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## TRANSPORT AND FATE OF MICROORGANISMS IN POROUS MEDIA: A THEORETICAL INVESTIGATION

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### ABSTRACT

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Bacteria and viruses found in groundwater are a proven health hazard as evidenced by the large number of outbreaks of water-borne diseases caused by contaminated groundwater. To analyze the fate of biological contaminants in soils and groundwater, we studied various transport processes including dispersion, convection, Brownian motion, chemotaxis and tumbling of bacteria. The differences between bacteria and viruses in their transport mechanisms, decay and growth kinetics have also been investigated. It has been shown that the rate of deposition terms can be incorporated by a first-order and an adsorption isotherm for bacteria and viruses, respectively. The movement of bacteria is coupled with the transport of a bacterial nutrient present in seeping wastewater.

### 1. INTRODUCTION

Approximately half of the population in the U.S.A. relies on groundwater for its drinking water. Despite its importance, groundwater is one of the least understood of our major natural resources. Groundwater will be vulnerable to even more widespread contamination in the future as population and industry continue to grow. New instances of domestic pollution of groundwater are reported with increasing frequency and are attributed to the following causes: accidental breakage of sewers, filtration from an estimated 16.6 million residential septic tanks and cesspools, rain infiltrating through sanitary landfills, artificial recharge of groundwater aquifers by treated sewage water and, most important of all, land application of municipal wastewater. This problem is also crucial in developing municipal water supplies by extracting water from wells located adjacent to polluted streams and rivers.

Within the past decade, there has been an increased interest in land application which is the practice of applying wastewater to the natural soil surface (Harlin, 1981). The Federal Water Pollution Control Act Amendments of 1972 encourage the use of this practice by stating:

"it is the national goal that the discharge of pollutants into the navigable waters be eliminated by 1985."

Following the passage of this legislation, the Environmental Protection Agency (E.P.A.) adopted the policy that land application must be evaluated as an alternative for all wastewater treatment systems funded by the federal government. So, the U.S.A. is strongly committed to the use of land application where feasible and appropriate. In addition to this kind of application, recharge of groundwater aquifers by treated sewage effluents has been practiced in numerous places in the country where water resources are scarce and reclamation is necessary (Shean, 1977). Also, most residential septic tanks and cesspools are not lined with concrete or other materials to prevent the pollutants from leaking into the soil and, because all earth materials are permeable to some degree, wastes leak into the sub-surface. Once in the ground the wastes move through the soil until they enter the groundwater system that may be in use as sources of water supply (Fig. 1). While natural processes can in some cases help to reduce the pollution, most biological contaminants can travel through the earth until they either enter someone's water well or are discharged into a stream. It is evident that microbial contamination of groundwater does occur when human wastes enter into the soil and can travel long distances in groundwater under proper conditions. Geraghty and Miller (1978) report that across the U.S.A. there are four counties (Nassau, Suffolk, New York; Dade, Florida; Los Angeles, California) each with more than 100,000 housing units served by septic tanks, with an additional 23 counties with more than 50,000 house installations.

Mack et al. (1972) isolated polio virus from a 30-m deep drinking water

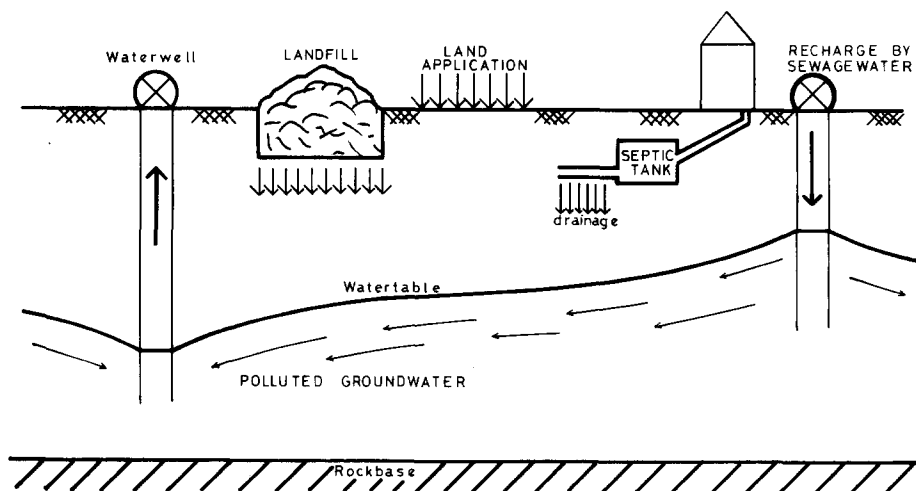


Fig. 1. Biological contamination sources of groundwater.

well located ~90 m from the edge of a wastewater drain field. Schaub and Sorber (1977) and Moore et al. (1981) reported virus isolations in groundwater beneath land application sites receiving sewage effluents. Viruses were recovered at depths of 30 m and lateral distances of 180 m. In a field study in Long Island, New York (Vaughn et al., 1981), 12 m deep monitoring wells yielded enteroviruses with relatively high frequency, even at a distance of 45 m from the septage source. Gerba et al. (1975) noted in a review the movement of coliform bacteria in a variety of soils for distances ranging from 1 to 450 m. Hagedorn (1981) listed various field studies where bacterial-contaminated groundwater traveled laterally 1 to 830 m through soil. Romero (1970) reviewed the case studies of biological groundwater pollution until 1970.

Although some authors found the microbial mass transport negligible for granular filters and soil-microbial mass systems (e.g., Wollum and Cassel, 1978; Sykes et al., 1982), many others considered the transport of microorganisms the most significant and used bacteria and viruses to trace groundwater movement in much the same manner as chemical tracers are used. A review presented by Keswick et al. (1982) finds that bacterial viruses appear to be the microorganisms most suited as a microbial tracer because of their size, ease of assay and lack of pathogenicity.

The present trend of disposing of wastewater by allowing it to infiltrate the soil brings a new dimension to our traditional environmental problems. Sewage contains many pathogenic microbial organisms with bacteria and virus probably being the most prevalent. De Walle et al. (1980) reported that even deep aquifers (150 m) that are predominantly used by public systems have been contaminated by septic tank leachfields. The results of a groundwater study in the Milwaukee, Wisconsin, area show that bacterial contamination is, in part, the result of leakage from sewer lines (Eisen and Anderson, 1979). Hain and O'Brien (1980) documented movement of indigenous enteric viruses into shallow groundwater. These few examples are among many other cases reported in the literature. Due to the seriousness of the problem, E.P.A. recently sponsored a conference (Conference on Microbial Health Considerations of Soil Disposal of Domestic Wastewaters, May 11–12, 1981, Norman, Oklahoma) on this topic. Speakers emphasized that the microbial contamination of groundwater is a serious problem that can result in large outbreaks of waterborne disease. The most important pathogenic bacteria and viruses which might be transported in the groundwater are *Salmonella* sp., *Shigella* sp., *Escherichia coli*, *Vibrio* sp., and viruses, hepatitis, Norwalk-like agent, echovirus, poliovirus, and coxsackievirus (Gerba, 1981). These pathogens have been associated with a wide variety of diseases, including typhoid, gastroenteritis, diarrhea, hepatitis, gastrointestinal illness, etc. Wilson et al. (1981) reported that of all water-borne disease outbreaks of 1971–1979 in the U.S.A., 42% were attributed to contaminated groundwater.

As is apparent from this short review, bacteria and viruses in groundwater

pose a proven hazard to health as evidenced by the large number of outbreaks of water-borne disease caused by contaminated groundwater.

It is the purpose of this paper to study the governing mechanisms for the transport and fate of biological pollutants (bacteria and viruses) in a porous medium.

## 2. TRANSPORT AND RETENTION MECHANISMS

The discussion in this study is at the macroscopic level. Without any special notation, all variables are at the macroscopic level. As an alternative the macroscopic mass-conservation equation can be developed starting from microscopic considerations and averaged over a representative elementary volume of the porous medium (see Bear, 1979; Bear and Corapcioglu, 1981).

The starting point is the mass-conservation equation for particles in a porous medium in a three-dimensional space:

$$R_a + \partial(\theta C)/\partial t = -\nabla \cdot J + R_{df} + R_{gf} \quad (1)$$

where  $R_a$  is the rate of deposition of particles on grains;  $R_{df}$  and  $R_{gf}$  are the decay and growth terms of the suspended particles, respectively;  $J$  is the specific mass discharge of suspended particles; and  $\theta$  denotes the volume occupied by the flowing suspension per unit total volume. Eq. 1 describes the temporal and spatial variations of particle concentration in a porous medium. The removal mechanisms of small suspended water-borne particles and the transport processes summed up in the term denoted by  $J$  will be described. Decay and growth kinetics will also be discussed.

The capture of suspended particles from water passing through soil (Spielman, 1977):

“is characterized by the simultaneous action of forces of fluid-mechanical origin along with the forces of other origin that act between the particle and collector.”

As bacteria and viruses are transported by a liquid flowing through porous media, they are removed by various mechanisms. The removal of bacteria is attributed to straining in the contact zones of adjacent pores, sedimentation in the pores, and adsorption. Adsorption is the primary removal mechanism for viruses due to their very small size.

### 2.1. Straining in the contact zones of adjacent pores

Straining takes place when a particle in suspension flowing through a pore is larger than the pore opening, so suspended microorganisms are accumulated on the soil grains. Theoretically, a particle of any diameter may wedge in a crevice site (the void between two tangent soil grains); this interpretation is no longer valid if the ratio of suspended particle diameter to soil grain diameter is small because it can be assumed that the particle lies

on a surface site due to some other mechanisms (e.g., surface forces). Although this process is not important in many filtration problems, it has been reported to be one of the limitations for bacteria traveling through soils (Krone et al., 1958; Gerba et al., 1975). To estimate the significance of this effect, Herzig et al. (1970) gave the following expression for the volume of deposited particles with uniform shape per unit volume of total porous medium based on purely geometric considerations:

$$\sigma = \frac{1}{2}(1 - n_0)\pi Z(d/d_g)^2 [(1 + d/d_g)^2 - 1]^{1/2} \quad (2)$$

where  $n_0$  is the initial porosity;  $d$  and  $d_g$  are suspended particle and grain mean diameters, respectively; and  $Z$  is the coordination number which indicates the interconnectedness in the network of a porous medium. Herzig et al. (1970) have shown that for  $n_0 = 0.40$  and  $Z = 7.0$ , the retention by this mechanism is important if  $d/d_g \geq 0.05$ . For bacteria with  $d = 1 \mu\text{m}$  and silt with a mean grain diameter  $0.01 \text{ mm}$ , eq. 2 would give  $\sigma = 3.02\%$  which is not a negligible amount. For viruses, the limit could hardly be reached. For polio virus with a mean diameter of  $0.01 \mu\text{m}$  in the same soil, eq. 2 would give  $\sigma = 3 \cdot 10^{-5}\%$  which is practically negligible. Therefore, for bacteria the effect of straining should be included in the formulation, but for viruses this term can be neglected.

## 2.2. Adsorption

The adsorption of viruses and bacteria through soils has been reviewed by Bitton (1975) and Gerba et al. (1975). The small size of the virus and its surface properties indicate that removal is primarily through adsorption to the particles rather than straining and other effects, as with bacteria.

Three types of adsorption are referred to in the literature (Weber, 1972), namely, "physical" adsorption, "chemical" adsorption (chemisorption), and "exchange" adsorption. The term "physical" is applied to adsorption resulting from van der Waals forces. In this kind of adsorption, the adsorbate (adsorbed particles) molecule is not affixed to a specific site at the surface but, rather, is free to undergo translational movement within the interface. Herzig et al. (1970) have shown that for particles of diameter larger than  $0.1 \mu\text{m}$  the molecular attraction energy of van der Waals forces overcomes the random movement energy of Brownian motion, but for smaller particles it is the contrary. Although these forces act over a range of the order of  $0.01 \mu\text{m}$ , Machrle and Machrle (1961), Spielman and Goren (1970) have built filtration models by assuming that particles are attracted towards grains by van der Waals forces and move towards them at the limit velocity given by Stokes law. Fluid flow and other transport processes would bring particles into the range of these forces.

The term "chemisorption" is applied is the adsorbate undergoes chemical interaction with the adsorbent. Chemical adsorption is believed to be unlikely when the porous medium is soil and non-reactive (Weber, 1972):

"Adsorption of the third type falls within the realm of ion-exchange. Exchange adsorption is a process in which ions of one substance concentrate at a surface as a result of electrostatic attraction of charged sites at the surface"

The range of electrokinetic forces is generally greater than that of van der Waals forces but is not considered to exceed  $0.1\ \mu\text{m}$  (Ives and Gregory, 1966). Viruses are amphoterically charged particles, negative at most soil pH-values. They are adsorbed by anionic resins at pH's below and by cationic resins at pH's above their isoelectric points. Negatively charged viruses can be adsorbed by cationic resins if the ionic strength of the medium is adequate to neutralize both the charge on the virus and the resins.

Virus adsorption to soil surfaces is largely governed by electrostatic double-layer interactions and van der Waals forces. The order of magnitude of each force is close. Therefore, it is necessary to combine both surface effects to calculate the total adsorption phenomena.

Several adsorption models have been designed, the applicability of any particular model being dependent upon the following factors: (a) the physical and chemical nature of adsorbate (viruses) and adsorbent (soil); (b) the pH of the solution; (c) the characteristics of the flow; and (d) the degree of saturation. Soil type, ionic strength of soil solution, amount of organic matter and humic substances are all considered in the first category. High salt content in groundwater would increase the adsorption due to double-layer compression. Also, it is usually agreed that fine-textured soils like clay retain more viruses (Drewry and Eliassen, 1968; Gerba et al., 1975; Bitton et al., 1979) and bacteria (Hagedorn, 1981) than do sandy soils. Scheuerman et al. (1979) found that the water-soluble humic substances present in sediments were responsible for the low adsorption of poliovirus. Also, increasing adsorption occurs with the reduction of pH below 8.0 and with the addition of cations, especially the divalent species (Bitton, 1980). Subtle differences among human viruses were observed regarding their adsorption pattern. Echovirus 1 was the least adsorbed among all the enteroviruses considered (Charles, 1979). This adsorption pattern shows the inability of a specific virus to predict the sorptive behavior of all enteric viruses. Filmer and Corey (1966) studied the movement of virus-sized particles (albumin molecules,  $d = 0.015\ \mu\text{m}$ ) in a soil column and reported that significant quantities of albumin were found in the effluent although most were retained. It was concluded that retention mechanism by soil was due to an adsorption mechanism which increased with decreasing soil moisture. Bitton et al. (1979) critically examined the various methods frequently used to assess soils' potential to retain viruses.

The adsorption models describe the equilibrium value of the dispersed phase (concentration) at a particular temperature. A curve of the concentration of adsorbed phase  $S$  as a function of concentration of dispersed phase  $C$  is called an adsorption isotherm. A discussion of various types of adsorption isotherms are given by Helfferich (1962) and Weber (1972).

Various studies (Drewry and Eliassen, 1968; Filmer et al., 1971; Burge

and Enkiri, 1978) have shown that adsorption data of viruses to soils were found to fit a Freundlich isotherm and were not describable by a Langmuir isotherm. But, Cookson (1970) has utilized the Langmuir isotherm in his study for virus removal through packed beds. The Freundlich equation is basically empirical and has the general form:

$$S = k_F C^{1/m} \quad (3)$$

where  $k_F$  and  $m$  are constants, and  $m > 1$ . Data are usually fitted to the logarithmic form of the equation:

$$\log S = \log k_F + m^{-1} \log C \quad (4)$$

which gives a straight line with a slope of  $m^{-1}$  and an intercept equal to the value of  $\log k_F$  for  $C = 1$ . The intercept is roughly an indicator of sorption capacity and the slope,  $m^{-1}$ , of the adsorption intensity. Since the slope of the isotherm is close to 1, one may assume that the percent virus removal at low virus concentration is similar to that obtained at higher virus concentrations. As pointed out by Bitton et al. (1979), this assumption may or may not be a reasonable one.

Filmer et al. (1971) obtained a Freundlich isotherm as:

$$S = 0.0543 C^{0.427} \quad (5)$$

Adsorption is also a factor in the removal of bacteria by soil. Gerba et al. (1975) and Weaver (1981) have noted that adsorption phenomena play an important role in the removal of bacteria in soils that contain clays.

### 2.3. Sedimentation in the pores

Gravitational deposition on grains can occur if the particles have a density different from that of the liquid. Due to their extremely small size, viruses and some bacteria are neutrally buoyant and therefore do not tend to settle. Hence, any term in the conservation of mass equation characterizing the effects of gravitational settling can sometimes be neglected. But, Gerba et al. (1975) reported that the sedimentation could be a mechanism of removal for some bacteria. Then its effect has to be included in formulation. Matthess and Pekdeger (1981) believed that the kinetic energy of a small particle which is transported by the groundwater flow to the surface of a soil grain is not high enough to overcome the repulsive surface forces. Yao et al. (1971) noted that gravitational settlings play a significant part only in the capture of relatively large particles ( $> 5 \mu\text{m}$ ) (Fig. 2); for these particles the removal efficiency is proportional to  $d^2$ . The gravitational velocity as expressed by Yao et al. (1971) can be used as a criterion to measure the significance of sedimentation:

$$v_g = (1 - \rho_w / \rho)(m_d g / 3\pi\mu_w d) \quad (6)$$

where  $\rho$ ,  $d$  and  $m_d$  are the density, diameter and mass of the particles,

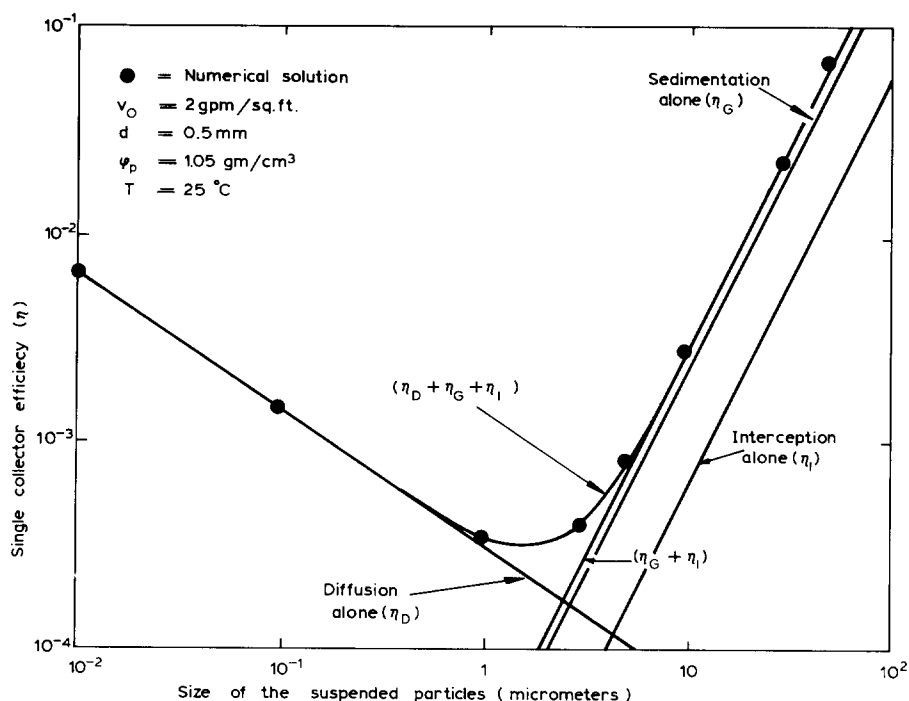


Fig. 2. Various capture mechanisms (after Yao et al., 1971).

respectively; and  $\rho_w$  and  $\mu_w$  are the density and viscosity of the water, respectively. For a bacterium with  $\rho = 1.02$  g cm<sup>-3</sup> and  $d = 5$   $\mu$ m, eq. 6 gives  $v_g \approx 4 \cdot 10^{-5}$  cm s<sup>-1</sup>. This value is comparable to groundwater flow velocity.

#### 2.4. Interception

Even with exactly the same density as the fluid, the suspended particles, owing to their size, would not be able to follow the smallest tortuosities of the streamlines of the fluid and they will thus collide with the walls of the convergent areas of the pores. The interception seen in Fig. 2, is not an effective mechanism because of particle size of the microorganisms considered in this study.

Based on the discussions given above we will give a different rate of deposition expressions for bacteria and viruses.

When suspended bacteria accumulate on the soil surface (or adjacent to the recharge well), the straining effect increases with bacterial accumulation and eventually captured particles behave like a filter and remove finer particles. When the accumulations grow and become unstable, clusters break off which are transported by the flow and may be removed by straining



and sedimentation. When the clusters break off, bacteria saturate the straining sites below, then the saturated front progresses. The rate of removal depends on the flow rate and the size and density of the bacterial clusters (Krone et al., 1958). If the deposition (clogging) of the bacteria by various mechanisms (straining, sedimentation, adsorption) and sloughing off clusters (declogging) are simultaneous, the conservation equation for the deposited material may be written as:

$$\partial \rho \sigma / \partial t = R_a + R_{d_s} + R_{g_s} \quad (7)$$

where  $R_{d_s}$  and  $R_{g_s}$  are the growth and decay terms, respectively, in the deposited state. These terms are discussed in Sections 2.7 and 2.8.  $\sigma$  is the volume of deposited bacteria per unit volume of porous medium. The term  $R_a$  can be expressed by a kinetic equation:

$$R_a = k_c(n - \sigma)C - k_y \rho \sigma^h \quad (7a)$$

Then, eq. 7 would yield to:

$$\partial \rho \sigma / \partial t = k_c(n - \sigma)C - k_y \rho \sigma^h + R_{d_s} + R_{g_s} \quad (7b)$$

where  $k_c$  and  $k_y$  are the clogging and declogging rate constants, respectively; and  $h$  is a constant. When  $h = 1$ , eq. 7 reduces to the kinetic equation proposed by several researchers (e.g., Mints, 1951; see Sakthivadivel and Irmay, 1966 for a review). The power term  $h$  which has to be determined experimentally scales the importance of  $C$  on  $\sigma$ . For the values of  $h$  greater than 1, the concentration has a decreasing effect on the amount of deposited material.

The clogging rate constant,  $k_c$ , is a function of flow rate, porosity and clogged pore volume. The declogging rate constant,  $k_d$ , is a function of concentration,  $C$ , and probably flow rate. Although eq. 7 or its counterparts in the literature has a single form, their application to the real problem is not straightforward. Especially, the problem of spontaneous declogging has created considerable debate among researchers working on filtration. Herzig et al. (1970) have reviewed these views. The widely accepted conclusion is that declogging is normally improbable at the beginning of filtration, but when the filter is nearly saturated with deposited material and the flow rate is considerably increased, declogging may occur.

In case of viruses, since the adsorption is the major removal mechanism, eq. 7 should be replaced by:

$$\partial \rho S / \partial t = R_a + R_{d_s} \quad (8)$$

$S$  is related to  $C$  by eq. 3. Note that  $R_{g_s} = 0$  for viruses as will be explained later.

In summary,  $R_a$  is a rate-controlled reaction for bacteria (eq. 7a), and an equilibrium controlled reaction due to surface and electrokinetic forces for viruses (eq. 3).

## 2.5. Diffusion by Brownian motion

Like other colloids, bacteria and viruses also rely on Brownian motion partially for their movement. Brownian motion is a random motion caused by the thermal motion of molecules following their collision with other molecules or with colloids. In Brownian motion, a particle moves in response to the instantaneous difference in impact made by bombarding molecules on different sides of the particle. This difference can be interpreted as a pressure gradient only when averaged over many particles or over a long time interval. The path of the individual particle appears quite erratic, while the average particle flux is proportional to the gradient in pressure. The pressure gradient, in turn, is proportional to the gradient in concentration of the fluid in which the particle is immersed. Then, the mass discharge of bacteria or viruses by Brownian motion is expressed by:

$$J_B = -D_B \theta \nabla C \quad (9)$$

where  $D_B$  is the diffusion coefficient of the suspended particles (bacteria or viruses), which could be estimated by the Stokes—Einstein equation:

$$D_B = k_b T / 3\pi \mu_w d \quad (10)$$

where  $k_b$  is the Boltzmann constant (energy per degree);  $T$  is the absolute temperature;  $\mu_w$  is the groundwater's viscosity; and  $d$  is the diameter of the suspended particles. The ratio of  $D_B$  to the average grain diameter  $d_g$  would give the mean velocity imparted by Brownian motion over a distance of  $d_g$ . Then  $d_g v_f / D_B$  would give the Péclet number of the diffusion process by Brownian motion in a porous medium flow. Ives (1975) reported that in water filtration this dimensionless ratio changes from  $2 \cdot 10^5$  to  $10^8$ . Although Brownian particle diffusivities are generally smaller than molecular diffusivities, they represent the diffusion characteristic for submicron particles like bacteria and viruses. Smaller particles are collected more efficiently due to their greater Brownian motion. Yao et al. (1971) have shown that for suspended particles smaller than  $1 \mu\text{m}$ , removal efficiency increases with decreasing particle size which is accomplished by Brownian diffusion. Many bacteria ( $7\text{--}0.2 \mu\text{m}$ ) and viruses ( $0.5\text{--}0.01 \mu\text{m}$ ) in groundwater are within this range.

## 2.6. Systematic (chemotaxis) and random (tumbling) motion of bacteria

The existence of concentration gradients in natural systems is commonplace. We consider for example the concentration profile of solutes in waters seeping through soil. Some microbes move systematically toward a richer food supply, and this motion, induced by presence of a solute gradient, is termed chemotaxis. In other words, chemotaxis may be defined as directed movement of a cell toward an attractant. The organism detects and responds to a difference in substrate concentration on a gradient of the stimulant. Their flagella propel them through the medium and normally they swim

smoothly, then tumble. Tumbling is uncoordinated, chaotic motion which randomly reorients the bacterium for the next swim (Berg and Brown, 1972). Swimming occurs for some types of bacteria (e.g., *Escherichia coli*) when their flagella rotate counterclockwise, and tumbling occurs when several of the flagella start to rotate clockwise and thus disrupt the swimming. Chemotaxis occurs when bacteria headed toward higher concentrations of attractant swim longer than normal and, to a lesser extent, when bacteria headed toward lower concentrations tumble sooner than normal (Berg and Brown, 1972). Thus, an otherwise "random walk" is biased so that bacteria "gravitate" toward higher attractant concentrations.

Bacteria swim longer if heading toward high attractant concentrations than in an isotropic (no gradient) medium because they have specific chemoreceptors, equipped with active sites like enzymes, that bind the attractant (Adler, 1969). As the bacteria encounter increasing concentrations of attractant, more of the chemoreceptors become titrated with attractant. Somehow, increasing the number of chemoreceptors titrated with attractant signals the flagella to keep swimming and postpone tumbling.

Bacteria appear to depend upon this temporal mechanism, since the difference in concentration of attractant over the diameter of the cell is probably too small to be sensed by a spatial mechanism. Macnab and Koshland (1972) have shown that bacteria sense gradients by a "temporal" mechanism. They appear to "remember" the concentration of attractant to which they were previously exposed and to "compare" it with their current environment. As a result of this comparison, if the latter is lower, the cells increase their rate of tumbling until adaptation (normal frequency of swimming and tumbling) occurs as indicated by reduced tumbling. If it is higher, the cells show more smooth swimming and less frequent tumbling until adaptation results with increased tumbling.

The attractants (substrate) for bacteria are simple molecules such as sugars (glucose, galactose, ribose) and amino acids (serine, aspartic acid). These organic matters are present in sewagewater as carbohydrates and proteins (Metcalf & Eddy, Inc., 1972). In general, wastewater seeping or being injected into groundwater contains nutrients required for proper cell growth.

Dahlquist et al. (1972) studied the chemotactic response of *Salmonella typhimurium* in time-dependent concentration difference of serine. The superposition of a linear gradient of serine which runs from zero to  $10^{-3}$  M on an initially uniform distribution of the bacteria shows that the bacteria accumulate at the high serine concentration region, but a broad peak is formed in the middle of the serine gradient. According to Dahlquist et al. (1972), this can only be explained if the velocity of the bacteria moving up the gradient depends on the absolute concentration  $C_F$  as well as the rate of change of concentration. The gradient,  $dC_F/dx$ , is constant throughout, so the bacteria cannot be responding solely to the absolute gradient of serine. Following Dahlquist et al. (1972):

"An explanation of the complicated response to linear gradients is that the bacteria actually respond to proportional changes in concentration, that is,  $dC_F/C_F$ . In this case, an exponential gradient:

$$\frac{dC_F/C_F}{dx} = \frac{d \ln C_F}{dx} = \text{constant}$$

should elicit a constant bacterial response throughout its length. The result of superposing such a distribution of serine on an initially uniform bacterial distribution shows that the bacteria accumulate at the top of the gradient as a well defined peak. The bacterial concentration on either side of this peak remains fairly constant, however, unlike the response to a steep increase which shows a distinct trough adjacent to the peak. Thus, bacteria are moving through the gradient region at a steady rate. This in turn means that the average velocity,  $v_m$ , of the bacteria is determined by the proportional changes in concentrations,"

Therefore, one can write:

$$v_m = k_m \nabla \ln C_F = (k_m / C_F) \nabla C_F \quad (11)$$

where  $k_m$  is called the migration rate constant or chemotactic coefficient. It is a weakly varying function of  $C_F$  (Nossal and Weiss, 1973). For the problem we are considering, it may be safely assumed as a constant.

The chaotic, random movement of motile bacteria which was referred to as "tumbling" above gives rise to an effective diffusivity or motility coefficient  $D_T$ . This random movement may be assumed to be superimposed upon any systematic migration induced by substrate, so the two effects (random and systematic) may be considered to be additive. Although  $k_m$  may be positive or negative,  $D_T$  is always positive. These two parameters can be determined by laboratory experiments.

In chemotaxis the motion of a cell is influenced by the molecules of the substrate through a chemical interaction rather than by the direct impact characteristic of Brownian motion. The bacteria respond to fluctuations in estimates made of the concentration of the substrate, rather than to the average concentration. This assumption seems necessary when one considers the extremely small dimensions that a chemical receptor of a microscopic organism must have. The number of molecules in the vicinity of the receptor at any given time would inevitably exhibit a great deal of fluctuation, although fluctuations would be diminished if the receptor effectively averages its readings on concentration over some time interval.

If the chemotactic movement is initiated by the local concentration in the vicinity of a receptor, then the path of the individual cell would reflect the fluctuation characteristics of the samples assayed by the receptors. Keller and Segel (1971) found that individual cell paths can result in an average cell flux which is proportional to the macroscopic substrate gradient. Then, the total flux due to chemotactic movement and tumbling can be expressed by:

$$J_{CT} = \theta(v_m \cdot C - D_T \cdot \nabla C) \quad (12)$$

or by inserting eq. 11:

$$J_{CT} = \theta(Ck_m \nabla \ln C_F - D_T \cdot \nabla C) \quad (13)$$

The anatomy of viruses is very much different than that of bacteria. Viruses are colloidal infectious particles which are composed of a protein coat (capsid) containing a nucleic acid core (Bitton, 1980). Viruses are known to parasitize animal cells, bacteria, plant cells, blue-green algae, and fungi. According to the infected host-cell, they are classified as animal viruses, bacterial phages, blue-green algal viruses and fungal viruses. Human enteric viruses which are our concern in this study fall into the first category.

Viruses reproduce only inside an appropriate host-cell. Once in the host-free state, they are exposed to a variety of adverse environmental factors. Therefore human enteric viruses do not usually have host-cells for reproduction in a groundwater environment and they are inactivated. Therefore, the formulation given by eqs. 11–13 for bacterial chemotaxis is irrelevant for viruses.

## 2.7. Die-off

Death of microorganisms plays an important role during the larger retention periods and/or especially unsaturated flow conditions. Survival times for bacteria in soil normally range from a few weeks to a few months, and sometimes longer. The factors affecting survival of enteric bacteria in soil have been reviewed by Gerba et al. (1975). They concluded that, in most cases, 2–3 months is sufficient for reduction of pathogenic bacteria to negligible numbers once they have been applied to the soil, although survival times as long as 5 years have been reported.

Beard (1940) stated that soil moisture was the most important determining factor in the survival of *Salmonella typhosa*. Their survival in all types of soil tested was found to be greatest during the rainy season. Beard (1940) has also found that the death was very rapid in peat soil with a pH between 3 and 4. Warm temperature is an adverse factor in the viability of bacteria. Similar conclusions were reached for the bacterial survival in groundwater too (Gerba et al., 1975). Kudryavtseva (1972) reported that bacteria survived in groundwater for 3 months after they had been introduced. In the laboratory, pathogenic and saprophytic strains of *E. coli* in groundwater samples kept in total darkness survived 4 and 5½ months, respectively.

Gerba and Schaiberger (1975) have shown that viruses adsorbed to kaolinite clay survived longer in seawater than unadsorbed virus. Sobsey (1981) noted that poliovirus and reovirus in suspensions of eight different soil materials did not always survive longer than in soil-free control. Schaub et al. (1974) also reported that viruses in adsorbed state were infectious as much as free entities. Gerba et al. (1975) concluded that the survival of

the enteroviruses in soil is dependent on the nature of the soil, temperature, pH and moisture.

The death of microorganisms is a first-order irreversible reaction, hence it can be expressed by:

$$R_{df} = -k_d \theta C; \quad R_{ds} = -k_d \rho \sigma \quad \text{for bacteria} \quad (14)$$

or

$$R_d = R_{df} + R_{ds} = -k_d(\theta C + \rho \sigma) \quad (14a)$$

and, similarly:

$$R_d = -k_d(\theta C + \rho S) \quad \text{for viruses} \quad (15)$$

where  $k_d$  is the specific decay rate, which is a constant; and  $R_{df}$  is the decay term in free state in water. Based on the discussions above, we assume that the decay rate is the same in free and adsorbed states. There is considerable variation in the values of  $k_d$  in the literature. The value of  $k_d$  for heterogeneous microbial populations in municipal sewage varies from 0.025 to 0.098 hr.<sup>-1</sup> (Gaudy and Gaudy, 1980). The decay mechanisms of viruses is similar to that of bacteria, but certain types which are more resistant to environmental changes might survive longer (1–6 months) than their bacterial counterpart. Matthess and Pekdeger (1981) considered only the decay in the free state in their study.

## 2.8. Growth of bacteria

In various studies, bacterial growth has been reported. Matthess and Pekdeger (1981) noted that parasitic bacteria can show slight increase in the germ number during the first 1–7 days after entering the groundwater due to contamination. *E. coli* multiply on organic matter filtered out from lake water (Goldshmid, 1974). Similar results were also previously reported by Rebhun and Schwarz (1968). Rittmann et al. (1980) presented a biofilm model considering the kinetics of substrate (nutrient) utilization and growth of biofilm. Their computer simulations based on reasonable assumptions of kinetic constants lead to the conclusion that biofilm growth takes place for the most part within 1 m of the recharge injection well, even when injected water is a high-quality advanced treatment effluent.

The growth of bacteria is assumed to follow Monod's equation. Monod's equation describes a relationship between the concentration of a limiting nutrient and the growth rate of microorganisms. As stated earlier, nutrients proper needed for biological growth can be present in a sewagewater. The restrictions of Monod's equations are discussed in various microbiology textbooks (e.g., Gaudy and Gaudy, 1980). Since the bacterial growth in a groundwater environment is slow, Monod's equation may safely be used. Similar to the decay process, we assume that bacteria can grow in deposited state as well as in the suspension in the same rate. Then, a generalized Monod's equation can be written as:

$$R_{gf} = \mu\theta C \quad \text{and} \quad R_{gs} = \mu\rho\sigma \quad (16)$$

or

$$R_g = \mu(\theta C + \rho\sigma) \quad (16a)$$

where  $\mu$  is the specific growth rate; and  $R_{gf}$  and  $R_{gs}$  denote the growth terms in free and adsorbed states, respectively. The functional relationship between  $\mu$  and an essential nutrient's concentration  $C_F$  was proposed by Monod (1942). Of the same form as the Langmuir adsorption isotherm and the standard rate equation for enzyme-catalyzed reactions with a single substrate, it states that:

$$\mu = \mu_{\max} C_F / (K_i + C_F) \quad (17)$$

where  $\mu_{\max}$  is the maximum growth rate achievable when  $C_F \gg K_i$  and the concentration of all other essential nutrients is unchanged.  $K_i$  is that value of the concentration of the substrate where the specific growth rate has half its maximum value; roughly speaking, it is the division between the lower concentration range where  $\mu$  is linearly dependent on  $C_F$ , and the higher range, where  $\mu$  becomes independent of  $C_F$ .  $K_i$  for *E. coli* strains growing in tryptophan media is  $1.1 \cdot 10^{-3} \text{ g ml}^{-1}$  (Stainer et al., 1970). Gaudy et al. (1971) reported the value of  $K_i$  and  $\mu_{\max}$  for a heterogeneous microbial population of sewage origin growing on glucose as  $0.1 \cdot 10^{-3} \text{ g ml}^{-1}$  and  $0.38 \text{ hr}^{-1}$ , respectively. Gaudy and Gaudy (1980) noted that the smaller the value of  $K_i$ , the more closely Monod's equation (13) will be able to reproduce the curve of growth.

Rittmann et al. (1980) introduced the concept of secondary utilization which states that a secondary substrate present below a minimum concentration level is utilized by a biofilm that is supported by one or more primary substrates that are present above that minimum level. They observed that during groundwater recharge of advanced treatment effluent chemical oxygen demand (COD) or total organic carbon (TOC) serves as the primary substrate. However, the land application by primary treatment effluent or the seepage of raw sewagewater from septic tanks can provide enough substrate concentration to support the microbial growth in soil and groundwater.

### 3. SUMMARY OF MECHANISMS FOR BACTERIAL CONTAMINATION

The governing equations can be summarized by combining eq. 1 with eqs. 7, 14 and 16 to yield:

$$\partial(\rho\sigma)/\partial t + \partial[(n - \sigma)C]/\partial t = -\nabla \cdot J + [(n - \sigma)C + \rho\sigma][\mu - k_d] \quad (18)$$

where  $\theta = n - \sigma$ . The flux term,  $J$ , can also be expressed as the sum of mechanisms discussed earlier:

$$J = -D_d(n - \sigma)\nabla C + (n - \sigma)v_f C + (n - \sigma)v_g C + J_B + J_{CT} \quad (19)$$

where  $D_d$  and  $v_f$  are the coefficient of convective dispersion and the flow velocity, respectively. The term  $v_f C$  expresses the transport by convection. The transport by dispersion is given by  $D_d \nabla C$ . Inserting eqs. 9 and 13 into eq. 19 would give:

$$\begin{aligned} \partial(\rho\sigma)/\partial t + \partial[(n - \sigma)C]/\partial t = \\ - \nabla \cdot [-D(n - \sigma)\nabla C + (n - \sigma)\{v_f + k_m \nabla \ln C_F \\ + (1 - \rho_w/\rho)(m_d g/3\pi\mu_w d)\}C] + [(n - \sigma)C + \rho\sigma](\mu - k_d) \end{aligned} \quad (20)$$

The coefficient of hydrodynamic dispersion,  $D$ , is defined as the sum of the coefficients of mechanical (convective) dispersion,  $D_d$ , of Brownian diffusion,  $D_B$ , and of effective diffusivity,  $D_T$ , due to tumbling of bacteria. Eq. 20 would couple with eqs. 7 and 7a, and the mass-conservation equation for substrate  $C_F$  for a complete solution.

The balance equation for an adsorbing solute (substrate) flowing through a porous medium is expressed as:

$$\begin{aligned} \partial(\rho_s S_F)/\partial t + \partial[(n - \sigma)C_F]/\partial t = \\ - \nabla \cdot [-(D'_d + D_m)(n - \sigma)\nabla C_F + (n - \sigma)v_f C_f] + R_F \end{aligned} \quad (21)$$

where  $C_F$  is the mass of the substrate per unit volume;  $S_F$  denotes the mass of adsorbed substrate per unit mass of the solid part of the porous medium.  $D_m$  and  $D'_d$  are the coefficients of molecular diffusion and mechanical dispersion, respectively; and  $\rho_s$  is the bulk mass density of dry soil.

The adsorption term ( $\partial\rho_s S_F/\partial t$ ) for the substrate is related to its concentration,  $C_F$ , in the liquid phase by an adsorption isotherm.

The rate of substrate consumption due to bacterial growth (in deposited and free states),  $R_F$ , can be written as:

$$R_F = -\mu Y^{-1}[(n - \sigma)C + \rho\sigma] \quad (22)$$

where  $Y$  is the true or maximum cell yield which is the mass of cells produced per unit mass of substrate removed. For heterogeneous populations of wastewater origin grown on various carbon sources,  $Y$  ranges from 0.29 to 0.68. A value of  $\sim 0.5$  would be a fairly good average (Gaudy and Gaudy, 1980). Sykes et al. (1982) has determined  $Y$  as 0.04 for a landfill study.

Eqs. 7a, 20 and 21 would contribute the set of governing equations for bacterial transport in a porous medium. The dependent variables are  $C$ ,  $\sigma$  and  $C_F$ . The equations are highly nonlinear.

The transport equation given by Matthess and Pekdeger (1981) contains some of the terms of eq. 20. The mechanisms considered by them were dispersion, convection, diffusion (Brownian), adsorption, "active mobility" (motility), and decay which was considered only for the free state and neglected for deposited microorganisms. No solution of the equation with realizable coefficients are given. The additional mechanisms to be considered for bacteria are deposition, sedimentation, chemotaxis, and growth processes



are given in eq. 20. The growth and chemotaxis processes are coupled with a transport equation (21) for bacterial nutrients. Also a first-order kinetic deposition equation (7a) is incorporated to the governing equation (20).

Hagedorn (1981) has noted the use of a longitudinal dispersion equation to interpret the dispersal of *E. coli* cells through a porous medium by McCoy and Hagedorn (1980). The equation itself is not given in either publication. Sykes et al. (1982) has presented a model to predict the concentrations of leachate organics, measured as chemical oxygen demand, in groundwaters below sanitary landfills. Simultaneous substrate utilization and microbial mass production equations, with convection and dispersion included for the former, are used for modeling of biodegradation. In other words, with these assumptions eqs. 18 and 21 would reduce to:

$$\partial C / \partial t = \mu C - k_d C \quad (23)$$

$$\partial C_F / \partial t = -\nabla \cdot [-D'_d \nabla C_F + v_f C_f] + R_F \quad (24)$$

#### 4. SUMMARY OF MECHANISMS FOR VIRAL CONTAMINATION

In the case of viruses, eq. 1 combined with eqs. 8 and 14a would yield for a saturated porous medium:

$$\partial(\rho S) / \partial t + \partial(nC) / \partial t = -\nabla \cdot J - k_d(nC + \rho S) \quad (23)$$

In the development of eq. 23, the growth terms for viruses ( $R_{g_f} = R_{g_b} = 0$ ) were neglected. We also neglected the volume of deposited viruses by assuming  $\theta \simeq n$ . The flux term  $J$  would contain:

$$J = -D_d n \nabla C + n v_f C + J_B \quad (24)$$

Since the viruses are neutrally buoyant particles, the settling velocity term,  $v_g$ , has vanished. Substitution of eq. 9 into eq. 24 would yield:

$$\partial(\rho S) / \partial t + \partial(nC) / \partial t = -\nabla \cdot [(D_d + D_B) n \nabla C + n v_f C] - k_d(nC + \rho S) \quad (25)$$

The adsorption term,  $\partial(\rho S) / \partial t$ , can be evaluated by employing the adsorption isotherm given by eq. 3.

#### 5. SUMMARY AND CONCLUSIONS

We introduced the governing mechanisms for transport and fate of microbial particles in a water-saturated porous medium. So far we have been discussing the case of a single liquid saturating the porous medium. In principle, the methodology presented is applicable also to unsaturated flow (e.g., the zone above the water table); however, certain modifications have to be introduced. The mass conservation should be treated separately for air and water. This means one mass-conservation equation will be

developed for each fluid. Each fluid will have its own pressure and saturation. This, in turn, might create different flow velocities for air and water. The effect of degree of saturation on deposition or adsorption also has to be considered. As observed by various researchers (e.g., Filmer and Corey, 1966), the volume of captured particles increases with decreasing degree of saturation due to the fact that in case of unsaturated pores the distance traveled by the particles is shorter than that of saturated pores.

In the derivation of governing equations, we have started from the three-dimensional macroscopic conservation of mass equation for particles. The mass-dispersive and mass-diffusion fluxes have been neglected. In addition to the transport by dispersion, convection and Brownian motion, we included the transport due to the systematic (chemotaxis) and random (tumbling) motion of bacteria. The decay and growth of the microorganisms has been included as source and sink terms in the mass-conservation equation. The differences between bacteria and viruses with respect to their transport mechanisms, decay and growth kinetics due to their anatomies and sizes have been considered and treated separately. The rate of deposition terms have been incorporated by a rate (kinetic) equation and an adsorption isotherm for bacteria and viruses, respectively. The values of the various coefficients and parameters appearing in the equations have to be determined basically by laboratory experiments or sometimes in the field.

In this study we did not consider the effect of water potential on microbial growth and activity. The primary response of microbial cells to water potential stress is dominated by the biophysical need of the cells to achieve thermodynamic water potential equilibrium with their environment. This topic was studied extensively by Harris (1981). For a detailed discussion, the reader is referred to that publication.

The methodology developed in this study can be employed to study the microbial soil and groundwater contamination due to septic-tank seepage or land application of municipal wastewater or recharge of groundwater aquifers by treated sewagewater. In other words, it can be used as a practical tool in the long-term evaluation of the risk of accumulation of pollutants in top soils of agricultural importance and of contamination of groundwater used for drinking water supply. Results also can be applied to determine the suitability of septic-tank locations and planning of future sanitary landfills and cesspools. The solutions of the governing equations will be presented in a separate paper.

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