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Modeling Transport of Microbes in Ten Undisturbed Soils under Effluent Irrigation

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The HYDRUS-1D mobile–immobile water model (MIM) was used to evaluate the transport of fecal coliforms, *Salmonella* bacteriophage, and Br in 10 soils. At a flux of 5 mm h^{-1}, a pulse of dairy shed effluent was applied to 30 large undisturbed lysimeters, followed by water irrigation. Soil types included clayey gley soil, clay loam, silt loam, silt loam over gravels, fine sandy loam, dune sand soil, pumice soil, and allophanic soil. Except for dune sand, modeling results showed lower mobile water contents and dispersivities for microbes than for Br, indicating the exclusion of microbes from smaller pores. The MIM-derived removal rates were in the order: volcanic soils > greywacke-derived silt loams > granular young sandy soils, and were the most variable in clayey gley loam and silt loam over gravels. Microbial reduction was 100% in allophanic soil, 16 to 18 log m^{-1} in pumice soil (where the unit log is the $\log_{10}$ reduction in maximum concentration compared with the original concentration), and was lowest in clayey gley soil (0.1–2 log m^{-1}). For most of the other soils, the reduction was 2 to 3 log m^{-1}, except for 9 to 10 log m^{-1} for fecal coliforms in a fine sandy loam. The detachment rate was only 1% of the attachment rate, indicating irreversible attachment of microbes. Soil structure (macroporosity) appeared to play the most important role in the transport of microbes and Br, while soil lithology had the greatest influence on attenuation and mass exchange. The general pattern of predicted mobile water content agrees with the measured macroporosity, which is positively related to leaching vulnerability but negatively related to dispersivity.

ABBREVIATIONS: BTC, breakthrough curve; cfu, colony-forming unit, a unit indicating the number of bacteria present in a water sample; DSE, dairy-shed effluent; PV, pore volume.

Irrigation of dairy-shed effluent (DSE) onto land to fertilize pasture and safely dispose of effluent is an integral part of New Zealand's farming practice; however, the use of inappropriate soils and practices could result in an increased risk of contaminating waterways and groundwater with DSE. Animal wastes contain many pathogenic microorganisms such as *Cryptosporidium*, *Giardia*, *Campylobacter*, toxigenic *Escherichia coli*, *Salmonella*, and some viruses (e.g., rotavirus) (Pell, 1997), and some of these are zoonotic (i.e., can cause human diseases). New Zealand has the highest reported incidence of campylobacteriosis of any developed country (Baker et al., 2006; Gilpin et al., 2006), and there is concern about the possible link between these outbreaks and contamination of drinking water sources due to pathogen migration through soil leaching and overland flow. Apart from harmfully affecting domestic water supplies, contamination of waterways by fecal microorganisms may also adversely affect shellfish harvesting and can pose a health risk to recreational water users. In addition, the health and productivity of livestock may be compromised when they consume water contaminated with fecal material (Pell, 1997).

Soils act as natural filters that can attenuate microbial contaminants, but they vary widely in their ability to remove them. The ability of New Zealand soils to remove microorganisms has been evaluated previously in a few studies. Wells (1973) rated the properties of New Zealand soils in relation to effluent disposal and concluded that young (2000–7000 yr of age), fine-grained tephric soils had the best characteristics for effluent disposal, while the gleyed soils were unsuitable because of their anaerobic conditions. There were no experimental data to support this conclusion, however. Childs et al. (1977) performed some lysimeter leaching experiments examining the ability of some New Zealand soils (peat, clay, silt loam, sandy loam, sand, and stony soil) to remove chemicals and fecal coliforms from sewage effluent. They found that concentrations of fecal coliforms derived from effluent were >100 colony-forming units (cfu) L^{-1} in leachate collected at 50-cm depth for all soils under both spray and flood irrigation, except for dune sand and a silt loam derived from volcanic ash under spray irrigation. In a field study of a border strip (flood) effluent irrigation scheme in Canterbury, New Zealand, Sinton et al. (2005) observed rapid leaching of fecal coliforms and F-RNA phages through alluvial gravel soils (15.7–39.2 m h^{-1}), and the estimated reduction in the bacteria and phages was only $\log_{10}(c_{max}/c_o)$ = 1.42 and 2.63, respectively (the $\log_{10}$ reduction in maximum concentration compared with

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original concentration), through a 16.8-m vadose zone. In recent years, lysimeter studies were performed to examine the leaching of fecal coliforms and bacteriophages through a range of key New Zealand soils commonly used for effluent disposal (Aislabie et al., 2001; McLeod et al., 2001, 2003, 2004). The results of these studies suggest that the vertical movement of bacteria and viruses varies significantly with soil type.

Although experimental data were collected in some of these studies, no modeling work was performed. Thus parameters (such as removal rates) that describe the fate and transport of microbes in New Zealand soils remains unknown, limiting the applicability of the data in the field. Although some overseas studies have derived parameters of microbial transport in undisturbed soils (Shelton et al., 2003; Guber et al., 2005; Frazier et al., 2002), the physicochemical properties of some New Zealand soils (e.g., allophanic soils) are quite unique. In addition, it is difficult to compare the results from different studies because the removal of microbes in soils changes according to the experimental conditions, especially the rate of irrigation (Guber et al., 2005).

Contaminant transport models are useful tools for the quantitative assessment of microbial transport in soils and the elucidation of the importance of factors that control microbial concentrations in receiving waters. By calibrating transport models with observed data, we can characterize microbial attenuation and transport in a specific soil system using descriptive parameters (e.g., removal, attachment, and detachment rates). These calibrated parameter values together with transport models can then be used by others (e.g., government agencies, regional authorities, researchers, and consultants) to manage and predict similar systems for various purposes (e.g., resource management, land-use planning, design of monitoring programs, and risk analysis).

New Zealand has a diverse landscape with a wide range of contrasting soil types occurring across short distances (McLeod et al., 2001). As dairy farming is widespread across the nation, it is important to evaluate the leaching of microorganisms in the different soils in New Zealand so that best farming practices can be implemented. The objectives of this study were (i) to better understand microbial transport in major New Zealand soils under DSE irrigation, and (ii) to establish values of important parameters for describing microbial transport and attenuation in these soils by applying an appropriate contaminant transport model to the data obtained from lysimeter experiments generated in this and previous studies (Aislabie et al., 2001; McLeod et al., 2001, 2003, 2004). These results will be of critical importance to regulatory agencies that need scientific information about microbial removal in different soils to enable them to effectively manage development and evaluate the risk of groundwater contamination.

Materials and Methods

Selected Soils and Microbial Tracers

Ten major New Zealand soils were evaluated in this study, including two soils (Lismore silt loam over gravels and Templeton deep silt loam) investigated in this study and eight other soils presented in Aislabie et al. (2001) and McLeod et al. (2001, 2003, 2004). These soils are widely distributed in areas of intense dairy farming and are commonly irrigated with DSE. The selected soils cover a wide range of physical, chemical, and mineralogical properties. The major features of the selected soils are summarized in Table 1, and some physical and chemical properties are listed in Table 2.

In this study and those by Aislabie et al. (2001) and McLeod et al. (2001, 2003, 2004), fecal coliforms and a host-specific bacteriophage *Salmonella typhimurium* 28B (Lilleengen, 1948) were used as the microbial indicators, and Br was used as a tracer for water movement. Fecal coliforms are always present in animal and human wastes and are the most widely used bacterial indicator. Fecal coliforms have typical dimensions of 0.3 to 1 μm in diameter and 0.6 to 6 μm in length. The transport of fecal coliforms in soils has been reported in many studies (e.g., Rahe et al., 1978; McCoy and Hagedorn, 1980; Jamieson et al., 2002; Unc and Goss, 2003; Shelton et al., 2003). Phage 28B does not occur in environmental samples or in feces, and it has been used as a viral tracer to determine the source of fecal contamination (Stenström, 1996). Phage 28B is 60 nm in diameter, and is persistent at elevated pH values (Carlander and Westrell, 1999). Carlander et al. (2000) used Phage 28B to evaluate the impact of viruses on groundwater quality from wastewater irrigation of a coppice. In their field lysimeter study, rapid transport of Phage 28B was observed in clay soils, with breakthrough at a depth of 1.2 m after 2 to 24 h, probably due to the presence of macropores and bypass flow. They observed that phage transport through sandy soils varied considerably, with a generally much slower rate and higher retention compared with the clay soil.

Lysimeters and Leaching Experiments

The methods used for obtaining intact cores of Lismore and Templeton soils and leaching experiments were the same as those used for the other soils by Aislabie et al. (2001) and McLeod et al. (2001, 2003, 2004). The description below applies for all 10 soils used for modeling in this study.

Undisturbed soil cores were hand carved in situ from the ground surface, and the lysimeter casings were progressively pressed down over the cores. The lysimeters were made of high-density polyethylene (HDPE) pipe, with an internal diameter of 46 cm and a length of 47 to 70 cm. A 1-cm internal annulus was filled with petroleum jelly to prevent water preferentially flowing at the soil-casing interface. The bottom of the lysimeter was welded to a 1-cm-thick HDPE base plate, with a sampling port installed in the center to allow the collection of leachate. The pore volume (PV) for each set of lysimeter cores was estimated from previous analyses of the total porosity of these soils at the same site. One PV is the amount of space in the soil core occupied by soil pores or cracks. This ranged from 45 to 67% of the total soil volume (Table 2).

In the laboratory, the lysimeters were initially irrigated for 5 d with tap water to field saturation with leachate emanating from the sampling port, then were allowed to drain for 7 d. Seven days is similar to the return period commonly used at effluent irrigation sites. Following this, each lysimeter was irrigated, at a constant rate of 5 mm h^{-1}, with a 25-mm depth of DSE, containing native fecal coliforms (typically 10^5–10^9 cfu mL^{-1}) and spiked with a tracer solution containing *Salmonella* bacteriophage (10^9 plaque-forming units [pfu] mL^{-1}) and Br (2 g L^{-1}). The lysimeters were then irrigated continuously with tap water at a rate of 5 mm h^{-1} using a drip-type rainfall simulator with drippers spaced on a 20-mm triangular grid, approximately 170

TABLE 1. Major features of the selected soils.

Soil	Structure	Soil classification		Parent material	Clay mineralogy	Drainage capacity
		New Zealand†	U.S.			
Netherton clayey gley loam	Topsoil: medium or coarse polyhedral structure. Subsoil: coarse prismatic structure with low porosity within the prisms. Large cracks lined with organic matter and clay cutans, large worm holes and root channels.	Typic Orthic Gley	Typic Endoaquept	estuarine alluvium	mixed	poor
Hamilton clay loam	Topsoil: strongly pedal with fine and medium polyhedral peds. Subsoil: strongly pedal and clayey. Medium and coarse polyhedral peds; abundant continuous clay coatings on ped faces; large nearly vertical earthworm channels throughout, up to 5-mm diameter	Mottled Orthic Granular	Typic Haplohumult	strongly weathered rhyolitic and andesite tephra	halloysitic	imperfect
Waikiwi silt loam	Topsoil: well structured with fine polyhedral peds. Upper subsoil: moderately well developed fine and medium blocky peds. Below 50 cm: firm apedal horizon.	Typic Firm Brown	Typic Dystrudept	tuffaceous greywacke loess	mixed	good
Waikoikoi silt loam	Topsoil: fine polyhedral structure over a worm-mixed transitional horizon. Upper subsoil: strongly mottled and well structured with medium blocky peds. About 50 cm: dense, compact fragipan with very low permeability.	Fragic Perch-gley Pallic	Aeric Fragiaquept	loess derived from schist and greywacke rocks	mixed	poor
Lismore shallow silt loam over gravels	Top 36 cm: silt loam, strongly to weakly developed fine to coarse subangular blocky structure with few indistinct clay coats. 36–70 cm: gravelly silt loam and loamy sand, >50% stones with silt coating. Many fine roots throughout, very weak soil strength.	Typic Orthic Brown	Typic Dystrustept	greywacke loess on greywacke gravels	mixed	good
Templeton deep silt loam	Topsoil: very firm with a weakly developed blocky structure over a moderately firm transitional horizon with a weakly developed subangular blocky structure. Upper subsoil: moderately firm with weakly developed coarse blocky structure. Below about 50 cm: massive soil structure.	Typic Immature Pallic	Typic Dystrustept	greywacke alluvium	mixed	imperfect
Manawatu fine sandy loam	Topsoil: moderately pedal with extremely fine and fine polyhedral peds. Subsoil: weakly or moderately pedal silt loam and fine sandy loam. Medium polyhedral peds tending to massive at depth.	Weathered Fluvial Recent	Fluventic Eutrudept	recent soil, quartzo-feldspathic alluvium	mixed	good
Waitarere recent sandy soil	Minor pedological development with a single-grain structure throughout, except for some weak aggregation in the topsoil.	Typic Sandy Recent	Typic Udipsamment	dune sand	mixed	good
Atiamuri pumice soil	Massive but porous, welded pumice flow tephra at about 40-cm depth, which prohibits deep rooting by plants. Coarse sand with some gravels, fine to single-grain soil structure.	Typic Orthic Pumice	Typic Udivitrand	sandy rhyolitic tephra	rhyolitic and with small fraction of allophane	good
Waihou allophanic soil	Weakly developed, porous, subangular blocky and crumb structure. Very fine structure, single grain. Below 40 cm: welded pumice.	Typic Orthic Allophanic	Typic Hapludand	rhyolitic tephra	allophanic	good

† New Zealand soil classification (Hewitt, 1998).

TABLE 2. Major chemical and physical properties of the selected soils.

Soil	Layers in HYDRUS	Depth†	pH	C		CEC‡	Clay		ρ_b§	Total θ¶	Macropore θ#	*d*††	Dp/d‡‡	
				Topsoil	Subsoil		Topsoil	Subsoil					Fecal coliforms	Phages
	no.	cm		—— % ——		$cmol_c\ kg^{-1}$	—— % ——		$g\ cm^{-3}$			μm	—— % ——	
Netherton clayey soil	5	70	5.1–6.1	5.4	0.8	27–32	52–55	60–69	0.98	0.61	0.03	<2	73.00	3.00
Hamilton clay loam	5	70	4.9–5.3	3.0	0.8–1.1	9.9–17.2	29–30	35–79	1.16	0.51	0.05	9	16.11	0.66
Waikiwi silt loam	4	47	5.7–6.1	4.1	1.6–2.7	9.1–15.4	24	12–18	1.22	0.53	0.07	15	9.48	0.39
Waikoikoi silt loam	3	50	5.9–6.0	3.2	1.2–1.7	9.0–14.2	23	16	1.42	0.45	0.09	14	10.40	0.43
Lismore shallow silt loam over gravels	5	36	5.6	2.2		11.7	21–22		1.73	0.47	0.21	19§§	7.68§§	0.32§§
		70	5.9		0.8	8.3		12–24	2.20	0.56				
Templeton silt loam	4	40	5.6–5.9	3.1	0.3	15.2–6.7	24	15–23	1.37	0.47	0.12	23	6.35	0.26
Manawatu fine sandy loam	5	70	5.5–6.4	2.0	0.5–0.7	8.7–11.3	10	20	1.31	0.49	0.12	75	1.95	0.08
Waitarere sandy recent soil	4	70	5.0–5.7	6.0	0.6	3–19	6	2	1.18	0.56	0.19	212	0.69	0.03
Atiamuri pumice soil	4	70	5.7–6.2	8.1	0.4	5.5–6.2	5	5	0.76	0.50	0.23	122	1.19	0.05
Waihou silty allophane soil	4	70	5.1–6.5	6.8	1.7	12–28	26	34	0.75	0.67	0.16	22	6.67	0.27

† Depth of soil lysimeter taken from ground surface.
‡ Cation exchange capacity.
§ Length-averaged dry bulk density.
¶ Length-averaged porosity.
# Pores ≥59 μm in diameter determined at 5-kPa water potential.
†† Length-averaged mean particle size.
‡‡ D_p = 60 nm for phage and 1.46 μm for fecal coliforms.
§§ 36-cm soil depth only; below 36 cm, gravel size >2 mm.

mm above the soil surface. Typically the chemical properties of the tap water were as follows: pH = 7.7, electrical conductivity = 21.2 mS m^{-1}, and ionic strength = 0.003 mol L^{-1}. Experiments were performed on triplicate intact cores for each soil type.

To obtain the background levels of Br and microbial tracers in the leachate, samples were first taken at the end of the wetting period. During the leaching experiments, soil leachate was collected into sterile bottles and subsampled. Subsamples for microbial assays were stored at 4°C and analyzed within 2 h of collection. Bromide tracer samples were analyzed within 1 wk of collection.

Batch tests were also performed in the current study to determine the inactivation rates of the microbial indicators in DSE under similar experimental conditions to those used in the lysimeter studies (background solution, pH, concentrations of microbial indicators, and Br). The reactors were incubated at 15°C in the dark for 14 to 17 d. Phages were sampled daily for 14 d, while fecal coliforms were sampled twice a day for the first 5 d and then once a day between Days 5 and 17.

Sample Analysis

The bacteriophage propagation and assay method used was detailed in McLeod et al. (2001). Essentially, Phage 28B was grown overnight on its host strain *S. typhimurium* Type 5 in tryptic soy broth at 37°C. The phages were isolated by chloroform lysis of the bacterial host, then passed through a 0.45-μm mixed cellulose ester-based membrane filter to remove cell debris. To obtain a clean virus preparation free of organic material, the filtrate was centrifuged at 25,000 × *g* (Sorval T21, Thermo Fischer Scientific, Waltham, MA) for 2 h at 4°C, the supernatant was poured off, and the phages were resuspended in 1 to 2 mL of phage storage buffer (Sambrook et al., 1989) and stored at 4°C until required. Phage stocks were enumerated using a soft agar overlay method. Leachate samples were mixed with the host-strain culture and poured onto nutrient agar plates. After incubation for 18 to 24 h at 37°C, well-formed, clear plaques were counted and reported as plaque-forming units per milliliter. Each reported phage concentration is the average of three replicates.

Fecal coliforms were determined in soil leachates using a membrane filtration technique (American Public Health Association, 1998). Samples were diluted in phosphate-buffered water (pH 7.0), then filtered according to standard procedures. The filters were placed on mFC agar (Difco, BD Diagnostics, Auckland, NZ) and blue colonies were counted after incubation for 24 h at 44.5°C. Bromide concentrations in the leachate samples were measured using an ion-selective electrode (Metrohm 6.0502.100, Herisau, Switzerland).

Modeling and Data Analysis

The Richards equation (Richards, 1931) was used to describe variably saturated one-dimensional vertical water flow in the soil lysimeters:

$$\frac{\partial \theta(h)}{\partial t} = \frac{\partial}{\partial x}\left[K(h)\left(\frac{\partial h}{\partial x}+1\right)\right] \quad [1]$$

where h is the water pressure head [L], θ is the volumetric water content [$L^3\ L^{-3}$], t is time [T], x is the spatial coordinate [L], and $K(h)$ is the unsaturated hydraulic conductivity function [$L\ T^{-1}$]. The variable $K(h)$ is described by the van Genuchten–Mualem hydraulic model (van Genuchten, 1980). The parameters used in the van Genuchten–Mualem hydraulic model (Table 3) were obtained by analyzing measured water retention curves (θ vs. h) using the RETC program (van Genuchten et al., 1991) or by using soil texture data and the Rosetta program (Schaap et al., 2001) if water retention data were absent.

Concentration breakthrough curves (BTCs) for Br are generally asymmetrical with significant tailing. This indicates the presence of physical nonequilibrium processes in the experimental systems as a result of media heterogeneity. Thus a two-region mobile–immobile model (van Genuchten and Wagenet, 1989; Toride et al., 1995) was used to simulate transport of Br:

Table 3. Parameters† used in the van Genuchten–Mualem hydraulic model.

Soil	Depth	θ_r	θ_s	α	*n*	ρ_b
	cm			cm^{-1}		g cm^{-3}
Netherton clayey gley loam	0–10	0.114	0.650	0.026	1.287	0.870
	10–20	0.112	0.612	0.023	1.287	0.985
	20–38	0.116	0.616	0.027	1.247	1.020
	38–49	0.118	0.624	0.028	1.241	1.005
	49–70	0.115	0.628	0.026	1.263	0.980
Hamilton clay loam	0–9	0.090	0.532	0.008	1.528	1.110
	9–19	0.085	0.481	0.008	1.551	1.210
	19–29	0.090	0.471	0.010	1.479	1.320
	29–46	0.099	0.500	0.014	1.371	1.330
	46–70	0.120	0.611	0.029	1.220	1.030
Waikiwi silt loam	0–20	0.000	0.513	0.002	1.267	1.130
	20–26	0.000	0.462	0.001	1.244	1.210
	26–39	0.000	0.461	0.005	1.109	1.290
	39–47	0.000	0.413	0.002	1.112	1.460
Waikoikoi silt loam	0–20	0.000	0.430	0.003	1.256	1.270
	20–41	0.067	0.418	0.006	1.627	1.500
	41–50	0.000	0.364	0.003	1.148	1.570
Lismore shallow silt loam over gravels	0–17	0.078	0.476	0.005	1.664	1.200
	17–28	0.000	0.366	0.009	1.190	1.353
	28–36	0.071	0.415	0.006	1.628	1.420
	36–47	0.041	0.243	0.045	1.133	2.200
	47–70	0.046	0.233	0.036	1.624	2.200
Templeton deep silt loam	0–22	0.000	0.371	0.004	1.204	1.348
	22–29	0.076	0.445	0.006	1.636	1.330
	29–34	0.000	0.332	0.008	1.150	1.510
	34–40	0.061	0.406	0.005	1.688	1.370
Manawatu fine sandy loam	0–10	0.000	0.457	0.004	1.252	1.170
	10–23	0.067	0.453	0.005	1.713	1.245
	23–33	0.000	0.410	0.010	1.191	1.305
	33–60	0.073	0.451	0.005	1.656	1.345
	60–70	0.031	0.404	0.019	1.199	1.420
Waitarere recent sandy soil	0–10	0.049	0.596	0.055	1.499	0.825
	10–12	0.050	0.451	0.041	2.556	0.825
	12–30	0.052	0.468	0.040	2.618	1.290
	30–70	0.051	0.502	0.046	2.492	1.240
Atiamuri pumice soil	0–10	0.235	0.547	1.338	2.417	0.555
	10–23	0.155	0.511	2.297	2.138	0.745
	23–40	0.101	0.533	1.865	1.827	0.835
	40–70	0.149	0.568	1.756	1.946	0.800
Waihou allophanic	0–15	0.096	0.614	0.008	1.523	0.775
	15–31	0.100	0.642	0.010	1.503	0.710
	31–64	0.109	0.670	0.015	1.401	0.710
	64–70	0.076	0.512	0.008	1.563	0.995

† θ_r and θ_s, residual and saturated water contents; α and *n*, air-entry parameters in van Genuchten model; ρ_b, dry bulk density.

$$\frac{\partial \theta_m c_m}{\partial t} = \frac{\partial}{\partial x}\left(\theta_m D_m \frac{\partial c_m}{\partial x}\right) - \frac{\partial q c_m}{\partial m} - \alpha\left(c_m - c_{im}\right) \quad [2]$$

$$\theta_{im} \frac{\partial c_{im}}{\partial t} = \alpha\left(c_m - c_{im}\right) \quad [3]$$

where the subscripts m and im refer to the mobile and immobile water regions, respectively; c is the concentration in the liquid phase [M L^{-3}]; $\theta = \theta_m + \theta_{im}$ is the volumetric moisture content [L^3 L^{-3}]; D is the dispersion coefficient [L^2 T^{-1}], which is the product of dispersivity ξ [L] and the spatial coordinate x (i.e., $D = \xi x$); q is the water flux [L T^{-1}]; and α is the first-order mass-exchange coefficient [T^{-1}] governing the rate of solute exchange between the mobile and immobile water regions.

The two-region mobile–immobile model (MIM) assumes that water flow and contaminant transport is limited to the mobile water region and that water in the immobile water region is stagnant, with a first-order diffusive exchange process between the two regions.

In our recent unpublished batch study, Br was adsorbed in soils that contain volcanic materials (Waihou allophanic soil, Atiamuri pumice soil, and Hamilton clay loam). Therefore, the distribution coefficient for linear adsorption, K_d [L^3 L^{-1}], was also considered for these soils. Assuming all adsorption sites equilibrate with the mobile water phase, Eq. [2] becomes

$$\frac{\partial\left(\theta_m + \rho_b K_d\right) c_m}{\partial t} = \frac{\partial}{\partial x}\left(\theta_m D_m \frac{\partial c_m}{\partial x}\right) - \frac{\partial q c_m}{\partial x} - \alpha\left(c_m - c_{im}\right) \quad [4]$$

in which ρ_b is the bulk density of the soil material [M L^{-3}]. The K_d values determined from the unpublished batch study were used in the model.

We assumed that microbes were excluded from the immobile water region (thus no mass exchange between two regions, $\alpha = 0$), and thus their attachment and inactivation occurred only in the mobile water region. Under this assumption, the two-region MIM for describing transport of microbes at a constant flow is expressed as

$$\frac{\partial \theta_m c_m}{\partial t} + \rho_b \frac{\partial S}{\partial t} = \frac{\partial}{\partial x}\left(\theta_m D_m \frac{\partial c_m}{\partial x}\right) - \frac{\partial q c_m}{\partial x} - \theta_m \mu c_m \quad [5]$$

$$\rho_b \frac{\partial S}{\partial t} = \theta_m k_{att} c_m - \rho_b k_{det} S \quad [6]$$

where S is the concentration attached to the soil media [cfu or pfu M^{-1}]; c_m is the concentration in the mobile phase [cfu L^{-3} for bacteria and pfu L^{-3} for phages]; k_{att} and k_{det} are the first-order rate coefficients for attachment and detachment [T^{-1}], respectively; and μ is the measured first-order inactivation rate coefficient in the liquid phase [T^{-1}]. We have assumed that the attachment rate, k_{att}, also includes the effects of the air–water interface and straining. The total removal rate, k_{tot} [T^{-1}], is then $k_{tot} = k_{att} + \mu$.

The inactivation rate for the microbial indicators in the liquid phase (μ) was determined by fitting the experimental data of the inactivation batch tests with an exponential function, $c = c_o \exp(-\mu t)$, where c_o is the influent concentration, using the Solver function of the Microsoft Excel spreadsheet.

Since an equal flux is applicable for both Br and microbial tracers, $q = (\theta_m v_m)_{microbe} = (\theta_m v_m)_{Br}$, where v is the pore-water velocity, velocity enhancement of a microbial tracer can be measured from the ratio

$$\frac{v_{m\text{-}microbe}}{v_{m\text{-}Br}} = \frac{\theta_{m\text{-}Br}}{\theta_{m\text{-}microbe}} \quad [7]$$

Equations [1–6] were solved numerically using HYDRUS-1D (Šimůnek et al., 2005). The HYDRUS model consisted of four to five layers for a lysimeter, each with variable inputs of soil hydraulic properties (using the van Genuchten–Mualem model) and bulk density. Values of ξ, θ_m (via θ_{im}), and α were optimized for the Br data, while ξ, θ_m (again via θ_{im}), k_{att}, and k_{det} were optimized for the microbial data. The measured inactivation rates in the liquid phase for the bacteria and phages, μ, were fixed in the model. Single values of ξ, θ_m, α, k_{att}, and k_{det} were assigned for all layers. The distinctive transport patterns of Br and the microbial tracers (Fig. 1), as a result of size exclusion in microbial transport, meant that ξ and θ_m for the microbes had to be independently estimated from those for Br. HYDRUS-1D was modified for this study to simultaneously assign the same values of optimized parameters to all layers and to keep the mobile water content the same in all layers. The reason for using of the same parameter values for multiple layers was to reduce the number of parameters to be optimized. This treatment is reasonable, as all BTC data were obtained from one sampling point at the end of the lysimeters.

The available data included 29 Br BTCs, 30 phage BTCs, and 18 bacteria BTCs (Fig. 1, Table 4). Some BTCs were incomplete, however, and some BTCs had largely zero values. The quality of the BTCs determined by their completeness is given in Table 4. Therefore, the number of BTCs suitable for modeling was 29 for Br, 27 for the phages, and 17 for the bacteria.

Results and Discussion

Transport of Bromide Solute Tracer

Figure 1 shows that the peak concentrations of all of the Br BTCs arrived earlier than 1 PV except for Waihou allophanic soil. These earlier Br breakthroughs, together with significant tailings in the Br BTCs, suggest the presence of preferential flow paths or dual flow regions due to zones of contrasting permeabilities within the undisturbed cores. When Br passes through the soil medium, a portion of the solute preferentially goes through high-permeability zones (or the macropore network), providing that soil moisture is sufficiently high to fill these pores, and the other portion of the solute diffuses into the low-permeability zones (or matrix). After the solute has moved through the high-permeability zones, the solute in the low-permeability zones slowly diffuses back to the high-permeability zones, driven by the concentration gradient. This water exchange between zones of contrasting permeability causes the observed "tailing" in Br BTCs. Therefore, the front of a BTC reflects solute transport

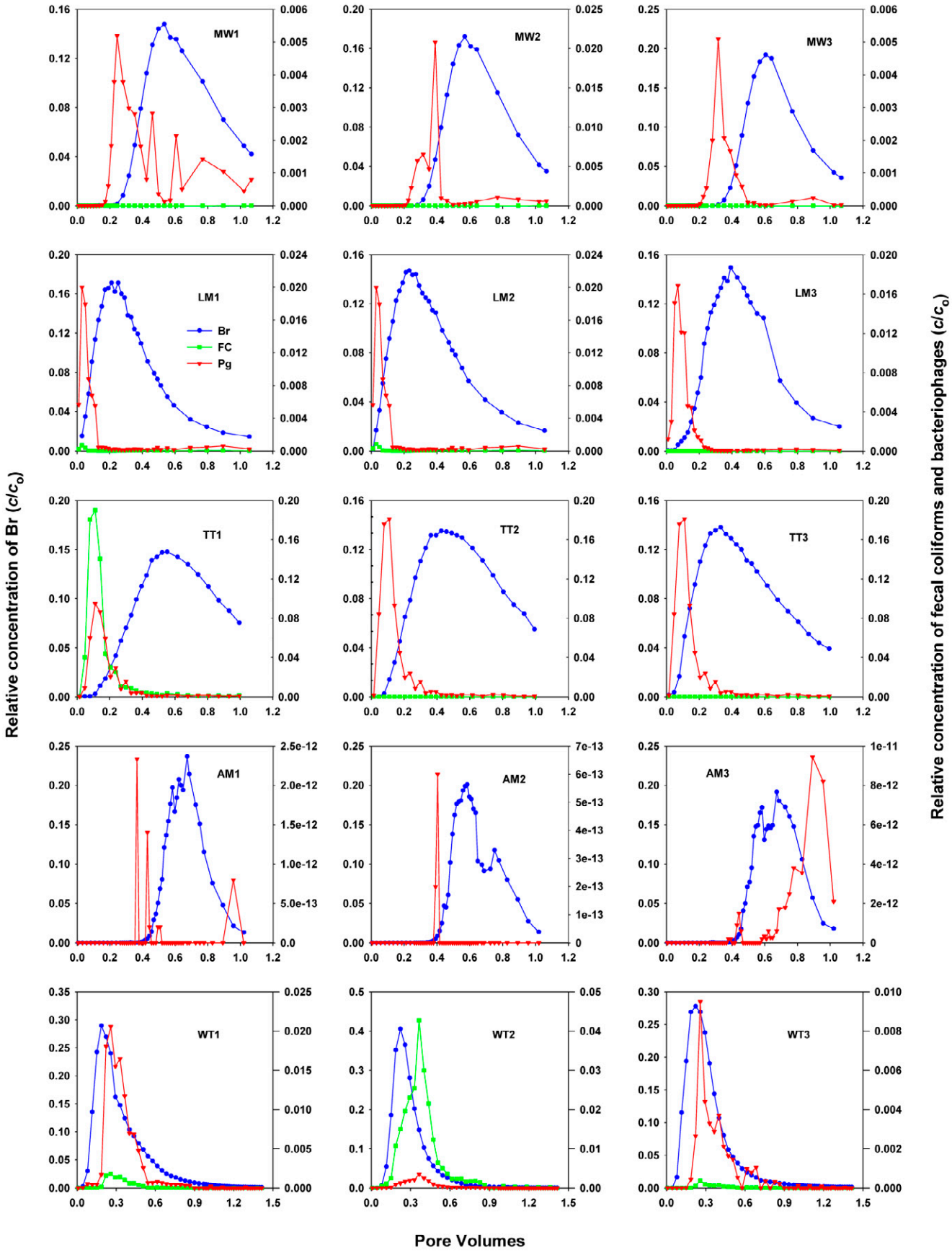

FIG. 1. Concentrations of fecal coliforms, *Salmonella* bacteriophages, and bromide in the leachate of soil lysimeters (FC = fecal coliforms Pg = bacteriophages). Soils are Manawatu fine sandy loam (MW), Lismore shallow silt loam (LM), Templeton silt loam (TT), Aliamuri pumice soil (AM), Waitarere sandy recent soil (WT), Hamilton clay loam (HT), Netherton clayey soil (NT), Waihou silty allophane soil (WH), Waikiwi silt loam (WK), and Waikoikoi silt loam (WO); 1, 2, and 3 refer to lysimeter numbers. The pore volumes were calculated based on the total porosity of the soils.

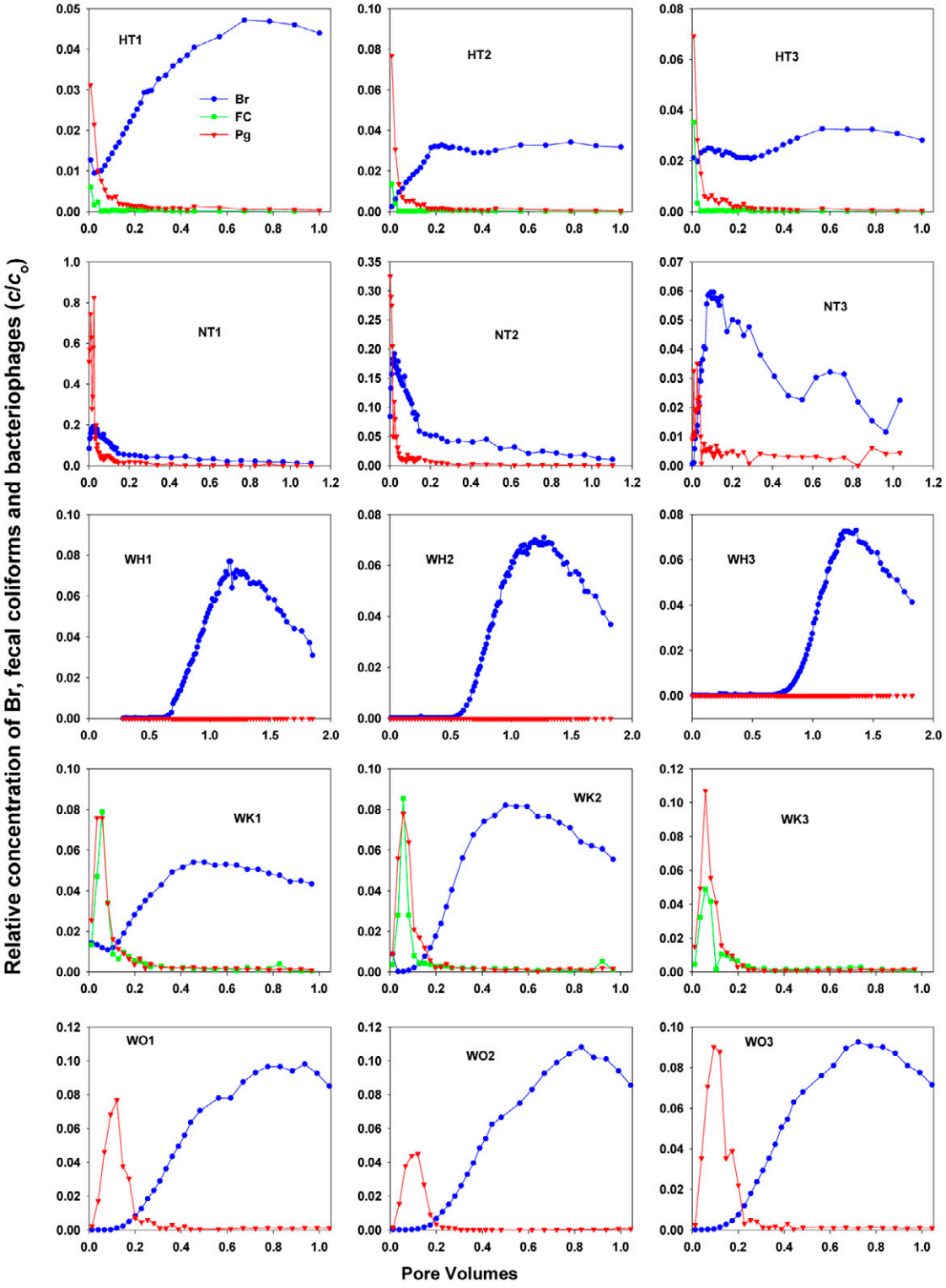

FIG. 1. Continued.

through high-permeability zones, while the long tail reflects solute exchange with low-permeability zones. The presence of aggregates, earthworm and root channels, and finger flow due to water repellency is believed to be responsible for the observed preferential flow in these soils.

In contrast, the peak concentrations of Br BTCs arrived later than 1 PV in Waihou allophanic soil. This later breakthrough of Br indicates that Br is not conservative in allophanic soil. Results of a batch test (our recent unpublished data) showed that Br is adsorbed on Waihou allophanic soil, with a distribution coefficient (K_d) of 0.06 L kg^{-1} in the top 15 cm of soil and 0.33 L kg^{-1} in the subsoil (these values were used in the modeling). Sorption of Br in an allophanic soil could be explained by its soil surface charge. Amorphous allophanic clays have a high isoelectric point of pH 6.0 (Cooper and Morgan, 1979), and Waihou soils have a topsoil field pH that is typically less than that (Table 2), so the allophane has a net positive surface charge (McLeod et al., 2001) and thus has an affinity for anions. Sorption of Br in allophanic soils has already been reported

TABLE 4. Observed relative transport and concentration reduction of microbes. There was complete removal in the Waihou silty allophanic soil with very fine structure.

Exp.†	Microbe pore vol. at c_{max}‡	Max. Br pore vol./ max. microbe pore vol.	c_{max}/c_o	Spatial coordinate	$\log(c_{max}/c_o)$	Removal rate	Quality of BTC§
				m		log¶ m^{-1}	
Netherton clayey soil (no bacteria data available)							
Pg1	0.01	2.30	8.25×10^{-1}	0.70	0.08	0.12	poor
Pg2	0.01	9.40	3.25×10^{-1}	0.70	0.49	0.70	poor
Pg3	0.01	7.30	3.50×10^{-2}	0.70	1.46	2.08	poor
Hamilton clay loam							
Pg1	0.01	86.54	3.13×10^{-2}	0.70	1.50	2.15	poor
Pg2	0.01	20.90	7.70×10^{-2}	0.70	1.11	1.59	poor
Pg3	0.01	50.77	6.94×10^{-2}	0.70	1.16	1.66	poor
FC1	0.01	86.54	6.07×10^{-3}	0.70	2.22	3.17	poor
FC2	0.01	20.90	1.36×10^{-2}	0.70	1.87	2.67	poor
FC3	0.01	50.77	3.51×10^{-2}	0.70	1.45	2.08	poor
Waikiwi silt loam (no Br data available for Lysimeter 3)							
Pg1	0.03	10.32	7.59×10^{-2}	0.47	1.12	2.38	good
Pg2	0.06	7.81	7.80×10^{-2}	0.47	1.11	2.36	good
Pg3	0.06	7.81	1.07×10^{-1}	0.47	0.97	2.07	good
FC1	0.06	6.19	7.87×10^{-2}	0.47	1.10	2.35	good
FC2	0.06	7.81	8.54×10^{-2}	0.47	1.07	2.27	good
FC3	0.06	7.81	4.88×10^{-2}	0.47	1.31	2.79	good
Waikoikoi silt loam (no bacteria data available)							
Pg1	0.12	6.47	7.68×10^{-2}	0.50	1.11	2.23	good
Pg2	0.12	6.91	4.51×10^{-2}	0.50	1.35	2.69	good
Pg3	0.09	7.12	9.02×10^{-2}	0.50	1.04	2.09	good
Lismore shallow silt loam over gravels							
Pg1	0.03	6.37	2.02×10^{-1}	0.70	0.69	0.99	poor
Pg2	0.03	8.11	2.00×10^{-2}	0.70	1.70	2.43	poor
Pg3	0.07	5.60	1.69×10^{-2}	0.70	1.77	2.53	good
FC1	0.03	6.37	2.02×10^{-2}	0.70	1.69	2.42	poor
FC2	0.03	8.11	5.75×10^{-3}	0.70	2.24	3.20	poor
FC3	0.03	13.52	2.86×10^{-5}	0.70	4.54	6.49	poor
Templeton silt loam							
Pg1	0.11	4.72	9.50×10^{-2}	0.40	1.02	2.56	good
Pg2	0.11	3.86	1.81×10^{-1}	0.40	0.74	1.85	good
Pg3	0.08	3.80	2.39×10^{-1}	0.40	0.62	1.56	ok
FC1	0.11	4.72	1.90×10^{-1}	0.40	0.72	1.80	good
FC3	0.08	3.80	3.07×10^{-1}	0.40	0.51	1.28	ok
Manawatu fine sandy loam							
Pg1	0.24	2.18	5.19×10^{-3}	0.70	2.28	3.26	poor
Pg2	0.39	1.47	2.08×10^{-2}	0.70	1.68	2.40	ok
Pg3	0.32	1.91	5.10×10^{-3}	0.70	2.29	3.28	good
FC1	0.67	0.80	6.07×10^{-7}	0.70	6.22	8.88	poor
FC2	0.28	2.03	2.02×10^{-7}	0.70	6.69	9.56	poor
FC3	0.15	3.94	2.02×10^{-7}	0.70	6.69	9.56	poor
Waitarere sandy recent soil (dune sand)							
Pg1	0.26	0.72	2.06×10^{-2}	0.70	1.69	2.41	good
Pg2	0.37	0.61	3.51×10^{-2}	0.70	1.45	2.08	good
Pg3	0.26	0.86	9.49×10^{-3}	0.70	2.02	2.89	good
FC1	0.26	0.72	2.50×10^{-2}	0.70	1.60	2.29	good
FC2	0.37	0.61	4.27×10^{-2}	0.70	1.37	1.96	good
FC3	0.26	0.86	1.15×10^{-2}	0.70	1.94	2.77	good
Atiamuri pumice soil (no bacteria data available)							
Pg1	0.37	1.84	2.33×10^{-12}	0.70	11.63	16.62	poor
Pg2	0.42	1.37	6.00×10^{-13}	0.70	12.22	17.46	poor
Pg3	0.89	0.76	9.44×10^{-12}	0.70	11.03	15.75	ok

† Pg, bacteriophages; FC, fecal coliforms; 1, 2, and 3 refer to lysimeter numbers.
‡ c = concentration.
§ Determined by the completeness of the breakthrough curve (BTC).
¶ The unit log is $\log_{10}$ reduction in maximum concentration compared with the original concentration, $\log_{10}(c_{max}/c_o)$.

(Close et al., 2003). Although Br was also adsorbed in Atiamuri pumice soil (K_d = 0.01 L kg^{-1} for topsoil and 0.03 L kg^{-1} for subsoil) and Hamilton clay loam (K_d = 0.00 for topsoil and 0.03 L kg^{-1} for subsoil), the degree of adsorption was not high enough to cause a shift in the concentration peak.

Model-simulated Br concentrations are compared with observed concentrations in Fig. 2, selecting one lysimeter per soil type. The MIM-derived dispersivity for Br transport (Table 5) is on average 7% of the lysimeter length (SD = 6%, n = 29). Although there is a lot of variability in the data, dispersivity of Br transport appears to be inversely correlated with the measured macroporosity (Fig. 3). This is expected, as with an increase in macropores in the soils, convection flow would be more dominant and less dispersion would occur.

The fraction of mobile water, θ_m/θ, (Table 5) accessible for Br transport is on average 0.45 (SD = 0.23, n = 29). The θ_m/θ values are lowest for Netherton clayey soil and Hamilton clay loam, implying that, in highly heterogeneous soils, only a small fraction of water is needed to leach solute contaminants. This suggests that the fraction of mobile water reflects the degree of soil heterogeneity. The fact that the pattern of model-predicted mobile water content agrees with measured macroporosity (Fig. 3) also supports this comment.

The mass exchange rate, α, (Table 5) is much higher for allophanic and pumice soils than for other soils, indicating that Br transport is not at equilibrium in volcanic soils. In contrast, the α value is close to zero for dune sand, suggesting that Br transport in uniform dune sand was under equilibrium conditions. The relatively symmetric shape of Br BTCs with little tailing for this soil also supports this conclusion. The relationship between α and macroporosity is not clear in this study (Fig. 3).

Transport of Fecal Coliforms and Bacteriophages

Breakthrough Curves

The BTCs of microbial tracers show a very different pattern from those of Br (Fig. 1). In comparison with Br, the BTCs of microbial tracers commonly peak much earlier and are much less spread out. Many others (e.g., Shelton et al., 2003; Guber et al., 2005; Levy et al., 2007) have also observed an earlier breakthrough (or velocity enhancement) of microbes than conservative solutes in undisturbed soils. Except for Waitarere dune sand and phages in one lysimeter in Atiamuri pumice soil, all microbial BTCs finished ahead of the front of the Br BTCs. This suggests that microbes are transported through a macropore network and that the soil matrix has little impact on their transport. Similarly, others have also commented that microbial transport occurs primarily through interconnected large pores (Wollum and Cassel, 1978) and almost follows a piston flow (Germann et al., 1987). The effect of macropores on colloid transport in intact cores was also suggested in the review of DeNovio et al. (2004).

Figure 4 shows that MIM-simulated BTCs fit well the observed data. As it is superfluous to present all model-simulated BTCs here, we present only one lysimeter for one soil type in

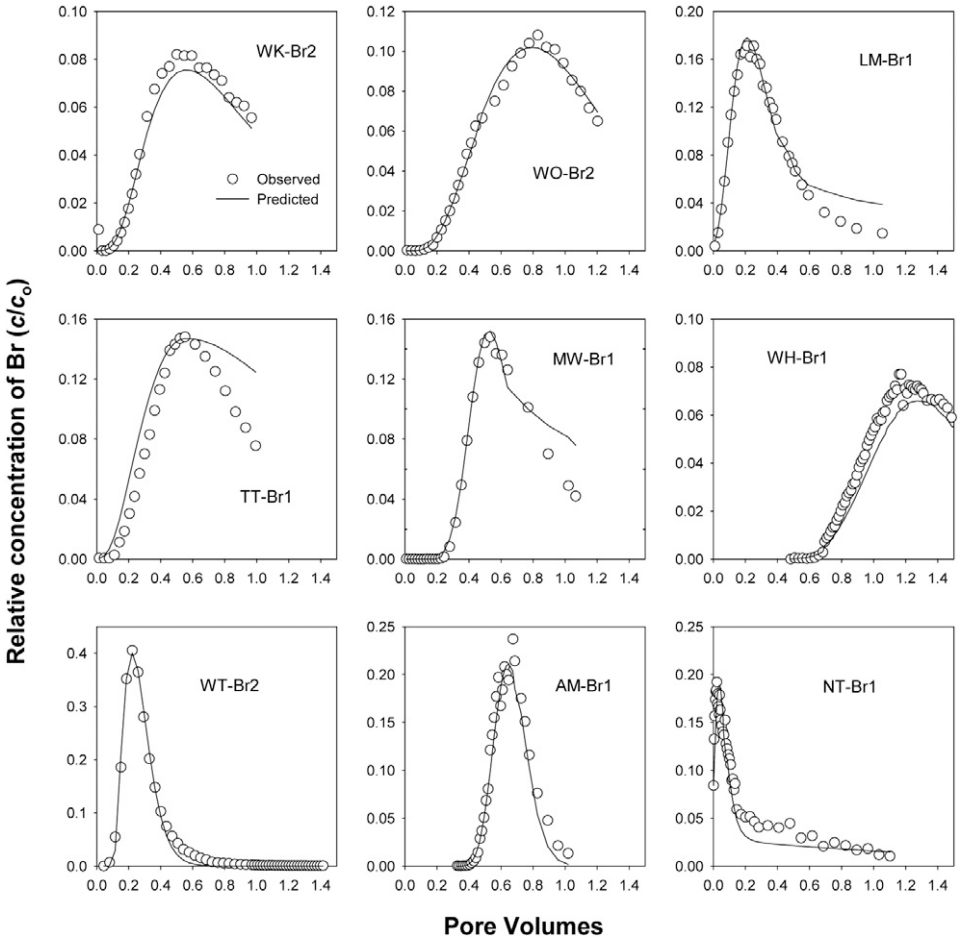

FIG. 2. Comparison of observed and mobile–immobile model (MIM) simulated Br concentrations in the leachate of soil lysimeters. Circles are observed data and solid lines are model-simulated data. Soils are Waikiwi silt loam (WK), Waikoikoi silt loam (WO), Lismore shallow silt loam (LM), Templeton silt loam (TT), Manawatu fine sandy loam (MW), Waihou silty allophane soil (WH), Waitarere sandy recent soil (WT), Aliamuri pumice soil (AM), and Netherton clayey soil (NT); 1, 2, and 3 refer to lysimeter numbers.

TABLE 5. Parameters† derived from Br breakthrough curves using the HYDRUS mobile–immobile model (MIM).

Soil	Lysimeter	Optimized from MIM				Calculated	
		ξ	θ_m	α	r^2	θ_m/θ	ξ/L
		cm		d^{-1}			
Netherton clayey gley loam	1	10.00	0.07	0.21	0.92	0.11	0.14
	2	4.00	0.10	0.15	0.87	0.15	0.06
	3	6.50	0.10	0.43	0.83	0.15	0.09
Hamilton clay loam	1	9.32	0.08	1.14	0.94	0.15	0.13
	2	4.76	0.08	0.67	0.91	0.15	0.07
	3	7.62	0.08	0.93	0.39	0.15	0.11
Waikiwi silt loam‡	1	11.87	0.43	0.15	0.97	0.84	0.25
	2	9.54	0.43	1.20	0.99	0.84	0.20
Waikoikoi silt loam	1	4.16	0.23	1.23	0.99	0.53	0.08
	2	2.37	0.14	1.73	0.99	0.32	0.05
	3	7.69	0.32	1.20	1.00	0.74	0.15
Lismore shallow silt loam over gravels	1	6.01	0.17	0.08	0.97	0.36	0.09
	2	6.77	0.18	0.12	0.95	0.38	0.10
	3	4.18	0.24	0.21	0.86	0.50	0.06
Templeton deep silt loam	1	3.00	0.17	1.85	0.93	0.45	0.08
	2	2.61	0.18	0.85	0.91	0.49	0.07
	3	4.30	0.19	0.53	0.91	0.51	0.06
Manawatu fine sandy loam	1	0.40	0.23	0.27	0.97	0.50	0.01
	2	0.35	0.23	0.26	0.95	0.51	0.01
	3	0.13	0.24	0.28	0.94	0.52	0.00
Waitarere recent sandy soil	1	5.07	0.28	0.00	0.98	0.46	0.07
	2	3.00	0.28	0.00	0.95	0.46	0.04
	3	5.00	0.28	0.01	0.99	0.47	0.07
Atiamuri pumice soil	1	0.43	0.44	2.36	0.97	0.80	0.01
	2	0.22	0.46	0.34	0.95	0.84	0.00
	3	0.88	0.44	3.82	0.93	0.80	0.01
Waihou allophanic	1	1.00	0.12	3.76	0.98	0.20	0.01
	2	1.79	0.13	4.00	0.98	0.20	0.03
	3	0.74	0.25	3.76	0.93	0.41	0.01

† Data for Waikiwi silt loam Lysimeter 3 were not available.
‡ ξ, dispersivity; θ_m, mobile water content; L, column length; θ, total porosity; α, mass exchange rate between two regions.

Fig. 4, which was typical (representative) of other BTCs.

Double peaks are displayed in the bacteria BTCs of Hamilton clay loam and the phage BTCs of Atiamuri pumice soil, with the first peaks showing velocity enhancement and second peaks showing later arrival than Br. The first peaks of the bacteria BTCs of Hamilton soil are concurrent with those of phages that do not show any second peak. As the concentrations of the bacteria in the second peaks are about two orders of magnitude lower than those of the first peaks, they are not noticeable on the scale shown in Fig. 1. For the pumice soil, the first peak was the major peak in one column and the minor peak in another column. Multiple peaks in a microbial BTC suggest the presence of multiple flow paths of contrasting permeability for microbial transport within the heterogeneous medium or detachment of adsorbed microbes associated with a change in leaching conditions. As leaching conditions were stable, multiple flow paths are the probable explanation.

Mobile–Immobile Water, Velocity Enhancement

In comparison with Br, the fraction of mobile water, θ_m/θ (Table 6), is much smaller for microbial BTCs that exhibit velocity enhancement (i.e., excluding Waitarere sandy recent and Atiamuri pumice soil), on average only 0.19 (SD = 0.09, n = 29), in a narrow range of 0.08 to 0.24, except for Manawatu fine sandy loam (0.40–0.45). This is because microbes can only access a smaller range of larger pores due to size exclusion, unlike the Br solute, which can access a wider range of pore sizes. Slower velocity zones near the solid surfaces, which are accessible to Br and might be considered mobile for Br, would become inaccessible to microbes and would thus be treated as immobile. Water will then travel faster if going through a smaller porosity. Shelton et al. (2003) estimated that the porosity available for fecal coliform transport in undisturbed stony silt loam is 15% of the total water content, which is similar to our findings. In contrast, θ_m/θ is much larger for microbial BTCs that arrived later than Br (Table 6), on average 0.80 (SD = 0.10, n = 7). Like the results from Br, the pattern of predicted mobile water content agrees with the measured macroporosity (Fig. 3).

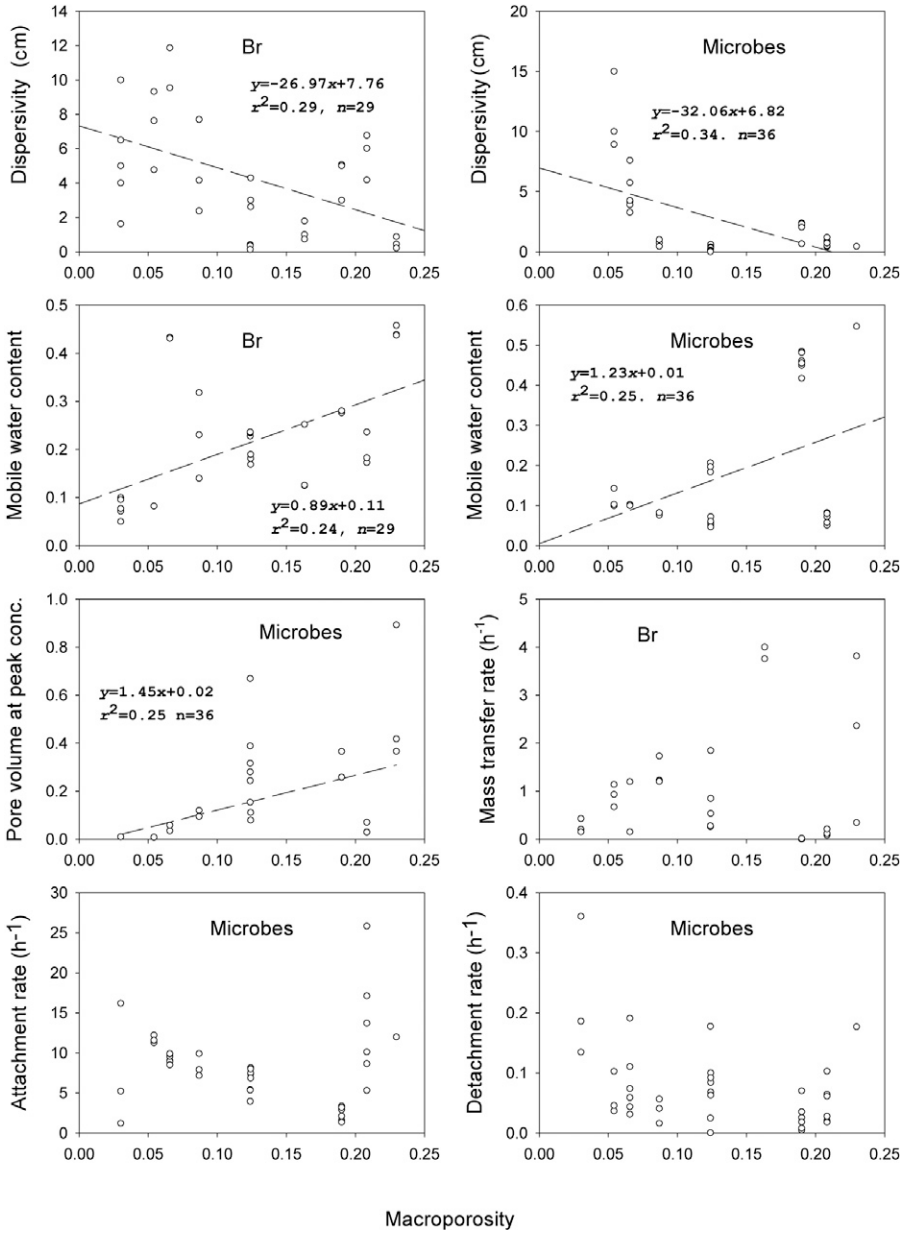

FIG. 3. Correlations of macroporosity with transport parameters.

The $\theta_{m\text{-}Br}/\theta_{m\text{-}microbe} = v_{m\text{-}microbe}/v_{m\text{-}Br}$ ratios derived from MIM (Table 7) suggest that the transport of the microbes was always faster (mostly two to four times) than that of Br, except in Waitarere dune sand and one lysimeter of Atiamuri pumice soil. This indicates that velocity enhancement is a common phenomenon in the structured soils investigated here because the microbes are excluded from smaller pores in the soil medium. The degree of velocity enhancement, as measured by $\theta_{m\text{-}Br}/\theta_{m\text{-}microbe}$, is in the order of: Waikiwi silt loam (4) > Lismore shallow silt loam, Waikoikoi silt loam, and Templeton silt loam (2–4) > Hamilton clay loam, Manawatu fine sandy loam, and Netherton clayey soil (1–2) > Waitarere sandy recent soil (<1). For a particular lysimeter, there was little difference in speed between bacteria and phages as reflected in their similar $\theta_{m\text{-}Br}/\theta_{m\text{-}microbe}$ ratios (Table 7). Our finding on the faster velocity of microbes than Br is similar to the observation of Shelton et al. (2003), who conducted a lysimeter study. They found the average velocity of coliform bacteria leaching from 90-cm-long undisturbed stony soil was about seven times greater than the average pore velocity.

The above relative speed, as measured by the $\theta_{m\text{-}Br}/\theta_{m\text{-}microbe}$ ratio, relates to the mean velocity or center of mass. If the relative speed is examined using the ratio of pore volume at peak concentration of Br to that of a microbial tracer (Table 4), which reflects macropore velocity (De Jonge et al., 2004), the relative speed for microbial transport is much greater than for Br. It is in the order of Hamilton clay loam (21–87) > Lismore shallow silt loam over gravels, Waikiwi silt loam, and Waikoikoi silt loam (6–14) > Templeton silt loam (4–5) > Manawatu fine sandy loam and Atiamuri pumice soil (1–2) > Waitarere sandy recent soil (<1). The ratio for heterogeneous Netherton clayey soil varies between 2 and 9.

The $\theta_{m\text{-}Br}/\theta_{m\text{-}microbe}$ ratios for Waitarere dune sand were <1 (Table 7), suggesting that the transport velocity of microbes in Waitarere dune sand was slower than that of Br. This is probably because this soil has only minor pedological development, with a single-grain structure throughout. It is therefore more uniform than other soils, thus matrix flow plays an important role in microbial transport in the dune sand. The $\theta_{m\text{-}Br}/\theta_{m\text{-}microbe}$ ratio was also <1 for one lysimeter of Atiamuri pumice soil, suggesting that transport of phages was slower than that of Br in one of the pumice soil lysimeters.

Dispersion

Dispersivity values derived from most microbial BTCs (Table 6) are much smaller than those from Br BTCs (average $\xi_{Br}/\xi_{microbe}$ = 4.69, SD = 3.85, n = 35, Table 7) as a result of a narrower pore network involved in microbial transport. The lower dispersion in microbial transport compared with solute transport was also observed in the lysimeter study of Shelton et al. (2003) with an undisturbed silt loam. There are a few exceptions of greater ξ values for microbes than for Br, but this is believed to be an artifact of the erratic data for microbial BTCs.

The dispersivity/transport distance ratio for microbe transport was less than that for Br, on average 5% (SD = 6%, n = 36, Table 6), mostly ranging from 1 to 3%. As for Br, the dispersivity of microbe transport appears to be inversely correlated with macroporosity (Fig. 3).

Leaching Vulnerability of Fecal Coliforms and Bacteriophages

The PV at maximum concentration (c_{max}) (Table 4), the normalized volume of water required for reaching maximum concentration in the leachate, reflects the leaching vulnerability of

microbes through soils. The leaching vulnerability for microbes is in the order of: Netherton clayey soil and Hamilton clay loam (0.01) > Lismore shallow silt loam over gravels (0.03) > Waikiwi silt loam (0.06) > Templeton silt loam and Waikoikoi silt loam (0.08–0.12) > Manawatu fine sandy loam, Waitarere dune sand, and Atiamuri pumice soil (0.24–0.89) > Waihou allophanic soil (not leached). This sequence is consistent with a general trend of decreasing heterogeneity of soil structure. This suggests that heterogeneous soils, due to the presence of preferential flow paths, are more vulnerable to microbial leaching through soils. The small PV values at c_{max} (Table 4) suggest that, in structured heterogeneous soils, even a very small amount of water in the unsaturated soils could lead to a rapid and significant leaching of microbes through bypass flow. This result suggests that the leaching vulnerability of microbes into shallow water bodies could be the greatest in clayey soils, which often crack, followed by silt loam over gravels, silt loam, sandy loam, dune sand, pumice soil, and allophanic soil. Figure 3 shows that there is a positive relationship between macroporosity and leaching vulnerability of microbes.

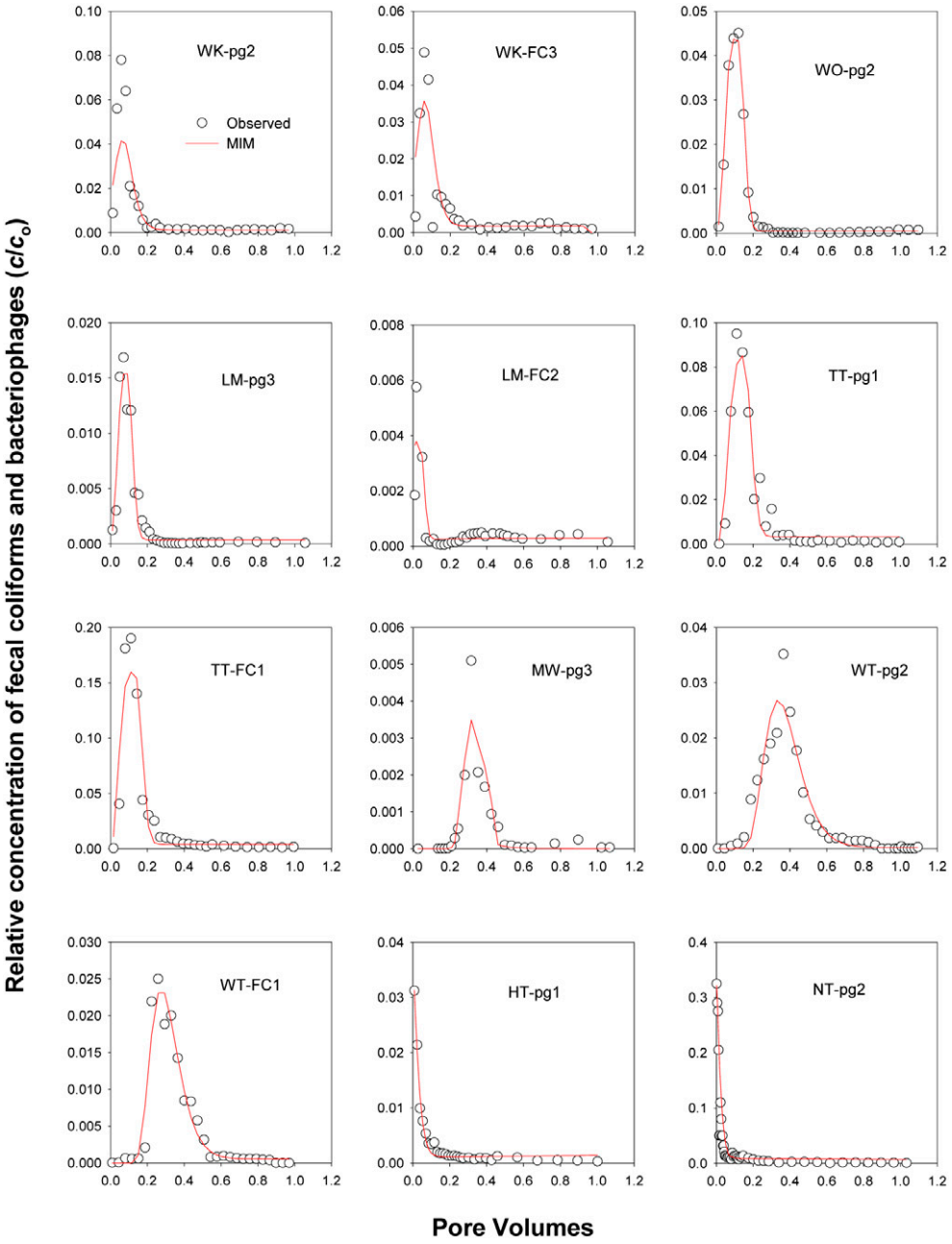

FIG. 4. Comparison of observed and simulated concentrations of fecal coliforms (FC) and *Salmonella* bacteriophages (pg) in the leachate of soil lysimeters. Empty dots are observed data. The red solid lines are data fitted with the mobile–immobile model. Soils are Lismore shallow silt loam (LM), Templeton silt loam (TT), Manawatu fine sandy loam (MW), Waitarere sandy recent soil (WT), Hamilton clay loam (HT), and Netherton clayey soil (NT); 1, 2, and 3 refer to lysimeter numbers.

Removal of Fecal Coliforms and Bacteriophages

Reduction in Concentration and Mass

The greatest reduction in phage concentration was observed in Waihou allophanic soil—no phages were detected in the leachate from this soil. As mentioned above, previous studies have suggested that allophanic clays have a net positive charge in the topsoil, which prompts bonding of negatively charged phages. In addition, allophane has a very large surface area, 700 to 900 m^2 g^{-1} (Aislabie et al., 2001), further enhancing the bonding of phages with the soil medium. The second greatest reduction in phage concentrations occurred in the Atiamuri pumice soil [16–18 log m^{-1}, where the unit log is $\log_{10}(c_{max}/c_o)$, Table 4]. The presence of allophanic clay, even though just a small fraction in the pumice soil (Table 1), is believed to have played an important role in removal of phages in Waihou allophanic soil and Atiamuri pumice soil.

For most other soils, the reduction in fecal coliform and phage concentrations is about 2 to 3 log m^{-1} (Table 4), except for a greater removal of fecal coliforms in Manawatu fine sandy loam (9–10 log m^{-1}). The clayey gley soil had the least reduction in microbial concentration (0.1–2 log m^{-1}). This finding agrees with the inference of Wells (1973) that tephric soils have the best characteristics for effluent disposal, and soils with cracks, such as gley soils, are not suitable for effluent disposal.

Attachment and Removal Rates

The MIM-derived k_{att} rates (Table 6) are the highest for the soils of volcanic origin (complete removal in Waihou allophanic soil, 11–12 d^{-1} for Atiamuri pumice soil and Hamilton clay loam). The high level of attachment associated with volcanic soils is attributed to their large surface areas, which are variably charged, as discussed above. Silt loams of greywacke ori-

gin (Templeton silt loam, Waikoikoi silt loam, and Waikiwi silt loam) have the second highest k_{att} rates, ranging from 7 to 10 d^{-1}. In these soils, soil particles are often coated with a thin layer of Fe and Mn oxides, which promote attachment of microbes. The lowest k_{att} rates are found for the granular young soils (1–3 d^{-1} for Waitarere dune sand soil, 4–5 d^{-1} for Manawatu fine sandy loam). This is expected because granular soils are generally weaker adsorbents for microbes than clays and minerals (Sobsey et al., 1980; Moore et al., 1981). In addition, much less coating is present on the grain (largely silica) surfaces of these young soils. Heterogeneous soils have the most variable k_{att} rates (1–16 d^{-1} for Netherton clayey gley loam, 5–26 d^{-1} for Lismore shallow silt loam), probably due to an uneven distribution of readily available attachment sites. The explanation for their possibly high k_{att} rates is that clays and metal oxides have a strong affinity for microbes. In Lismore silt loam over gravels, >50% of the stones (derived from greywacke) show coatings. The relationship between k_{att} and macroporosity is not clear (Fig. 3). These results suggest that attachment and thus removal of microbes is predominantly influenced by soil chemistry, particularly the lithologic origin.

The above patterns are also applicable for k_{tot} rates determined from MIM (Table 6). The k_{tot} rates derived from MIM are the highest in soils of volcanic origin (allophanic soil > 0.51 h^{-1}, pumice soil 0.51 h^{-1}, and clay loam containing tephra 0.48–0.52 h^{-1}), second in silt loams of greywacke origin (0.30–0.43 h^{-1}), weakest in granular young sandy soils (fine sandy loam 0.17–0.23 h^{-1}, dune sand soil 0.07–0.15 h^{-1}), and the most variable in heterogeneous soils (clayey gley loam 0.06–0.68 h^{-1}, silt loam over gravels 0.23–1.09 h^{-1}).

The k_{det} rate (mean 0.08 d^{-1}, SD = 0.07, n = 36, Table 6) is on average only 1% of the k_{att} rate (mean 8.28 d^{-1}, SD = 4.95, n = 36). The very low k_{det} rates suggest that detachment of microbes in the structured soils investigated here was negligible and that microbial attachment could be considered to be irreversible. Other researchers have also commented that microbial attachment to porous media is primarily an irreversible process (Yao et al., 1971; Rajagopalan and Tien, 1976; Pieper et al., 1997).

Relative Contribution of Individual Processes

The inactivation rate derived from the batch incubation tests conducted at 15°C (a similar temperature to that used in the lysimeter experiments) was 0.298 d^{-1} for fecal coliforms and 0.214 d^{-1} for the phages. The contribution of inactivation to the total removal rate was, on average, 3.78% for phages (SD = 3.17%, n = 25) and 5.54% for fecal coliforms (SD = 5.34%, n = 11).

Although the contribution of inactivation to total removal could be examined, the individual contributions of straining and air–water interaction effects could not be quantified. The effects of these processes are combined in the total removal rates. We could only identify the possible presence of these processes.

Straining occurs when the ratio of the colloid to medium grain diameter, D_p/d, is >0.5% (Bradford et al., 2004), and will be significant when the ratio is >8% (McDowell-Boyer et al., 1986). Using these criteria, straining may be expected in the transport of fecal coliforms through all of the lysimeters (D_p/d > 0.69%, Table 2), and could be significant in clayey soil, clay loam, and silt loam ($D_p/d \geq 10\%$). Based on these criteria, straining may have also occurred in the transport of phages in clayey soil and clay loam (D_p/d = 0.7–3%). It should be noted that the criteria for straining described above were developed from media with uniform grain sizes and that these criteria do not consider the effect of soil aggregates and macropores. Although the grain diameter may be small in aggregated soils, the effective diameter may be a lot larger because the grains are clumped and the microbes are not interacting with single grains. The large interconnected macropores developed in aggregated soils could allow the rapid transport of microbes with little straining. Therefore, the effect of soil aggregation and macropores on straining is unknown, especially for clayey soils, which naturally develop

TABLE 6. Transport parameters† derived from bacteriophage (Pg) and fecal coliform (FC) breakthrough curves using the HYDRUS mobile–immobile model (MIM).

Soil	Microbe	Lysimeter	Optimized from MIM					Calculated			
			ξ	θ_m	k_{att}	k_{det}	r^2	θ_m/θ	ξ/L	k_{tot}	k_{det}/k_{att}
			cm		d^{-1}	d^{-1}				h^{-1}	
Netherton clayey gley loam	Pg	1	1.63	0.05	1.21	0.19	0.81	0.08	0.02	0.06	0.153
		2	10.00	0.05	5.23	0.13	0.87	0.08	0.14	0.23	0.026
		3	5.00	0.08	16.18	0.36	0.37	0.12	0.07	0.68	0.022
Hamilton clay loam	Pg	1	8.92	0.14	12.21	0.10	0.98	0.24	0.13	0.52	0.008
		2	10.00	0.10	11.28	0.04	0.99	0.17	0.14	0.48	0.003
		3	15.00	0.10	11.58	0.05	0.99	0.17	0.21	0.49	0.004
Waikiwi silt loam	Pg	1	7.60	0.10	9.02	0.11	0.87	0.19	0.16	0.38	0.012
		2	4.16	0.10	8.91	0.04	0.87	0.20	0.09	0.38	0.005
		3	3.28	0.10	8.50	0.03	0.54	0.19	0.07	0.36	0.004
	FC	1	5.74	0.10	9.54	0.06	0.77	0.19	0.12	0.41	0.006
		2	3.90	0.10	9.85	0.19	0.60	0.19	0.08	0.42	0.019
		3	4.26	0.10	9.90	0.07	0.74	0.19	0.09	0.43	0.007
Waikoikoi silt loam	Pg	1	0.74	0.08	7.91	0.04	0.97	0.19	0.01	0.34	0.005
		2	0.44	0.08	9.91	0.02	1.00	0.18	0.01	0.42	0.002
		3	1.00	0.08	7.15	0.06	0.94	0.19	0.02	0.31	0.008
Lismore shallow silt loam over gravels	Pg	1	0.88	0.06	5.31	0.02	0.93	0.12	0.01	0.23	0.004
		2	1.19	0.07	10.11	0.02	0.98	0.15	0.02	0.43	0.002
		3	0.44	0.08	8.64	0.03	0.93	0.17	0.01	0.37	0.003
	FC	1	0.42	0.05	13.71	0.06	0.98	0.11	0.01	0.58	0.005
		2	0.49	0.06	17.12	0.06	0.77	0.12	0.01	0.73	0.004
		3	0.72	0.08	25.79	0.10	0.69	0.17	0.01	1.09	0.004
Templeton deep silt loam	Pg	1	0.45	0.07	7.38	0.07	0.93	0.20	0.01	0.32	0.009
		2	0.56	0.06	8.15	0.06	0.92	0.15	0.01	0.35	0.008
		3	0.60	0.05	7.49	0.10	0.94	0.14	0.01	0.32	0.013
	FC	1	0.39	0.06	8.00	0.08	0.90	0.16	0.01	0.35	0.011
		3	0.30	0.05	6.84	0.09	0.93	0.12	0.01	0.30	0.013
Manawatu fine sandy loam	Pg	1	0.13	0.18	5.41	0.18	0.80	0.40	0.00	0.23	0.033
		2	0.07	0.21	3.94	0.02	0.53	0.45	0.00	0.17	0.006
		3	0.00	0.20	5.35	0.00	0.86	0.43	0.00	0.23	0.000
Waitarere recent sandy soil	Pg	1	0.67	0.42	2.91	0.07	0.88	0.70	0.01	0.13	0.024
		2	2.38	0.48	1.58	0.00	0.90	0.81	0.03	0.07	0.003
		3	2.20	0.46	3.37	0.03	0.78	0.77	0.03	0.15	0.008
	FC	1	2.38	0.45	2.10	0.02	0.95	0.76	0.03	0.10	0.009
		2	2.33	0.48	1.36	0.01	0.89	0.81	0.03	0.07	0.006
		3	2.04	0.46	3.19	0.04	0.77	0.76	0.03	0.15	0.011
Atiamuri pumice soil	Pg	3	0.45	0.55	11.98	0.18	0.49	1.00	0.01	0.51	0.015

† ξ, dispersivity; θ_m, mobile water content; θ, total porosity; L, column length; k_{att}, attachment rate; k_{det}, detachment rate; k_{tot}, total removal rate.

cracks within the soil. This could not be assessed in this study as no attachment profiles were measured during the experiments.

The air–water interface plays an important role in the enhanced removal of microbes under unsaturated conditions compared with saturated conditions (Chu et al., 2003; Torkzaban et al., 2006). Retention of microbes increases as water content decreases (Jin et al., 2000; Han et al., 2006; Torkzaban et al., 2006). Evaluating the effect of the air–water interaction is difficult in this study, however, as water contents were not measured.

Conclusions

Our modeling results showed generally smaller values of mobile water content and dispersivity for microbial tracers compared with Br. This indicates that the transport of microbes in highly structured soils is predominantly controlled by macropores because they are excluded from small pores, resulting in the observed earlier breakthrough than Br (i.e., velocity enhancement). Thus transport of microbes is commonly more convective and less dispersive than that of Br. An exception to this was in uniform, single-grain dune sand, where the velocity of microbial tracers was slower than that of Br.

Breakthrough of both microbes and Br occurred at PV << 1, suggesting that rapid and significant leaching of microbial and solute contaminants could occur through bypass flow in structured soils with a small amount of water. An exception is for the allophanic soil, in which Br was adsorbed and the concentration peaked at >1 PV. Leaching vulnerability seems to relate to soil heterogeneity, being greatest in clayey and clay soils with cracks, followed by silt loam over gravels, silt loams, sandy soil, dune sand soil, pumice soil, and allophanic soil.

For both Br and the microbes, the general pattern of predicted mobile water content agrees with measured macroporosity, which is positively related to leaching vulnerability but negatively related to dispersivity. This suggests that soil structure plays the most important role in the transport of microbes and Br. The relationship between macroporosity and the mass exchange rate, however, was not clear in this study. The mass exchange rate between mobile and immobile water regions was greatest in volcanic soils and lowest in granular, young sandy soils.

Soil lithology has the greatest influence on the attenuation of microbes (attachment, removal, and reduction). The relationship between the attachment rate and macroporosity is not clear. The total removal rates (the sum of attachment rate and inactivation rate) derived from the MIM were highest in soils of volcanic origin (allophanic soil >0.51 h^{-1}, pumice soil 0.51 h^{-1}, and clay loam containing tephra 0.48–0.52 h^{-1}), second highest in silt loams of greywacke origin (0.30–0.43 h^{-1}), lowest in granular, young sandy soils (fine sandy loam 0.17–0.23 h^{-1}, dune sand soil 0.07–0.15 h^{-1}), and most variable in heterogeneous soils (clayey gley loam 0.06–0.68 h^{-1}, silt loam over gravels 0.23–1.09 h^{-1}).

The reduction in microbial concentration was the greatest in the allophanic soil (total removal) and second greatest in the pumice soil (16–18 log m^{-1}). The least reduction in phage concentrations occurred in clayey gley soil (0.1–2 log m^{-1}). For most other soils, fecal coliforms and phages had a concentration reduction of about 2 to 3 log m^{-1}, except that the fine sandy loam had a greater concentration reduction for fecal coliforms (9–10 log m^{-1}). These removal rates represent a reduction in microbial concentration on a natural log scale, and it needs to be divided by a factor of 2.3 to convert them to $\log_{10}$. The detachment rate of microbes was, on average, only 1% of the attachment rate, suggesting that detachment of microbes was negligible and attachment of microbes could thus be treated as irreversible.

The results of data analysis and modeling obtained in this study help our understanding of leaching vulnerability of microbes and the efficiency of effluent treatment for microbial removal in different soil media. We believe that this information could be applicable for soils under similar conditions, and that the results summarized from this study could provide useful information to regulatory agencies to more effectively manage land use and development and evaluate the risk of groundwater contamination.

These results, however, are applicable only for soils above the vadose zone (the zone between the soil root zone and the groundwater table). The ability of the vadose zone to remove microbes is expected to be much less effective. Thus, the reduction rates derived from soils cannot be extrapolated to vadose-zone media,

TABLE 7. Relative movement of bacteriophages (Pg) and fecal coliforms (FC) related to Br.

Soil	Microbe	Lysimeter	$\theta_{m\text{-}Br}/\theta_{m\text{-}micro}$†	ξ_{Br}/ξ_{micro}†
			Earlier arrival than Br	
Netherton clayey gley loam	Pg	1	1.44	6.14
		2	2.00	0.40
		3	1.25	1.30
Hamilton clay loam	Pg	1	1.04	1.05
		2	2.34	0.48
		3	2.11	0.51
Waikiwi silt loam	Pg	1	4.33	1.56
		2	4.19	2.29
		3	4.31	2.91
	FC	1	4.33	2.07
		2	4.31	2.45
		3	4.31	2.24
Waikoikoi silt loam	Pg	1	2.81	5.60
		2	1.83	5.40
		3	3.87	7.69
Lismore shallow silt loam over gravels	Pg	1	3.01	6.82
		2	2.53	5.67
		3	2.87	9.49
	FC	1	3.42	14.22
		2	3.16	13.74
		3	2.96	5.82
Templeton deep silt loam	Pg	1	2.32	6.59
		2	3.20	4.65
		3	3.65	7.16
	FC	1	2.79	7.71
		3	4.09	14.18
Manawatu fine sandy loam	Pg	1	1.24	3.10
		2	1.13	4.81
		3	1.20	–
			Later arrival than Br	
Waitarere recent sandy soil	Pg	1	0.66	7.60
		2	0.57	1.26
		3	0.60	2.27
	FC	1	0.61	2.13
		2	0.57	1.29
		3	0.61	2.45
Atiamuri pumice soil	Pg	3	0.80	0.98

† $\theta_{m\text{-}Br}/\theta_{m\text{-}microbe}$, ratio of mobile water content of Br to that of microbes, reflecting the degree of velocity enhancement in the transport of microbes (Eq. [7]); $\xi_{Br}/\xi_{microbe}$, ratio of dispersivity of Br to that of microbes, reflecting the degree of pore-size exclusion in the transport of microbes.

i.e., for estimation of vertical separation distances. Further study on microbial removal in vadose zone media is needed.

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