–Rose model, we have to assume a mass of cells associated with each CFU; we initially assume 1 cell = 1 CFU, but we test model's sensitivity to this assumption.

The pre-saturated soil column was packed on a shaking table. The ponded suspension was poured off. The column was then placed under the rainmaker and *E. coli*-free 1.08% KCl solution was gently added to pre-pond the experimental soil without disturbing the soil. The pre-ponding (1) set the initial condition of a particle concentration of zero in the ponded water and (2) establish an initial ponded water depth that was constant throughout each experiment, i.e., steady-state hydrologic conditions. Another 0.5 ml sample was extracted from the clean ponded water to characterize the particle concentration in the runoff at $t=0$.

We used sterilized rainwater (1.08% KCl solution) so that we did not introduce any unknown *E. coli*. A Marriott bottle was used (Fig. 1) to seal the rainwater from the environment while maintaining a constant rainfall rate.

The study area was protected with an umbrella until steady rainfall was established. A timer was started when the umbrella was removed and rainfall was continuous over the duration of each experiment. Samples of 0.5 ml were taken from the runoff at varying intervals depending on how rapidly we anticipated the changes in concentration based on trial experiments; the time between samplings changed at 0, 2, 5, 10, and 20 min from the beginning of rainfall - 15 s, 30 s, 1 min, 2 min, and 5 min, respectively. Rainfall lasted for a total of 30 min, which is when we could see no clay in the ponded water and *E. coli* measurements were near or below our detection limit. A total of five experiments were run.

The concentration of bacteria in each runoff sample was determined by a dilution and inoculation procedure. Sterilized 1.08% KCl solution was used as the diluent. Three aliquots from each runoff sample were diluted to three different end ratios. Sub-

sequently, each dilution was plated on *E. coli* media with 4-methylumbelliferyl- β -D-glucuronide (EC-MUG; Neogen corporation, Lansing, MI) and incubated for 20 h at 37 °C. CFUs were manually counted on each plate and converted to bacteria concentrations (CFU/ml). To verify that there were no unknown sources of *E. coli*, an additional experimental run was done without adding *E. coli* to the soil, and the results confirmed that there were no unaccounted for sources.

Despite the short duration of our experiments, we were concerned that the *E. coli* concentrations could be influenced by growth or die-off of the organisms. Growth or survival curves have been developed for *E. coli* in many different environments (Beversdorf et al., 2007; Gill and Delacy, 1991; Hwang et al., 2014; Muirhead and Littlejohn, 2009; Sagdic and Ozturk, 2013), but none exist for conditions similar to our experiment. We diluted our culture of *E. coli* in TSB with different doses of 1.08% KCl solution. Twenty microliters from each diluted sample was inoculated on an EC-MUG plate every 2 h over 10 h. The plates were then incubated for 20 h at 37 °C and enumerated to generate a population curve. There was a 40% increase in the population of *E. coli* over the 10 h period, but only a 5% increase in the population of *E. coli* during the first 2 h (data not shown). Thus, we assume growth or die-off of *E. coli* during our 30 min experiments is negligible, which is consistent with Oliver et al. (2007).

The concentration of clay in water samples was measured following Heilig et al. (2001) and Gao et al. (2003, 2004, 2005). A spectrometer (Spectronic 1001, Bausch and Lomb) was used to measure the runoff samples at 546.1 nm. Samples from one experimental run without clay in the soil were also analyzed by spectroscopy in order to determine if the presence of *E. coli*, TSB, or KCl in the water samples interfered with the measurements of clay concentration. These samples were all below our detection limit, so we assume these substances did not substantially affect our clay concentration measurements.

The rainfall intensity and the ponding water depth were measured before and after each experimental run. As in previous studies using this set-up (Gao et al., 2004, 2005; Wang et al., 2013), a layer of almost pure sand develops on the surface of the soil, i.e., referred to as a shield layer in the context of the Hairsine–Rose mode and as a mixing-layer or exchange-layer in the Gao model. The shield layer depth and the dry weight of shield layer were measured after each experimental run. The initial concentration of *E. coli* in the suspension which was then mixed with soil was also measured. Although we attempted to keep conditions identical between experimental runs, ponding depth, rainfall intensity, and initial *E. coli* concentrations (the initial concentration of the *E. coli*-TSB-KCl suspension before mixed with soil and the initial concentration of soil water) varied a little from run to run due to natural experimental variability; this was partially attributable to our need to set-up and run the experiments rapidly before the *E. coli* population could change. Also, sand size was larger than previous studies and may have increased the sensitivity of these experiments to other variables because the shield development was more rapid and the shield/exchange layer depth was thinner. The soil water content at saturation and the bulk density of the soil were measured in a separate experiment with the same artificial soil.

4. Model application

Despite the theoretical identity of the two models, their applications to the experimental data required some technical differences.

We applied Eq. 17 (Gao model) to the observed clay concentrations to obtain soil detachability, α ; this also allowed us a non-bacterial test to see if the Gao model would work as well as

Fig. 2. Measured and modeled clay and *E. coli*. The triangles are measured clay; the circles are measured *E. coli*; the bold solid red lines are clay modeled by Hairsine–Rose model; the bold dashed green lines are *E. coli* modeled by Hairsine–Rose model; and the thin dash-dot blue lines are *E. coli* modeled by Gao model. Each graph is for a separate experiment.

the Hairsine–Rose model for colloidal transfer between soil and runoff (see supplemental material Figure S1). All other parameters were either directly measured or inferred from measured parameters (Table 1). We found that the d_e calculated by $d_e = 10M_c^*/\rho_b$ worked better than trying to directly measure the exchange layer depth; this is consistent with previous applications of Hairsine–Rose model to these types of experiments where $9M_c^*$ was used to estimate M_d^* . Also, although we attempted to measure C_o directly, our measurements were systematically too high. We think this might be due to the heavy mineral particles displacing the lighter bacteria during shaking, which is consistent with the findings of Wang et al. (2013). So we let C_o to match the total CFUs lost divided by the pore-water volume of the exchange layer (results not shown); this is also consistent with the Hairsine–Rose model where N_e^* , the total CFUs eroded per unit area, was used. For the Hairsine–Rose model, it was not clear how to determine M_{cfu} , given possible aggregation of *E. coli* or aggregation of *E. coli*–clay, so we initially assume 1 cell = 1 CFU. Then, according to Neidhardt et al. (1990), $M_{cfu} = 9.5 \times 10^{-13} \text{ g/cell} \times 1 \text{ cell/CFU} = 9.5 \times 10^{-13} \text{ g/CFU}$.

5. Results

It can be seen from Fig. 2 that at the end of each run of the experiment, equilibrium state was reached, at which full shielding developed, and no more clay or *E. coli* is being released from soil into overland flow.

The clay data were well behaved (Fig. 2 and Table 3) and as previously shown in similar experiments, the Hairsine–Rose model captures the dynamics of the particle erosion process very well (e.g., Heilig et al. 2001; Gao et al. 2004).

As expected, the *E. coli* data were noisier than the clay data, in part because of the inherent variability in plate counts (Hedges, 2002; Oliver et al., 2005); we discuss this more in section 6. However, despite the relatively noisy data, the Hairsine–Rose particle erosion model and the Gao solute transport model work identically well for simulating *E. coli* concentrations in these experiments (Fig. 2 and Table 3); the two models are indistinguishable, as expected per our establishment that they were mathematically identical. Note also that the Hairsine–Rose model applied to the clay is identical to the application to the *E. coli*.

6. Discussion

6.1. Release mechanism of *E. coli*

The fact that the modeled curves of the release of *E. coli* are identical to the modeled curves of the release of clay shows that, as a particle, the mechanisms of *E. coli* transfer from soil into runoff is the same as mineral colloids, despite having neutral buoyancy in water.

The Hairsine–Rose model requires only one parameter; the main technical difficulty was how to interpret CFUs in the context of a particle mass. This was somewhat minimized by expressing our results normalized to the initial concentration. However, n is related to M_{cfu} by Eq. 7. As long as n is much larger than one the actual value of M_{cfu} does not really matter. Consider two extreme cases, one in which each single suspended cell in the sample grows into one colony and another where every aggregation as large as 1000 cells in the sample grows into one colony. As previously shown, if one cell = one CFU, then $M_{cfu} = 9.5 \times 10^{-13} \text{ g/cell} \times 1 \text{ cell/CFU} = 9.5 \times 10^{-13} \text{ g/CFU}$. If one

Table 3

Statistics of the model fit. Because the Hairsine–Rose and Gao models are ultimately identical when applied to *E. coli*, the statistics are the same for both.

	Clay						<i>E. coli</i>				
	run 1	run 2	run 3	run 4	run 5		run 1	run 2	run 3	run 4	run 5
RMSE	0.0135	0.00316	0.00122	0.00250	0.00235	RMSE	0.0104	0.00616	0.00442	0.00706	0.00305
Regression slope	0.966	0.900	0.910	0.966	1.05	Regression slope	1.08	1.08	0.753	0.478	1.00
R ²	0.790	0.873	0.973	0.927	0.904	R ²	0.875	0.530	0.627	0.420	0.826
Adjusted R ²	0.781	0.868	0.972	0.924	0.900	Adjusted R ²	0.870	0.511	0.612	0.397	0.818
p	6.08×10^{-10}	2.97×10^{-12}	2.09×10^{-20}	1.02×10^{-15}	3.16×10^{-14}	p	8.66×10^{-13}	1.66×10^{-5}	8.64×10^{-7}	2.59×10^{-4}	3.41×10^{-10}

colony equals to 1000 cells, then $M_{cfu} = 9.5 \times 10^{-13} \text{ g/cell} \times 1000 \text{ cell/CFU} = 9.5 \times 10^{-10} \text{ g/CFU}$. Then, by Eq. 7 and $I = 10n + 1$, we can get n and I (Table 1). We can see that in either of the extreme cases, $I \approx 10n$. In other words, the Hairsine–Rose model is insensitive to the estimated value of M_{cfu} .

In the Gao model, units of concentration (CFU ml^{-1}) are preserved, i.e., no assumption of M_{cfu} is required. Unfortunately, we were unable to make good *a priori* measurements of C_0 , which required us to estimate it *a posteriori*. We speculate that the ponded water after shaking the soil column was enriched with *E. coli* because the settling of the relatively dense mineral particles displaced the bacteria, similar to what Wang et al. (2013) observed with biochar using similar experiments.

6.2. Interaction between *E. coli* and clay

The identical model fits for *E. coli* and clay suggests that some *E. coli* could be attached to the clay particles. Neither model differentiates freely suspended *E. coli* from micro-aggregates of *E. coli* or micro-aggregates of *E. coli* and clay in overland flow. Mathematically and experimentally these forms are indistinguishable. In the Gao model, either one single *E. coli*, one micro-aggregate of *E. coli*, or one micro-aggregate of *E. coli* and clay were regarded as one CFU. While in Hairsine–Rose model, CFUs was converted to mass of *E. coli* via the mass of one cell and the assumed average number of cells per CFU. Oliver et al. (2005) found that the presence of clay resulted in “colloid facilitated transport” of *E. coli* but we did not see any solid evidence in our experiments to either support or contradict this.

6.3. Diffusion and adsorption

In our model derivation we assumed diffusion and adsorption of bacteria to sand were negligible. Olson et al. (2005) determined that diffusion coefficient, D_s in Gao et al. (2004), for a motile *E. coli* strain in a sand column (250–300 μm particles) was $3.0 \times 10^{-7} \text{ cm}^2/\text{s}$. If we include diffusion in the Gao model (see Gao et al. 2004), the fit to the data is almost the same as that for the model we derived here, which omits diffusion (Fig. 3). The inclusion of diffusion elevates the tail of the function a little, which does not seem to agree with our experimental data. This could be due to our relatively high rainfall intensity (twice that of Gao et al. 2004); note also that the diffusion coefficient of Olson et al. (2005) is less than one tenth of Gao et al. (2004)’s solute diffusion coefficient ($4.2 \times 10^{-6} \text{ cm}^2/\text{s}$). In fact, as our soil has clay mixed with sand so the actual diffusion coefficient would be less than that reported by Olson et al. (2005), which may partially explain why neglecting diffusion worked for the models as applied to our experimental data.

When non-zero K_p values are included in our model, the peak concentrations are lowered and the tail or recession concentrations are elevated, leading to disagreement between the model and the experimental data (Fig. 4); for all $K_p > 0$ the RMSE and R^2 were lower than for $K_p = 0$. However, we cannot exclude the possibility that some *E. coli* were adsorbed to sand particles. In fact, some

adsorption was likely to happen in the experimental environment with 0.145 M KCl. If this indeed occurred, it is possible that the reversibly adsorbed *E. coli* were too few to make a difference. Alternatively, if there was a substantial fraction of *E. coli* irreversibly adsorbed to sand particles, we may have inadvertently compensated for that in the way we estimated C_0 , i.e., it excludes the irreversibly adsorbed *E. coli* from the *E. coli* population from the beginning.

So, we understand that although *E. coli* are particulate, the Gao model with no diffusion simulates their release from soil into overland flow equally well as the Heilig et al. (2001)-simplified Hairsine–Rose particle model, despite having been originally derived as a solute model. As mentioned above, we compared the release of *E. coli* to that of clay, and showed that they show the same mechanism.

6.4. Soil detachability and exchange layer depth

One difference between our experiments and similar previously published experiments (Gao et al., 2004, 2005; Heilig et al., 2001) is that our curve-fitted or calibrated soil detachability parameter, a , varied for the same type of soil, especially for run 1 (Table 1). We believe this was due to the fact that we had to prepare the soil columns very gently and quickly so that the *E. coli* concentrations did not change due to growth or die-off. So, the soils were not packed with the same detachability from experiment to experiment; note, the experimental runs are numbered in the order they were run and it appears we were improving in our methodological consistency throughout this project, but we did not want to omit experiments without clear justification. It is also likely that our larger sand size relative to previous experiments may have made our experiments extra sensitive due to shallower shield/exchange layers and more rapid flushing of these layers.

In the Gao model, we calculated d_e (see model application), which was smaller than the d_e measured at the side of the column. Upon closer inspection, we found that the depth of deposited sand at the side of the column was systematically deeper than in the middle. We speculate that a sand grain deposited near the wall may be less likely to be re-ejected by a raindrop, thus there is a systematic accumulation along the column wall.

6.5. Consistency with previous published data

We compared our results to those of Ferguson et al. (2007)’s and, qualitatively, our data look similar to theirs for *E. coli* from bare soil. A figure of our data in a format similar to Fig. 2 (control part) from Ferguson et al. (2007) is included in supplemental material (Figure S2).

6.6. Noisy *E. coli* data

It is obvious that the relative concentration of *E. coli* showed more scatter than that of clay (Fig. 2, especially 2(b), (c) and (d)). This is, in part, because the method used to measure *E. coli* concentrations was less precise than that used to measure clay concentrations. For clay, each overland flow sample was usually di-

Fig. 3. The full Gao model with $D_s = 0$ (solid lines) and $D_s = 3.0 \times 10^{-7} \text{ cm}^2/\text{s}$ (dashed lines) comparing with data (circles). The experiments are in the same order as in Fig. 2..

Fig. 4. The full Gao model with $K_p = 0, 0.1, 0.2, 0.4, 0.8 \text{ (cm}^2/\text{s)}$. The bold black lines are when $K_p = 0$. The thin green, blue, magenta and red lines are when $K_p = 0.1, 0.2, 0.4, 0.8 \text{ cm}^2/\text{s}$, respectively. The arrows indicate the trends of change when K_p varies from 0 to $0.8 \text{ cm}^2/\text{s}$. The experiments are in the same order as in Fig. 2.

luted by a 1:5 or 1:10 ratio; whereas, for *E. coli*, the overland flow samples were usually diluted by 1:20 and then a 20 μ l sub-sample from the 1 ml diluted sample was inoculated on a plate. This means that the error of concentration of clay measured by the spectrometer was amplified by 5 or 10 times in the final results, while the error of the concentration of *E. coli* measured by counting the colony-forming units in the plates was amplified by $20 \times 50 = 1000$ times. And the dilution ratio had to be adjusted frequently from experiment to experiment as well as sample to sample in order to keep the CFU counts within method-approved ranges (Oliver et al., 2007). Indeed, the plate counting is especially imprecise when trying to relate it to a mass of *E. coli* (Hedges, 2002; Oliver et al., 2005). And it was the noisiness of *E. coli* data, especially for runs 3 and 4 (Figs. 2(c) and (d), respectively) regression slopes less than one and low R^2 values (Table 3).

7. Conclusions

We derived two models for colloidal transfer from soil into overland flow via raindrop impact, one based on soil erosion processes (Hairsine–Rose) and the other based on solute transfer processes (Gao). When applied to the assumptions that colloids have negligible diffusion and settling rates, the two models are identical. They both described *E. coli* concentration data from controlled rainfall-runoff experiments. Although there are unique challenges in defining model parameters in each model, neither model was overly sensitive to the necessary assumption that had to be made. The major challenge for the Gao derived model was determining the initial *E. coli* concentration in the near-surface pore water, which here we back-calculated from the observed cumulative release of *E. coli*. For the Hairsine–Rose derived model, we had to make assumptions about how many cells, on average, initiated a colony forming unit (CFU) and showed that the model is insensitive to this assumption over many orders of magnitude as long as the relative mass-fraction of the cells is much lower than any of the soil-particle mass-classes. Our findings suggested that management practices that reduce raindrop impact are most likely to reduce microbial release from soil into storm runoff, e.g., maintaining a vegetative or residue cover.

Future studies on microbial release from soil into overland flow should address some of the following unresolved issues: (1) do mineral-bacterial aggregates enhance or diminish soil-runoff transfer, (2) do non-destructive methods like magnetic resonance imaging provide better quality data and better characterization of the underpinning processes, (3) do other microorganisms, especially pathogens, behave differently than those of the *E. coli* (ATCC 25922) used here, (4) consider incorporating the important biological processes of population growth and die-off, (5) conduct similar experiment at a large scale and see if the assumptions are still valid and if the models can still capture the data well, and (6) use real soil which has multiple particle size classes to conduct the experiment.

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Appendix A. Analytical solution to the simplified no diffusion Gao solute model

Inserting Eqs. 14 into Eq. 13 and solving Eq. 13,

$$c \cdot C_e = \exp\left(-\frac{ap}{\rho_b d_e} t\right) \quad (A1)$$

$$\therefore C_e = C_o \text{ at } t = 0, \quad (A2)$$

$$\therefore C_e = C_o \cdot \exp\left(-\frac{ap}{\rho_b d_e} t\right) \quad (A2)$$

Inserting Eq. 14 into Eq. 12 and solving Eq. 12,

$$C_w = \left(\int \frac{ap\theta}{\rho_b d_w} C_e \cdot \exp\left(\int \frac{p}{d_w} dt\right) dt + c_2 \right) \cdot \exp\left(-\int \frac{p}{d_w} dt\right) \quad (A3)$$

Plugging in Eq. A2,

$$C_w = \left(\frac{ap\theta}{\rho_b d_w \left(\frac{p}{d_w} - \frac{ap}{\rho_b d_e}\right)} \cdot C_o \cdot \exp\left(\left(\frac{p}{d_w} - \frac{ap}{\rho_b d_e}\right)t\right) + c_2 \right) \cdot \exp\left(-\frac{p}{d_w} t\right) \quad (A4)$$

$$\therefore C_w = 0 \text{ at } t = 0 \text{ (Eq. 15)}$$

$$\therefore C_w = \frac{ap\theta}{\rho_b d_w \left(\frac{p}{d_w} - \frac{ap}{\rho_b d_e}\right)} \cdot C_o \cdot \left(\exp\left(\left(\frac{p}{d_w} - \frac{ap}{\rho_b d_e}\right)t\right) - 1 \right) \cdot \exp\left(-\frac{p}{d_w} t\right) \quad (A5)$$

Supplementary materials

Supplementary material associated with this article can be found, in the online version, at doi:10.1016/j.advwatres.2016.10.016.

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