Virus removal by soil passage at field scale and groundwater protection of sandy aquifers

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Abstract Virus removal from groundwater by soil passage often appears to be much higher during the first few metres due to the presence of more favorable sites for attachment than thereafter. A model is presented which interprets virus removal as a function of collision efficiencies α_β and α_λ, inactivation rate coefficient μ_I and rate parameter γ. Initial high removal is determined by ab, which decreases exponentially at a rate g to a constant base removal rate that is determined by α_λ and μ_I.

A hypothetical worst case was simulated to calculate the travel distance and time required for 9 $\log_{10}$ protection against virus contamination of groundwater wells in anoxic sandy aquifers. Unfavorable conditions for attachment were assumed. Virus was constantly leaking from a sewage pipe lying at the groundwater table. Mixing reduced virus concentration by 3.1 to 4.0 $\log_{10}$. For an additional 5.0 to 5.9 l $\log_{10}$ protection against virus contamination by attachment and inactivation, residence times of about three to seven times longer than the current guideline of 60 days are needed, depending on abstraction rates, aquifer thickness and grain size of the sand.

Key words Groundwater; protection; transport; virus

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Introduction

Theoretical and practical knowledge about virus removal from groundwater in terms of process parameters and boundary conditions at field scale was summarized. The aims were to evaluate prediction of virus removal by soil passage as a treatment in drinking water production and prediction of a zone that allows 9 $\log_{10}$ protection of groundwater wells against virus contamination.

Only relatively few well defined field studies on virus removal by soil passage exist. Removal data from these field studies as well as relevant conditions are compared. The aim is to identify some typical cases that can be used to predict virus removal at other sites under similar conditions. As pointed out by Schijven and Hassanizadeh (2000), removal rate of virus often appears to be higher nearer the source than further away due to the presence of more favorable sites for attachment near the source than thereafter. A model is presented to account for this chemical heterogeneity. This model is applied to obtain specific parameter values to quantify this heterogeniety. Worst case conditions for virus removal from these field studies were applied to calculate adequate residence times and setback distances that are required for 9 $\log_{10}$ protection of groundwater wells in sandy aquifers against virus contamination.

Methods

Virus removal model

A high initial removal can be ascribed to special attachment sites that are present in the first few metres of soil passage but rapidly decrease with travel distance or travel time in an exponential fashion, like sites formed by ferric oxyhydroxides in a deep well injection study (Schijven *et al.*, 2000), or by ferric oxyhydroxides and organic carbon in a dune recharge study (Schijven, 2001). Thus, under steady-state conditions and assuming negligible dispersion, virus removal can be described by:

$$\log_{10}\left(C/C_0\right) = \frac{1}{2.3}\left\{\frac{\beta}{\gamma}\left[\exp(-\gamma t) - 1\right] - \left(\lambda + \mu_l\right)t\right\} \tag{1}$$

where, $\log_{10}(C/C_0)$ defines removal, β is the maximum attachment rate coefficient to favorable attachment sites [T^{-1}], γ is the rate coefficient with which the attachment rate decreases due to an exponential decrease in the number of attachment sites with travel time [T^{-1}]. Parameter λ is the base removal rate coefficient due to attachment to a constant base level of attachment sites [T^{-1}], μ_l is the inactivation rate coefficient [T^{-1}].

Eq. (1) has been used to fit field data from Bales *et al.* (1995, 1997), Pieper *et al.* (1997), DeBorde *et al.* (1998, 1999), Ryan *et al.* (1999) and Schijven *et al.* (1998, 1999, 2000) to obtain values of parameters β, γ and λ. Corresponding values of collision efficiencies (α_β and α_λ) of the viruses were calculated applying the equations from colloid filtration theory according to Yao *et al.* (1971). In most of these studies, removal of bacteriophages MS2 or PRD1 was measured. These bacteriophages represent low adsorbing negatively charged viruses. A default value of 0.024 day^{-1} for the inactivation rate coefficient μ_l was applied. This is the low value that was found for MS2 in the deep well injection study by Schijven *et al.* (2000).

In some cases, only two to three measurements of removal with time were available (Bales *et al.*, 1997; Pieper *et al.*, 1997; DeBorde *et al.*,1999; Ryan *et al.*, 1999; Schijven *et al.*, 1998, 2000). For these data, Eq. (1) was not applied, but, for the sake of comparison, removal rates were calculated from the linear increase in time.

Worst case simulation for protection of groundwater wells

The following hypothetical case was simulated to calculate the residence and setback distance that are required for sufficient protection against virus contamination: water is abstracted continuously from a phreatic aquifer at a rate of Q_A $\text{m}^3.\text{day}^{-1}$. At a distance R from the abstraction well and right at the groundwater table, a sewage pipe is continuously leaking and thus contaminating the groundwater with viruses. A leakage rate q of 1 $\text{m}^3.\text{day}^{-1}$ was assumed, which may remain unnoticed. Only horizontal transport in the direction of the abstraction well is considered. The protective effects of confining or semi-permeable layers are not considered, because local differences in the thickness of confining layers due to irregularities and effects of erosion are regarded as a considerable source of uncertainty for protection.

Under steady-state conditions, neglecting dispersion, virus transport is described by:

$$\log_{10}\left(\frac{C_A}{C_0}\right) = -\frac{1}{2.3}\left(\frac{3}{5}\alpha k_1 R^{5/3} + \frac{1}{2}\mu_l k_2 R^2\right) + \log_{10}\left(\frac{q}{Q_A}\right) \tag{2}$$

where, protection $\log_{10}(C_A/C_0)$ is described by attachment to the soil grains (first right term), by inactivation (second right term), and dilution (third right term). Detachment is neglected, because it is usually much slower than attachment. C is the concentration of virus [L^{-3}].

$$k_1 = \frac{3}{2}\frac{(1-n)}{d_c}4A_s^{1/3}\left(\frac{D_{BM}2\pi h}{d_c Q}\right)^{2/3} \text{ and } k_2 = \frac{2\pi n h}{Q} \tag{3}$$

where, d_c is the average diameter of the single collector (soil grain), [L] and α is the collision efficiency. $A_s = 2(1 - \gamma^5)/(2 - 3\gamma + 3\gamma^5 - 2\gamma^6)$ is Happel's porosity dependent parameter, with $\gamma = (1 - n)^{1/3}$. $D_{BM} = K_B(T + 273)/(3\pi d_p \mu)$ is the diffusion coefficient,

$[L^2T^{-1}]$; $K_B = 1.38 \times 10^{-23}$ is the Boltzmann constant $[J.K^{-1}]$; T is the water temperature [°C]; d_p is the virus particle size [L]; μ is the dynamic viscosity $[ML^{-1}T^{-1}]$;

A selection was made of Dutch groundwater well systems located in phreatic sandy aquifers where the screens of the production wells are at a depth of less than 25 m. From these aquifers, a subset of six aquifers was selected, in which pH of the abstracted water is 7.1 to 7.3 and levels of dissolved oxygen and nitrate are less than 0.5 mg.l^{-1}. In addition, it was assumed that no pyrite is present. Under these anoxic conditions, one may assume that preferential sites for attachment of viruses are absent, as was found in the anoxic part of a deep well injection study (Schijven *et al.*, 2000). The absence of confining layers together with the shallowness of the aquifers and unfavorable conditions for attachment make it a reasonable assumption qualifying these groundwater well systems as relatively vulnerable. The collision efficiency α_λ for bacteriophage MS2 for this aquifer was used. In that field study, a value of 0.024 day^{-1} for μ_l of MS2 was measured, which may also be considered as a worst case value. At higher pH, electrostatic repulsion between the surfaces of viruses and soil grains is higher, which is reflected by lower values of the collision efficiency. The decrease in collision efficiency of MS2 due to an increase in pH was derived from the data from the column studies by Bales *et al.* (1991, 1993), Kinoshita *et al.* (1993) and Penrod *et al.* (1996): in the pH range of 3.5 to 7, α decreases by a factor 0.9 for every increase in pH by 0.1. This rule was applied to adjust α for the different pH.

In domestic wastewater at two large sewage treatment plants in The Netherlands, geometric mean concentrations of 34 and 190 enteroviruses per litre were found (Hoogenboezem *et al.*, 2000). Based on an infection risk of 1 per 10^4 persons per year and assuming rotavirus infectivity, the maximum allowable concentration level of viruses at the abstraction well would be 1.8×10^{-7} viruses per litre (Regli *et al.*, 1991). This implies that virus concentrations in domestic wastewater from a leaking pipe should be reduced by 9 $\log_{10}$ at the point of groundwater abstraction.

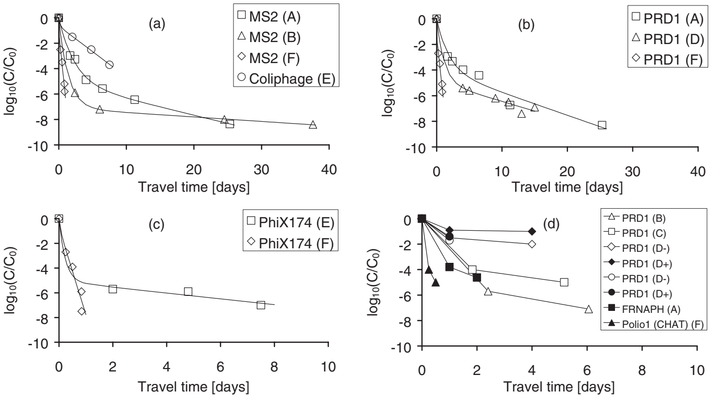
Results

Virus removal at field scale

Figures 1a–d show the removal data from the field studies. Aquifer A contains very fine dune sand. The initial removal rate was found to be strongly and positively correlated with soil organic carbon content and to a lesser extent with the content of ferric oxyhydroxides (Schijven, 2001). Due to the high pH, conditions for attachment are unfavorable, as evidenced by the low collision efficiencies (Table 1).

Aquifer B contains fine sand. Collision efficiency α_β is higher than in aquifer A due to the presence of favorable attachment sites in the form of ferric oxyhydroxides together with a lower pH. However, collision efficiency α_λ in aquifer B is lower than that in aquifer A, due to fewer sites for attachment in the anoxic part of aquifer B than in aquifer A. Eventually, 8-$\log_{10}$ removal was achieved in both aquifers. The sand in aquifer C is coarser, but otherwise similar to A or B. The collision efficiencies for PRD1 in aquifer C were not calculated because data on grain size and temperature were not reported. However, collision efficiencies of 0.0028–0.003 at pH 7.4 and of 0.00085–0.0016 at pH 8.4 were reported by the investigators (Bales *et al.*, 1997), similar to those in aquifers B and C. In aquifer C, pH is higher than in B. Probably, more favorable sites are available in aquifer C than initially in B. Aquifer D consists of a sewage contaminated (D+) and uncontaminated zone (D–). Organic matter limits attachment in D+ by occupying attachment sites, whereas a higher pH is the limiting factor in A and C. In aquifer D–, removal rates and collision efficiencies are higher than in aquifer D+, because in D–, attachment sites are not blocked by organic matter.

The sand in aquifer E is coarse and pore water velocity is high. The initial removal rate of



Figures 1a–d Removal of bacteriophages and poliovirus 1 as a function of travel time in sandy aquifers. See corresponding parameter values in Table 1

Table 1 Removal rate parameter values from field studies

Fig.	Virus	Aquifer	β	γ	λ	$\alpha_\beta \times 10^{-3}$	$\alpha_\lambda \times 10^{-3}$	Reference
1a	MS2	A	5.0	0.43	0.30	1.2	0.072	Schijven *et al.*, 1999
	MS2	B	12	0.71	0.081	2.6	0.015	Schijven *et al.*, 2000
	MS2	F	6.4	25	12	22	4.2	DeBorde *et al.*, 1999
	Coliphage	E	15	9.8	0.92	61	3.7	DeBorde *et al.*, 1998
1b	PRD1	A	8.5	0.53	0.41	3.8	0.069	Schijven *et al.*, 1999
	PRD1	D–	9.9	0.88	0.35	18	0.67	Bales *et al.*, 1995
	PRD1	F	103	32	11	67	6.9	DeBorde *et al.*, 1999
1c	ϕX174	E	129	11	0.56	520	2.2	DeBorde *et al.*, 1998
	ϕX174	F	0.62	0	18	0.24	6.6	DeBorde *et al.*, 1999
				λ_1	λ_2	α_1	α_2	
1d	PRD1	B		5.5	0.87	2.1	0.21	Schijven *et al.* 2000
	PRD1	C		5.1	0.69			Bales *et al.*, 1997
	PRD1	D–		3.5	0.39	7.3	0.78	Pieper *et al.*, 1997
	PRD1	D+		2.1	0.076	4.4	0.001	Pieper *et al.*, 1997
	PRD1	D–		3.9		8.1		Ryan *et al.*, 1999
	PRD1	D+		3.2		6.7		Ryan *et al.*, 1999
	FRNA phage	A		8.7	1.9	2.0	0.78	Schijven *et al.*, 1998
	Polio 1	F		37	9.2	13	3.3	DeBorde *et al.*, 1999

coliphages is similar to that of MS2 in B, but the base removal rate is ten times higher in E compared to that in B. The initial removal of ϕX174 in aquifer E is very high, but its base removal rate is only about twice that of MS2 in aquifer A.

Bacteriophage ϕX174 is expected to attach much more than MS2 because its surface charge is less negative than that of MS2. The relatively low base removal rates in aquifer E are probably due to the coarseness of the soil, which lead to low single collector efficiencies.

In aquifer F, we have the coarsest sand and, by far, the highest hydraulic conductivity and pore water velocity. In aquifer F we also find the highest collision efficiencies, suggesting the presence of many favorable attachment sites.

From the data shown in Figures 1a and 1b, it is possible to identify a worst case situation, i.e. where interaction with the soil grains is very low, due to the absence or blocking of preferable sites for attachment. In aquifers B and D+, very low base removal rates of 0.08 day^{-1} were found for MS2 and PRD1, respectively. In aquifer B, the low removal rate of MS2 took place in the anoxic part of the aquifer with few sites for attachment. In aquifer D+, attachment of PRD1 was limited due to the presence of organic matter occupying attachment sites. This latter condition may represent a situation of virus contamination from a wastewater source.

Groundwater well protection

The required residence times and setback distances for 9 $\log_{10}$ protection against virus contamination are listed in Table 2 and plotted in Figures 2a and 2b. Depending on the abstraction rate, the dilution factor varies between 3.1 and 4.0 $\log_{10}$, therefore an additional 5.0 to 5.9 $\log_{10}$ removal of virus is needed. To achieve this, residence times of 209 to 442 days are needed, depending on abstraction rates, aquifer thickness and grain size of the sand.

Conclusions

Virus removal by soil passage

To conclude, pH and the presence of favorable sites for attachment seem to be the major factors determining attachment of virus to the soil grains. This is in line with observations

Table 2 Hydrologic properties of selected Dutch phreatic aquifers

	Aq1	Aq2	Aq3	Aq4	Aq5	Aq6
Q [m^3.day^{-1}]	3096	1781	1370	8219	9589	4658
h [m]	30	25	23	20	25	20
$\log_{10}(Q/q)$	3.5	3.3	3.1	3.9	4.0	3.7
$9 - \log_{10}(Q/q)$	5.5	5.7	5.9	5.1	5.0	5.3
Required residence times for 9 $\log_{10}$ protection [days]	408	442	434	342	209	235
Corresponding setback distance [m]	196	169	153	357	270	223

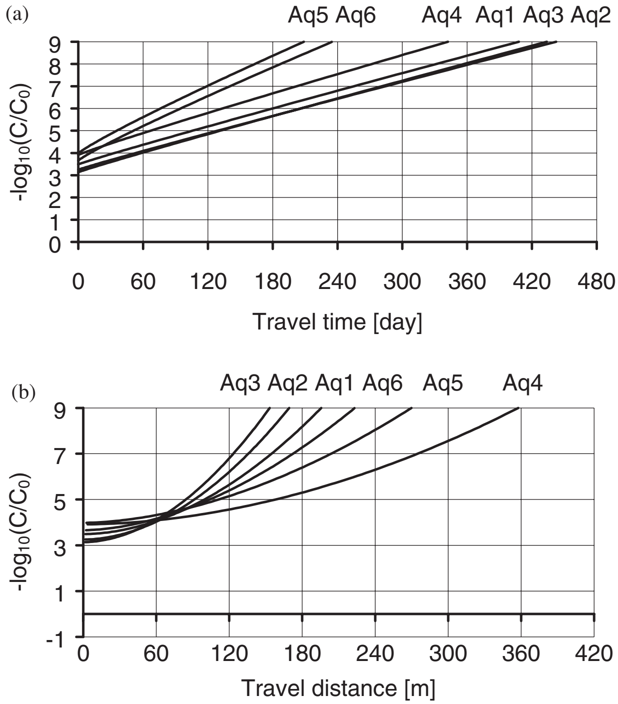


Figure 2 Virus removal in six phreatic aquifers as a function of travel time (a) and travel distance (b)

by Elimelech *et al.* (2000), where pH together with the presence of heterogeneously distributed patches of iron oxide coatings were found to be the key factors in determining attachment of colloids to soil.

Collision efficiencies α_β and α_λ are both determined by the fraction and nature of favorable attachment sites that are present. This requires detailed knowledge about the soil properties that may be obtained from geochemical analyses of soil samples and be supported by studying attachment of MS2 in column experiments. Similarly, from analysis of different soil samples along a flow line, one may estimate γ. E.g. γ may be related to a decrease in dissolved oxygen with travel time.

If for a given site, the fraction of favorable sites as a function of travel time is known, and pH, grain size, porosity and temperature are known, one should be able to derive values for β, γ and λ. Then, by applying Eq. (2) one can calculate (non-linear) removal of MS2 as a function of travel time. Of course, in the case of similar situations as the well characterized dune recharge site (Schijven *et al.*, 1999) and deep well injection site (Schijven *et al.*, 2000), one may also reasonably assume that removal rates are the same as at these sites.

Protection of groundwater wells aqainst virus contamination

Based on the worst case simulations, it can be concluded that the current guideline of a 60-day residence time is inadequate, and that residence times of about three to seven times the current guideline are needed for sufficient protection of groundwater wells in sandy aquifers against virus contamination.

The worst case depicted here may be even worse, because of higher leakage rates and the possibility of higher concentrations of pathogenic viruses in raw wastewater, like human caliciviruses (Lodder *et al.*, 1999). But these worsening factors may already have been compensated by protective effects of confining layers and vertical transport through (un)saturated zones.

Future studies on vulnerability of groundwater wells to virus contamination should include investigating the frequency, probability and extent of leakage rates. The physical and chemical characteristics of the porous medium should be studied in more detail in order to evaluate the extent of virus attachment. Finally, effects of confining layers and vertical transport through (un)saturated zones may also be taken into consideration.

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