ground water

# The Effect of Critical pH on Virus Fate and Transport in Saturated Porous Medium

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## Abstract

Several viral transport experiments were conducted in a model aquifer 1 m long, using bacteriophages MS2 and ϕX174 at various pH (4.6 to 8.3) conditions, to increase our understanding of virus behavior in ground water. The results indicate the existence of a critical pH at which the virus behavior changes abruptly. This is supported by data from field and batch experiments. The critical pH is determined to be 0.5 unit below the highest isoelectric point of the virus and porous medium. When water pH is below the critical pH, the virus has an opposite charge to at least one component of the porous medium, and is almost completely and irreversibly removed from the water. This suggests that electrostatic attraction at a subcritical water pH condition is an important factor controlling virus attenuation in ground water. The concept of critical pH can assist in the design of geologic barriers for preventing viral contamination in ground water.


## Introduction

In the last two decades, a number of studies have been done with batch, flowing column, and field experiments on viral behavior in the subsurface. Several processes may affect viral fate and transport in ground water, irreversible attachment, reversible attachment, and inactivation. Reversible attachment is defined as a process where the pre-attached virus re-enters the water in the time period of interest. Irreversible attachment is defined as the case where the attached viruses do not enter the water in the time period of interest. Only irreversible attachment can result in permanent removal of viruses from water. The viral behavior in ground water appears to be controlled by the properties of viruses (Dowd et al. 1998; Deborde et al. 1999; Schijven et al. 2001; Woessner et al. 2001), the properties of the porous medium (Loveland et al. 1996; Pieper et al. 1997; Ryan et al. 1999), and the properties of water transporting the virus (Loveland et al. 1996; Bales et al. 1993, 1997; Redman et al. 1999). An incomplete understanding of the processes governing virus fate and transport is achieved if the study does not consider all controlling factors from the three aspects listed. The electrostatic attraction and repulsion, van der Waals forces, and hydrophobic effects are three major forces responsible for interaction between the virus and the porous medium (Jin et al. 2000). In particular, viral attachment was observed to be a function of water pH (Bales et al. 1993, 1997), the isoelectric point ($pH_{iep}$) of the porous medium (Loveland et al. 1996), and the isoelectric point of the virus (Deborde et al. 1999; Dowd et al. 1998; Woessner et al. 2001). This suggests that electrostatic interaction is an important factor controlling virus attachment and detachment. Loveland (1996) reported a pH edge for electrostatic interaction between PRD1 and porous medium, which is 2.5 to 3.5 pH units above the $pH_{iep}$ of the mineral surface. PRD1 attachment is nearly complete below this pH, while minimal above that pH. More recently, Schijven et al. (2001) reported that bacteriophage PM2 with an isoelectric point of 7.3 shows different adsorption characteristics from four other viruses (MS2, Qβ, PRD1, and ϕX174) with lower isoelectric points. These

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results indicate that there are probably water pH ranges, depending on isoelectric points of the virus and the porous medium, between which the virus behavior in ground water changes abruptly. To verify this hypothesis, two bacteriophages with different isoelectric points were studied in a model aquifer. The pH was varied and monitored between experiments to evaluate the effects of pH on virus fate and transport in ground water.

## Materials and Methods

### Model Aquifer

A 109 cm × 40 cm × 2 cm model aquifer was constructed and filled with sand (Figure 1). Ninety percent of the sand grains were 0.5 to 1.0 mm in diameter with the remaining 10% less than 0.5 mm in diameter. One injection port (W0) and five major sampling ports (W1, 2, 3, 4, and 5) were installed in the model, all at the same depth. The distance between major ports was 18 cm. Four additional monitoring ports were installed beside the major ports, but ~6 cm above (W2a and 4a) or below (W3b and 5b) them. The sand consisted of 71% quartz, 9% feldspar, and 20% igneous rock fragments, and was free of iron oxide coating under optical microscope observations. The isoelectric point of quartz is estimated to be 2.9 to 3.0, and that of feldspar 5.2 to 6.1 (Sverjensky and Sahai 1996). The igneous rock fragment component had an elemental composition similar to feldspar when analyzed under the electron microscope. Its isoelectric point is estimated to be similar to that for feldspar. Thus, the sand has two major components in terms of the isoelectric point, one (quartz hereafter) at pH 3 and the other (feldspar hereafter) at pH 5.5. The porosity of the sediment was ~0.45, and the hydraulic conductivity 1.3 × $10^{-3}$ m/sec. The hydraulic gradient was controlled by manipulation of the water level in the inlet reservoir and outlet reservoir.

### Experiments

The model was flushed with deionized water (MiliQ) until the background ionic concentration sampled from injection and sampling ports was constant and below 0.2 mM. Two bacteriophages were selected for the study, MS2 and ϕX174. The two phages have similar sizes (24 and 27 nm, respectively), but significantly different isoelectric points (3.9 and 6.6, respectively), which allows possible effects of the viral isoelectric point to be observed at various pH conditions. Sodium bromide (between 5 and 12 mM)

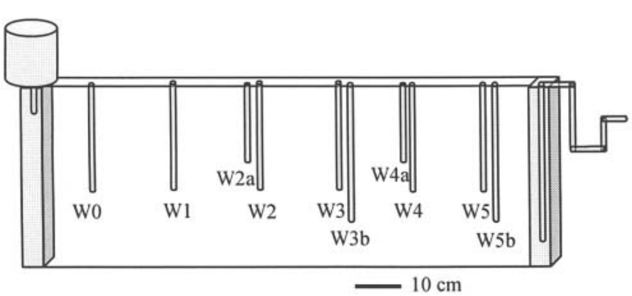


**Figure 1. Schematic map of the model aquifer (109 cm × 40 cm × 2 cm).**

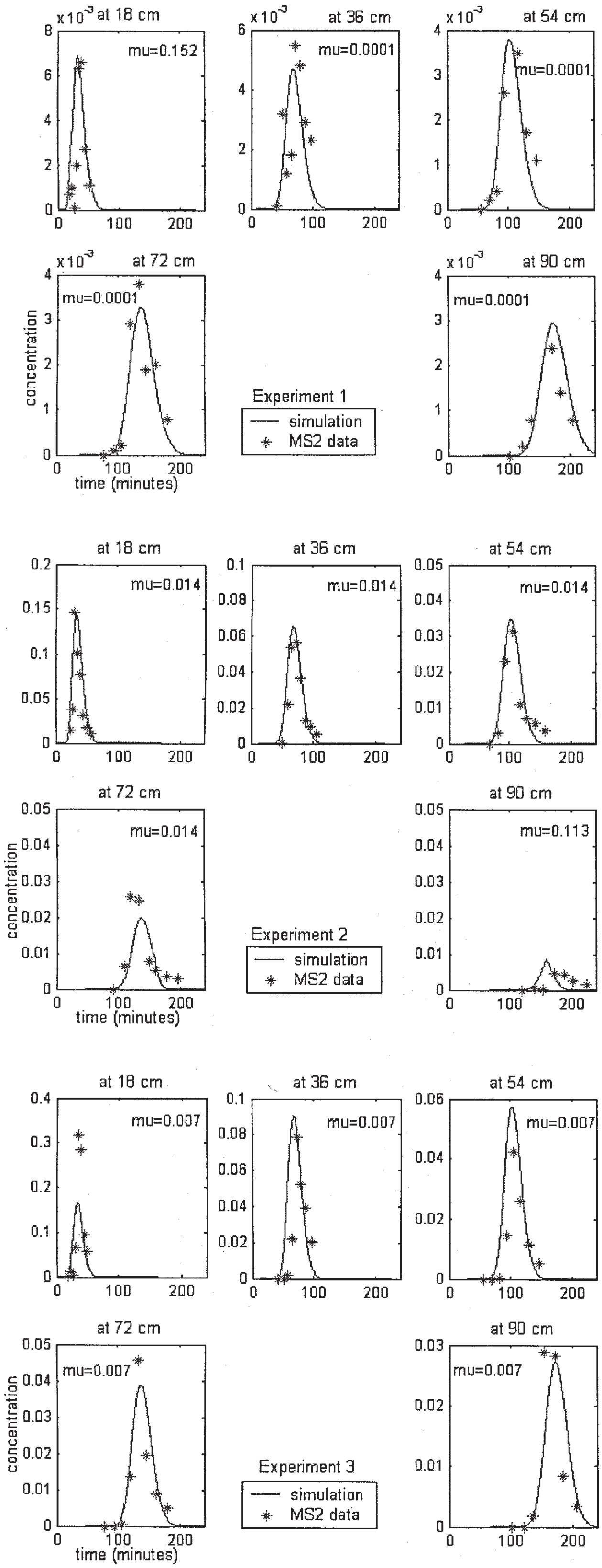


**Figure 2. Results of numerical simulations for MS2 breakthroughs of three experiments. Five panels in each experiment show MS2 breakthroughs at five major sampling ports, with horizontal axis of time and vertical axis of virus relative concentration ($C/C_0$). μ (mu) is the fitted removal rate coefficient (1/min).**

was used as a conservative tracer, and injected together with the virus suspension. After injection, the samples were collected from each of the sampling ports as well as monitoring ports. All samples were stored in a refrigerator at 4°C after the experiment. To avoid introducing complex ions, hydrochloric acid and sodium hydroxide were used to manipulate the water pH, which was measured on site immediately with an Orion Benchtop pH/ISE meter (model 290A). The concentration of bromide was measured using capillary electrophoresis with a detection limit of 0.5 mg/L. The viruses were assayed by the plaque-forming-unit (PFU) method described originally by Adams (1959), usually within two or three days. The analysis error was ±18% at a 95% confidence level. Two control samples of the source were obtained at the injection to observe the inactivation of the phages during the experiment (four hours). One was placed on site to experience similar temperature conditions to that of the water samples (room-temperature source). The other was stored in a refrigerator (4°C) immediately after injection (refrigerated source).

## Data Analysis and Numerical Model

### Problem of Viral Inactivation

Because only active phages were measured in the water samples, it is important to exclude the effect of phage inactivation on the data analysis when evaluating interactions of viruses with the porous medium under various pH conditions. It is reported that temperature is the major factor controlling viral inactivation in water (Yates and Gerba 1984; Yahya et al. 1993). Thus, in the reported experiments the measured phage concentration in the water samples was normalized to that of the room-temperature source. This procedure was thought to remove most of the viral inactivation effect because the phages in the room-temperature source experienced similar temperatures to those in the water samples. The normalized phage concentrations are dimensionless and comparable between experimental runs. The bromide concentration was normalized in the same way. Additionally, the room-temperature source and the refrigerated source were compared to make sure that the viral inactivation was not a dominant factor during the experiments.

### Relative Breakthrough and Collision Efficiency

Two key parameters of colloid filtration theory, relative breakthrough (RB) and collision efficiency (α), were used to analyze the viral breakthroughs. Relative breakthrough is a comparison of the time-integrated mass of bacteriophages relative to that of bromide (Harvey and Garabedian 1991), and is given by

$$RB(\%) = \left[ \frac{\int_{t_0}^{t_f} \frac{C_t}{C_0}\, dt}{\int_{t_0}^{t_f} \frac{[tracer]_t}{[tracer]_0}\, dt} \right] \times 100 \tag{1}$$

where $C_0$ and $[\text{tracer}]_0$ are the phages and bromide normalized concentrations in the injection source, respectively; $C_t$ and $[\text{tracer}]_t$ are the normalized concentrations of phages and bromide at time $t$; and $t_0$ and $t_f$ are the times of the beginning and end of the breakthrough. The RB data are comparable between sampling ports if it is assumed that the phage did not deviate from the bromide plume, and both phages and bromide exhibited similar dispersion characteristics at a distance less than 1 m. A derivative parameter, relative attenuation (100 – RB)%, was used in order to easily compare to the collision efficiency.

Collision efficiency relatively describes the probability of the efficient collisions of the suspended viruses with the stationary grains resulting in removal (or attachment), and is given by

$$\alpha = \frac{d\,\{[1-2(\alpha_l/x)\ln(RB)]^2 - 1\}}{6(1-\theta)\eta\alpha_l} \tag{2}$$

where $d$ is the average collector grain diameter (m), $\alpha_l$ is the longitudinal dispersivity (m), $x$ is the transport distance (m), $\theta$ is the porosity (dimensionless), and $\eta$ is the single collector efficiency (dimensionless) (Harvey and Garabedian 1991).

### Numerical Model

Under the assumption that the virus transport is advection dominated and that dispersion in the vertical

**Table 1**
**Summary of the Experimental Conditions**

| | Model Aquifer | | | Injection Source | | | | |
|---|---|---|---|---|---|---|---|---|
| **Experiments** | **Velocity m/s** | **pH[1]** | **DO[2] mg/L** | **Ionic Conc.[3] mM** | **Bromide mg/L** | **MS2 PFU/mL** | **φX174 PFU/mL** | **Volume mL** |
| 1 | 1.1E–4 | 6.1 | 5.0 | 0.15 | 973.2 | 3.12E+6 | none | 15 |
| 2 | 9.7E–5 | 6.0 | 4.3 | 0.12 | 436.2 | 1.95E+5 | 2.07E+5 | 15 |
| 3 | 9.7E–5 | 7.5 | 4.4 | 0.12 | 584.7 | 3.67E+5 | 6.25E+4 | 15 |
| 4 | 9.6E–5 | 7.9 | NA | 0.17 | 573.8 | 2.09E+5 | 2.08E+5 | 15 |

[1]pH is the average value during the experiment.
[2]DO is dissolved oxygen in water.
[3]Ionic concentration measured in the molar sum of major anions

**Table 2**
**Details of the Water pH for the Experiments**

| Ports \ Experiment | Distance to Injection | 1 b.g. | 1 pl. | 1 end | 2 b.g. | 2 pl. | 2 end | 3 b.g. | 3 pl. | 3 end | 4 b.g. | 4 pl. | 4 end |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| W0 | 0 cm | 5.6 | NA | NA | 5.6 | NA | NA | 6.8 | NA | 7.0 | 7.7 | NA | NA |
| W1 | 18 | 5.3 | 4.6 | 6.3 | 6.2 | 6.0 | 5.9 | 6.9 | 6.1 | 7.1 | 7.8 | 7.4 | 7.0 |
| W2 | 36 | 6.3 | 6.0 | 6.6 | 6.5 | 6.2 | 6.5 | 7.7 | 7.2 | 7.5 | 8.2 | 7.9 | 8.2 |
| W3 | 54 | 6.8 | 6.5 | 6.9 | 6.4 | 6.3 | 6.7 | 8.1 | 7.7 | 8.0 | 8.3 | 8.3 | 8.1 |
| W4 | 72 | 7.1 | 6.7 | 7.1 | 6.4 | 6.3 | 6.5 | 8.1 | 7.7 | 8.3 | 8.4 | 7.6 | 8.1 |
| W5 | 90 | 7.2 | 6.9 | 7.3 | 4.7 | 4.6 | 4.8 | 8.4 | 7.1 | 8.5 | 8.4 | 7.4 | 7.9 |
| Overall pH | | | 6.1 | | | 6.0 | | | 7.5 | | | 6.1 | |

b.g..: background, pl.: plume, end: at the end of the experiment

directions is negligible, the transport process can be modeled by a one-dimensional advection-dispersion equation with a first-order removal term. The advection-dispersion model is written in terms of a dimensionless virus concentration. The irreversible attachment of the virus to the porous medium was modeled through a linear removal term. The reversible equilibrium attachment mechanism was modeled through a proportionality factor to the changing rate of the viral concentration.

The one-dimensional model used here is a slight modification of that used by Jin et al. (2000). The model used here is given by

$$R\frac{\partial C}{\partial t} = D\frac{\partial^2 C}{\partial x^2} - v\frac{\partial C}{\partial x} - \mu C \qquad (3)$$

where $C$ is the dimensionless virus concentration in water, $R$ is the retardation factor of the virus that is related to the reversible attachment process, $D$ is the hydrodynamic dispersion coefficient ($cm^2$/min), $v$ is the steady-state ground water velocity (cm/min), and $\mu$ is the removal rate coefficient (1/min) which is related to the irreversible attachment processes. The initial and boundary conditions for the analytic model are given by

$$C(x,0) = 0, \quad 0 < x < \infty$$
$$\lim_{x\to\infty} C(x,t) = 0$$
$$C(0,t) = 1, \quad 0 \le t < T_c$$
$$C(0,t) = 0, \quad t \ge T_c$$

**Table 3**
**Measured Virus Concentration PFU/mL of Room-Temperature Sources (22º to 23°C) and Refrigerated Sources (4°C)**

| Experimental Runs | | 1 | 2 | 3 | 4 |
|---|---|---|---|---|---|
| MS2 | 22°– 23°C | 3.12E+6 | 1.95E+5 | 3.67E+5 | 2.09E+5 |
| | 4°C | 3.08E+6 | 2.20E+5 | 4.80E+5 | 2.21E+5 |
| ϕX174 | 22°– 23°C | NA | 2.07E+5 | 6.25E+4 | 2.08E+5 |
| | 4°C | NA | 2.24E+5 | 1.24E+5 | NA |

A standard finite difference code using upstream differencing for the advection terms was used to solve the one-dimensional parabolic partial differential equation on a truncated domain, $0 < x < X_\infty < \infty$. The far field boundary condition was approximated using $C(X_\infty,t) = 0$. The truncated boundary, $X_\infty$, was set far enough away from the sampling ports so that its effect on the time-varying solution could be assumed to be negligible for the duration of the simulation. The model simulated a uniform injection of the virus at the left boundary by prescribing to the boundary a unit (dimensionless) concentration for the time period, $0 \le t < T_c$, and zero concentration thereafter.

## Results

A total of four experimental runs were conducted at room temperature (22.5º ± 0.5°C) (Table 1). Detailed pH measurements are shown in Table 2. It should be noted that because no pH buffer was used, pH values tended to vary somewhat during each experiment. This provided a good opportunity for studying virus behaviors under multiple pH conditions. Comparison of room-temperature source and refrigerated source showed that no significant virus inactivation occurred during the experiment except for ϕX174 in experiment 3 (Table 3). Data from the four experimental runs was then plotted and simulated by a one-dimensional model, shown in Figures 2 and 3. The numerical model was calibrated with the bromide breakthrough curves for each experimental run. A constant removal rate coefficient was used to fit the data for experiment 4, which exhibited a small pH fluctuation along the flowpath. Multiple removal rate coefficients were applied if necessary for the experiments with a large pH fluctuation, e.g., experiments 1, 2, and 3. The simulated removal rate coefficient is shown in Figure 4c. Relative breakthrough and collision efficiency were calculated and are plotted in Figures 4a and 4b, where relative breakthrough is presented as relative attenuation for easy comparison. Both relative attenuation and collision efficiency describe the tendency for viral removal from the ground water, and thus are comparable to the removal rate coefficient from numerical simulations. The results from the numerical simulation and filtration theory calculation are in good agreement. It was clearly observed that there is a specific water pH at which the virus behavior changes abruptly (Figure 4). This specific pH is 5 ± 0.5 for MS2, and approximately 6 for ϕX174. At a pH below this value, the viruses were removed from water to a substantially larger degree, compared to a pH above this value.

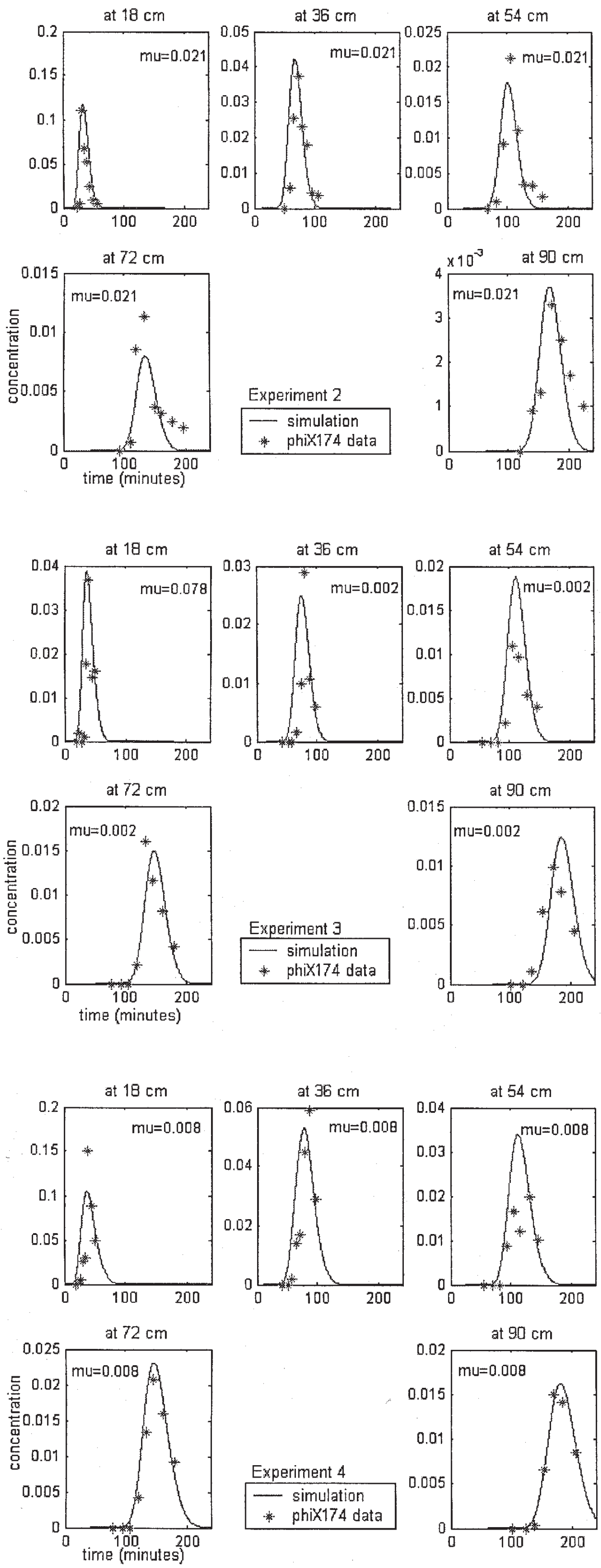


**Figure 3. Results of numerical simulations for ϕX174 breakthroughs of three experiments. Five panels in each experiment show ϕX174 breakthroughs at five major sampling ports, with horizontal axis of time and vertical axis of virus relative concentration ($C/C_0$). μ is the fitted removal rate coefficient (1/min).**

# Discussion

## Electrostatic Interaction and Critical pH

As described earlier, the viral data were normalized to remove the inactivation effect. Thus, the reversible and irreversible attachments are assumed to be the dominant processes controlling virus behaviors shown in our data. Schijven et al. (2001) classified two types of reversible attachment, equilibrium and kinetic. Equilibrium attachment does not distort the symmetric breakthrough, but retards the virus plume relative to the conservative tracer. This is modeled by a retardation factor in our numerical model. The lack of tailing in the virus breakthrough curves (Figures 2 and 3) indicates that reversible attachment due to kinetic processes was not significant in the experiments. To quantify irreversible attachment processes, we use relative attenuation and collision efficiency, which is calculated according to colloid filtration theory, and the removal rate coefficient from the numerical modeling. The results indicate that these parameters depend on water pH (Figure 4). The irreversible attachment processes can be caused by electrostatic attraction, van der Waals force, hydrophobic effect, and possibly straining (Jin et al. 2000; Schulze-Makuch et al. 2003). Among these factors, only electrostatic adsorption is sensitive to the water pH. Besides water pH, ionic concentration in the water may also play a role in electrostatic interaction. Compared to monovalent cations, multivalent cations have much stronger effects on colloid transport (Grolimund et al. 1998; Redman et al. 1999; McCarthy et al. 2002). Both Grolimund et al. (1998) and Redman et al. (1999) report that the critical concentration above which the ionic strength effect becomes significant in colloid attachment is ~10 to 100 mM for $Na^+$, and ~0.1 to 1 mM for $Ca^{2+}$. The ionic strength effect can be neglected in this study because of the very low ionic concentration (~0.1 mM) in background water, and the low $Na^+$ concentration (~10 mM) in the injection sources (Table 1).

Water pH affects the charge density and can even change the sign of the charge on the surface of the viruses and/or the porous medium based on the relationship between the isoelectric points ($pH_{iep}$) and the water pH. A particle (virion or sand) suspended in water is negatively charged if the pH is above its $pH_{iep}$, and vice versa. Particles with different $pH_{iep}$ exhibit a dramatic change of attachment behavior when a specific water pH (called critical pH in this paper) causes them to be oppositely charged. Both MS2 and ϕX174 experienced such a pH condition in this study. This is evident from the increase of more than an order of magnitude of the removal rates at pH 4.6 for MS2 and pH 6.1 for ϕX174, and the abrupt change of the relative attenuation and collision efficiency at pH 5 for MS2 and pH 6 for ϕX174 (Figure 4). When the water pH was below 5, the feldspar component in the porous medium was positively charged, and the MS2 was almost completely adsorbed irreversibly. When the water pH was below 6, ϕX174 was positively charged and was almost completely adsorbed irreversibly by the quartz component of the porous medium. Thus, our study indicates that the critical pH for a virus depends on the $pH_{iep}$ of both the virus and porous medium, and occurs at ~0.5 unit below the highest $pH_{iep}$. When the water pH is below the critical value (subcritical pH condi-

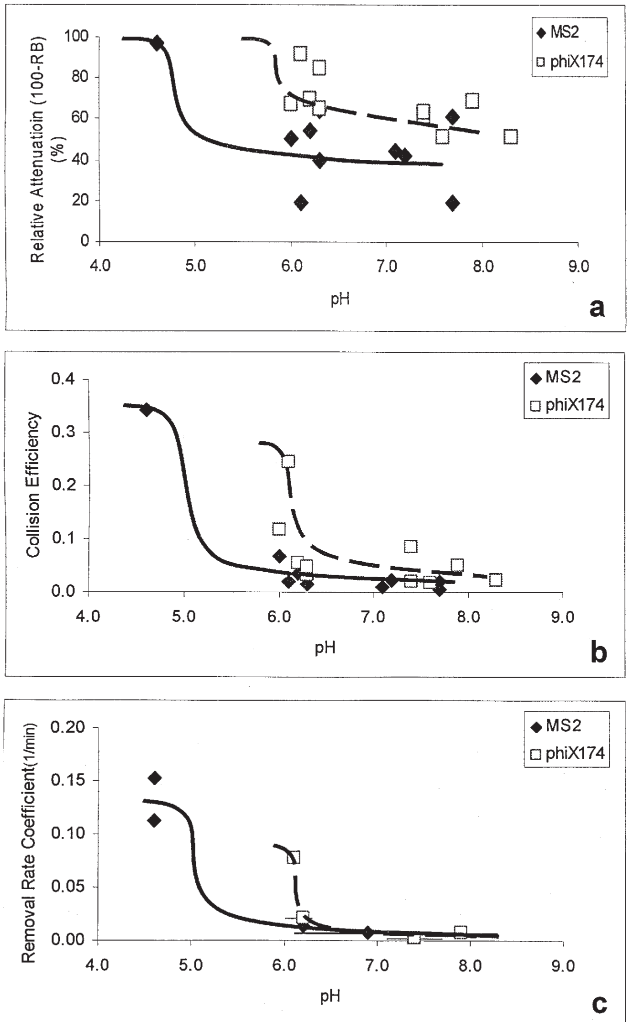


**Figure 4. (a) Calculated relative attenuation, (b) collision efficiency, and (c) simulated removal rate coefficient versus the plume pH.**

tion), the viruses are adsorbed almost completely. As the water pH increases above the critical value (supercritical pH condition), there is a dramatic decrease in virus removal due to both porous medium and viruses having negative or weakly opposite charges.

It is reasonable to assume that, at a supercritical pH condition, no adsorption occurs because viruses and the medium particles repulse each other. However, the experimental results show that relative attenuation rates are still 40% for MS2 and 60% for ϕX174 at a supercritical pH condition (Figure 4a). This suggests that some other processes, such as van der Waals force attraction, hydrophobic effect, and possibly straining also contribute to the removal of viruses from ground water. Schulze-Makuch et al. (2003) discuss these processes in detail. The electrostatic repulsion at a supercritical pH condition may hinder but not stop these processes. This is suggested by the observed slight decrease in relative attenuation and collision efficiency of the viruses with increasing water pH at supercritical pH conditions (Figures 4a and 4b).

The concept of critical pH is intended to describe a threshold pH condition at which electrostatic interaction between viruses and porous medium changes abruptly, without considering other components in the water. Thus the predicted results based on critical pH may deviate from actual results when the ground water is chemically complex. For example, organic molecules with similar $pH_{iep}$ in contaminated ground water may compete with viruses for adsorption at a subcritical pH condition, resulting in a lower attenuation rate (Pieper et al. 1997). Cations in the water, especially multivalent cations, may work as an electrostatic bridge to link viruses and porous medium particles of like charge, resulting in a higher attenuation rate than predicted at a supercritical pH condition (Loveland et al. 1996). Nonetheless, the estimated removal of viruses based on critical pH is observed for most uncontaminated fresh water aquifers (Table 4).

### Critical pH Observed in Other Studies

Some other studies with detailed description of porous medium and viruses are summarized in Table 4. For the iron oxide-coated sand, the isoelectric point of iron oxide (pH ~7 to 8) was used to estimate the critical pH, instead of the measured $pH_{iep}$ of a mixture of the quartz and ferric oxyhydroxides (pH 5.1 in Loveland et al. 1996). This is because viruses can be attached to sand surface with the iron oxide coating, and is not affected by the surface without the coating, just as in the case that MS2 was adsorbed almost completely by feldspar surfaces in the mixture of feldspar and quartz sand when pH is below 5. This effect of surface charge heterogeneity of porous media on colloid removal from ground water is discussed in detail by Ryan and Elimelech (1996).

The critical pH was clearly observed in the studies presented in Table 4 although it has not been explicitly pointed out. In batch experiments, Loveland et al. (1996) noticed that viral attachment changed abruptly at some pH levels. For the iron-coated sand experiment, this can be associated with the critical pH concept. For the clear sand experiment, the estimated critical pH value is one unit larger than the observed data. This might be due to the effect of static water in the experiments, which is different from the flowing water condition in this study. Two 1 m field experiments conducted by Ryan et al. (1999) and Pieper et al. (1997) had a subcritical pH condition for PRD1, and resulted in a relative attenuation comparable to the results we obtained for MS2 and ϕX174 at a subcritical pH condition. Deborde et al. (1999) and Woessner et al. (2001) conducted two field experiments with four viruses transporting 7.5 and 20 m, respectively. Polio virus experienced a subcritical pH condition and had an almost complete removal, while the other three viruses at supercritical pH conditions had significantly less attenuation.

### Some Implications of Critical pH

Removal of viruses from ground water increases significantly at a subcritical pH condition compared to that at a supercritical pH condition. The condition depends on water pH and $pH_{iep}$ of viruses and porous medium. Thus, a subcritical pH condition may be locally achieved using in situ barriers. For example, iron oxide-coated sand increased the critical pH for most viruses over 7.5, resulting in a subcritical pH condition in most natural ground water environ-

**Table 4**
**Critical pH Observed in Batch, Flowing Column, and Field Studies**

| References | Experiments | Porous Medium ($pH_{iep}$)[1] | Virus ($pH_{iep}$) | Water pH | Attenuation[2] (%) | Critical pH Based on This Study |
|---|---|---|---|---|---|---|
| This study | Flowing column (1-meter transport) | Quartz (3) and feldspar (5.5) sand | MS2 (3.9) | < 5 | > 90 (RA) | 5 ± 0.5 |
| | | | | > 5 | ~40 (RA) | |
| | | | ϕX174 (6.6) | ~ 6 | ~90 (RA) | 6 ± 0.5 |
| | | | | > 6 | ~60 (RA) | |
| Loveland et al. 1996 | Batch experiment | Iron oxide coated quartz sand (8)[3] | PRD1 (4.2) | < 7 | > 90 | 7.5 ± 0.5 |
| | | | | > 8 | < 5 | 7.5 ± 0.5 |
| | | Quartz without iron coating (2.5) | | < 5 | > 90 | 3.7 ± 0.5 |
| | | | | > 7 | < 10 | 3.7 ± 0.5 |
| Ryan et al. 1999 | Field experiment (1-meter transport) | Iron oxide coated sand (8) | PRD1 (4.2) | 5.4–5.6 | 95–99 (RA) | 7.5 ± 0.5 |
| Pieper et al. 1997 | Field experiment (1-meter transport) | Iron oxide coated sand (8) | PRD1 (4.2) | 5.0–5.7 | 78–88 (RA) | 7.5 ± 0.5 |
| Deborde et al. 1999 | Field experiment (7.5-meter transport) | Sand free of iron oxide coating (3–5) | MS2 (3.9) | 7.2 | 49 (RA) | 5 ± 0.5 |
| | | | PRD1 (4.2) | | 71 (RA) | 5 ± 0.5 |
| | | | ϕX174 (6.6) | | 65 (RA) | 6 ± 0.5 |
| | | | Attenuated polio (7.5) | | 99 (RA) | 7 ± 0.5 |
| Woessner et al. 2001 | Field experiment (20-meter transport) | Sand free of iron oxide coating (3–5) | MS2 (3.9) | 7.2 | 83 | 5 ± 0.5 |
| | | | PRD1 (4.2) | | 45 | 5 ± 0.5 |
| | | | ϕX174 (6.6) | | 93 | 6 ± 0.5 |
| | | | Polio type 1 (7.5) | | > 99 | 7 ± 0.5 |

[1]Isoelectric point of the porous medium is estimated based on the descriptions.
[2]Either percent of virus removal from the water or relative attenuation (marked as RA)
[3]Loveland reported 5.1; the $pH_{iep}$ of iron oxide is used here. Refer to text for details.

ments. Cationic surfactant modified zeolite also increases the critical pH for viruses, and may create a subcritical pH condition. Schulze-Makuch et al. (2002) succeeded in removing MS2 from ground water using this approach.

## Conclusion

Both numerical simulations and colloid filtration theory calculations on the experimental data indicate a critical pH at which the virus behavior changes abruptly. It is ~0.5 unit below the highest $pH_{iep}$ of the porous medium and the virus. When water pH was below the critical pH, most of the virus was removed from water irreversibly. The critical pH is also observed from other field and batch experiments. This suggests that electrostatic interaction is an important factor controlling virus attenuation in ground water. Additionally, some other processes also contribute to virus removal from the water. The concept of critical pH will assist in the design of geologic barriers for preventing viral contamination in ground water.


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