

Determination of protection zones for Dutch groundwater wells against virus contamination - uncertainty and sensitivity analysis

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ABSTRACT

Protection zones of shallow unconfined aquifers in The Netherlands were calculated that allow protection against virus contamination to the level that the infection risk of 10^{-4} per person per year is not exceeded with a 95% certainty. An uncertainty and a sensitivity analysis of the calculated protection zones were included. It was concluded that protection zones of 1 to 2 years travel time (206–418 m) are needed (6 to 12 times the currently applied travel time of 60 days). This will lead to enlargement of protection zones, encompassing 110 unconfined groundwater well systems that produce $3 \times 10^8 \text{ m}^3 \text{ y}^{-1}$ of drinking water (38% of total Dutch production from groundwater). A smaller protection zone is possible if it can be shown that an aquifer has properties that lead to greater reduction of virus contamination, like more attachment. Deeper aquifers beneath aquitards of at least 2 years of vertical travel time are adequately protected because vertical flow in the aquitards is only 0.7 m per year. The most sensitive parameters are virus attachment and inactivation. The next most sensitive parameters are grain size of the sand, abstraction rate of groundwater, virus concentrations in raw sewage and consumption of unboiled drinking water. Research is recommended on additional protection by attachment and under unsaturated conditions.

Key words | contamination, groundwater, protection, sensitivity, uncertainty, virus

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INTRODUCTION

The current Dutch legislation for drinking water states that the risk of infection by a pathogen due to drinking water consumption may not exceed the level of 10^{-4} per person per year ($\text{p}^{-1}\text{y}^{-1}$) (Staatsblad 2001). This implies that protection zones for groundwater wells should be large

enough to comply with this quality standard. Current protection of groundwater wells in The Netherlands is still based on the assumption that a travel time of 60 days is sufficient for die-off of pathogenic microorganisms in contaminated groundwater to the extent that no health

risks exists (CBW 1980). However, it has become clear that not only pathogenic viruses and protozoa but also bacteria can survive much longer than 60 days in soil and groundwater (Schijven *et al.* 1995). Furthermore, a health risk of zero is not feasible. Recently, sizes of protection zones for Dutch groundwater wells were calculated for a selection of shallow unconfined sandy aquifers (Schijven & Hassanizadeh 2002). Viruses leaking from a sewage pipe were simulated with slow virus inactivation and a low rate of virus attachment to the sand assumed. Viruses are the most critical pathogens in this regard. Viruses are very persistent and are easily transported because of their small size and because they may attach poorly.

Protection zones were calculated that are large enough in order to guarantee 9-log₁₀ reduction of virus concentrations and thereby not exceed a maximum allowable concentration (MAC) of 1.8×10^{-7} viruses per litre at the pumping well. This MAC was derived from a maximum infection risk of 10^{-4} p⁻¹y⁻¹, consumption of unboiled drinking water and the dose response relation of rotaviruses (Regli *et al.* 1991; Medema & Havelaar 1994). It was concluded that protection zones with a travel time of 470–885 days, *i.e.* three to seven times the current 60 days guideline, would be needed.

These protection zones were based on single, partly conservative, estimates of parameter values, therefore uncertainty of the size of these zones was not estimated. The present study was aimed at determining the size of required protection zones for the same selection of aquifers, but now including uncertainties of all model parameters. By including uncertainties, protection zones could be dimensioned such as to comply with the risk of infection of 10^{-4} p⁻¹y⁻¹ with 95% certainty.

Also, a sensitivity analysis was conducted in order to identify which of the model parameters are most important in determining the size of the protection zone. In addition, a conclusion was drawn on the vulnerability of deeper, completely or partially confined aquifers.

METHODS

Model description

Contamination of a single production well by viruses originating from a leaky sewage pipe was considered. The

well was assumed to be situated in a homogeneous unconfined aquifer with a thickness h [L]. These aquifers are sandy soils with a relatively high permeability, which is needed for producing sufficient quantities of drinking water and are therefore relatively homogenous, especially in the horizontal direction. The leakage was assumed to occur just below the groundwater table at a distance R [L] from the well. As the distance R is much larger than h , flow lines from the leakage point to the well were assumed to have the total length R and only horizontal movement of viruses was considered. Also, for typical aquifers the Peclet number is much larger than one and thus dispersion was neglected. This is also justified by the low dispersion that was found in the sandy aquifers in the field studies by Schijven *et al.* (1999, 2000). The groundwater table drawdown at the well was neglected as it was expected to be small compared to R . It was assumed that the leaking sewage water would be completely abstracted by the well. Under steady state conditions, virus transport is described by:

$$\frac{dC}{dr} + \frac{k_{att} + \mu_1}{v} C = 0 \quad (1)$$

where, C is the concentration of the virus [L⁻³] and r is the distance from the well [L]. k_{att} is the attachment rate coefficient [T⁻¹]. Detachment was neglected, because it usually proceeds much slower than attachment. Thus, attachment was assumed to be irreversible. μ_1 is the inactivation rate coefficient of free viruses in the water [T⁻¹]. Pore water velocity v [LT⁻¹] increases in the direction of the production well according to:

$$v = -\frac{Q}{2\pi nr} \quad (2)$$

where Q [L³T⁻¹] is the well production rate and n is the porosity.

According to filtration theory (Yao *et al.* 1971), but only considering diffusion, because transport of small viruses in the immediate vicinity of soil grains is dominated by Brownian diffusion (Penrod *et al.* 1996), k_{att} can be calculated as follows:

$$k_{att} = 6 \frac{(1-n)}{d_c} \alpha A_s^{1/3} \left(\frac{D_{BM}}{d_c n v} \right)^{2/3} v \quad (3)$$

where, d_c [m] is the average diameter of the so-called single collector (grain of sand); $A_s = 2(1 - \gamma^5)/(2 - 3\gamma + 3\gamma^5 - 2\gamma^6)$

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, [$\text{m}^2 \text{s}^{-1}$]; $K_B = 1.38 \times 10^{-23}$ is the Boltzmann constant [J.K^{-1}]; t is the water temperature [$^{\circ}\text{C}$]; d_p is the particle size of the viruses [m]; $\mu = \rho * 0.000947/(t + 42.5)^{1.5}$ is the dynamic viscosity [$\text{kg m}^{-1}\text{s}^{-1}$] with $\rho = 999.703 \text{ kg m}^{-3}$ the density of water.

Substitution of equations (2) and (3) into (1) gives:

$$\frac{dC}{dr} - \left(\alpha k r^{2/3} + \mu_l \frac{2\pi n h}{Q} r \right) C = 0 \quad (4)$$

where $k = 6 \frac{(1-n)}{d_c} A_s^{1/3} \left(\frac{D_{BM} 2\pi n h}{d_c Q} \right)^{2/3}$.

Thus equation (4) has the following analytical solution:

$$\ln(C) = \frac{3}{5} \alpha k r^{5/3} + \mu_l \frac{\pi n h}{Q} r^2 + C^* \quad (5)$$

where C^* is an integration constant.

Subject to boundary conditions $C_R = C_0 \frac{q}{Q_R}$ at $r = R$, where C_0 is the initial virus concentration in the sewage pipe [L^{-3}], C_R is the virus concentration in the aquifer at R [L^{-3}], q is the leakage rate of sewage water [$\text{L}^3 \text{T}^{-1}$] and Q_R is the flow rate in the aquifer at R [$\text{L}^3 \text{T}^{-1}$]. It is assumed that virus attachment is not affected by the size of the hole through which viruses are leaking. Therefore, the virus concentration at R is described by:

$$\ln(C_R) = \frac{3}{5} \alpha k R^{5/3} + \mu_l \frac{\pi n h}{Q} R^2 + C^* \quad (6)$$

Subject to boundary conditions $C_A = C_W \frac{Q_0}{Q}$ at $r = W$, where C_A is the virus concentration in the abstracted water [L^{-3}], C_W is the virus concentration at W [L^{-3}] and W is the radius of the well [L]. The virus concentration at W is described by:

$$\ln(C_W) = \frac{3}{5} \alpha k W^{5/3} + \mu_l \frac{\pi n h}{Q} W^2 + C^* \quad (7)$$

By substitution of boundary conditions, subtracting equation (6) from (7) and neglecting W , because $W \ll R$, one obtains the equation for calculating virus removal (logarithmic reduction of virus concentration):

$$\log_{10} \left(\frac{C_A}{C_0} \right) = - \frac{1}{2.3} \left(\frac{3}{5} \alpha k R^{5/3} + \mu_l \frac{\pi n h}{Q} R^2 \right) + \log_{10} \left(\frac{q}{Q} \right) \quad (8)$$

Here, removal is determined by attachment to grains of sand (first term) with collision efficiency α , by inactivation

of viruses (second term) with inactivation rate coefficient μ_l , [T^{-1}], and by dilution (third term).

The risk of virus infection due to consumption of unboiled abstracted groundwater was estimated, including various uncertainties in the model parameters as described below. First, the virus concentration at the well C_A was calculated. Next, the dose D , which is the number of ingested virus particles, was calculated:

$$D = \frac{C_A}{E} V \quad (9)$$

Here, E is the recovery efficiency of the virus enumeration method which is the detected fraction of the virus particles that are present in a sample of water, V is the volume of ingested unboiled drinking water in litres per person per year ($1 \text{ p}^{-1} \text{ y}^{-1}$) (excluding water that is previously boiled like in cooking, coffee, etc.).

Because D will commonly be low, the following approximation of the Beta Poisson dose response model for infection was applied (Teunis & Havelaar 2000):

$$p_{\text{inf}} = p_m D \quad (10)$$

Here, p_{inf} is the risk of infection and p_m is the infectivity of a virus.

Travel time T was calculated from R according to:

$$T = \frac{\pi n h R^2}{Q} \quad (11)$$

The set of equations (8) to (11) prescribe a relation between R and p_{inf} or T and p_{inf} , with various properties of water and aquifers as model parameters.

Uncertainty analysis

The uncertainty analysis was carried out through Monte Carlo simulations. A number of model parameters whose values were considered to be uncertain were chosen. These were grouped into those non-aquifer specific and aquifer specific. Non-aquifer-specific model parameters are virus concentration in raw sewage, C_0 , recovery efficiency, E , consumption of unboiled drinking water, V , leakage rate from a sewage pipe, q , size of virus particles, d_p , virus inactivation rate coefficient, μ_l and virus infectivity, p_m

Table 1 | Distributions of the non-aquifer-specific model parameters

Parameter		Distribution[par1; par1]	Average	95% interval
Enterovirus concentration in raw sewage	C_0 [n/l]	Lognormal[4.23; 1.32]	150	5.3–820
Norovirus concentration in raw sewage	C_0 [n/l]	Lognormal[11.9; 1.62]	5.3×10^5	5.6×10^3 – 3.6×10^6
Recovery	E	Constant	1	
Consumption of unboiled drinking water	V [$l\ p^{-1}d^{-1}$]	Lognormal [– 1.88391; 1.12209]	0.27	0.017–1.3
Leakage rate of sewage pipe	q [$m^3\ s^{-1}$]	Lognormal[0; 0.5]	1.1	0.38–2.6
Porosity	n	Uniform[0.25; 0.50]	0.37	0.25–0.50
Virus size	d_p [m]	Uniform [2.0×10^{-8} ; 3.0×10^{-8}]	2.5×10^{-8}	2.0×10^{-8} – 3.0×10^{-8}
Inactivation rate coefficient	μ_1 [d^{-1}]	Lognormal[log(0.024); 0.5]	0.027	0.0089–0.064
Infectivity (rotavirus)	p_m	$\hat{a} = 0.253$ and $\hat{b} = 0.422$, Beta-distributed	0.64	0.26–0.87

(Table 1). Aquifer specific model parameters are, porosity, n , aquifer thickness, h , abstraction rate, Q , groundwater temperature, t , grain size, d_c , pH and collision efficiency, α (Table 2).

As part of the uncertainty analysis, model parameter values were obtained from various sources.

Lognormal distributions were fitted to measured virus concentrations in sewage. For all other model parameters various types of distributions were assumed. From each of these distributions, 10 000 values were randomly drawn using Mathematica v 4.2.0.0 (Wolfram Research). Distributions of virus concentrations at the well (equation 8) and of risks of infection were obtained as a function of distance R and travel time T by applying equations (9), (10) and (11).

In addition, the size of the protection zone was calculated for $p_{inf} = 10^{-4} p^{-1} y^{-1}$. Uncertainty in the size of protection zone was evaluated by repeating this for all Monte Carlo samples of the parameters mentioned above. (To that aim the “Solve” command in Mathematica was used to obtain distributions of distance and travel time as a function of the risk of infection).

Non-aquifer-specific model parameters

Virus concentration in raw sewage, C_0

Virus concentration in raw sewage was based on measurements of enterovirus concentrations in raw sewage at two large sewage treatment plants (STP’s) in Rotterdam and Amsterdam (Hoogenboezem *et al.* 2000). These STP’s treat primarily domestic sewage. Enterovirus concentrations were measured for a period of one year. The virus concentrations were assumed to be lognormally distributed.

On the basis of a likelihood ratio test (Cox & Hinkley 1974), a small but significant difference between the average concentrations of both STP’s was found, but no difference between variances. The data of both STP’s were nevertheless combined to obtain a representative value for The Netherlands.

In sewage noroviruses may also be present. These viruses are a major cause of gastro-enteritis in The Netherlands (de Roda Husman 2001). Concentrations of noroviruses in sewage cannot be determined by means of tissue culture as is the case with enteroviruses, but by reverse transcription polymerase reaction (RT-PCR)

Table 2 | Distributions of the aquifer-specific model parameters

Parameter		Aquifer	Distribution[par1; par2]	Average	95% interval
Porosity	n	Aq1 - Aq6	Uniform[0.25; 0.50]	0.37	0.25–0.50
Aquifer thickness	h [m]	Aq1	Uniform[25; 35]	30	25–35
		Aq2	Uniform[20; 30]	25	20–30
		Aq3	Uniform[18; 28]	23	18–28
		Aq4	Uniform[15; 25]	20	15–25
		Aq5	Uniform[20; 30]	25	20–30
		Aq6	Uniform[15; 25]	20	15–25
Abstraction rate	Q [m ³ day ⁻¹]	Aq1	Constant, 3096		
		Aq2	Constant, 1781		
		Aq3	Constant, 1370		
		Aq4	Constant, 8219		
		Aq5	Constant, 9589		
		Aq6	Constant, 4658		
Temperature	T [°C]	Aq1	Uniform[10.0; 11.2]	10.6	10.0–11.1
		Aq2	Uniform[9.6; 10.6]	10.1	9.6–10.6
		Aq3	Uniform[9.5; 11.5]	10.5	9.5–11.5
		Aq4	Uniform[10.0; 11.4]	10.7	10.0–11.4
		Aq5	Uniform[9.5; 11.7]	10.5	9.4–11.7
		Aq6	Uniform[9.5; 10.1]	9.8	9.5–10.1
Grain size	d_c [m]	Aq1- Aq4	Lognormal[5.0×10^{-4} ; 0.4]	5.4×10^{-4}	2.3×10^{-4} – 1.1×10^{-3}
		Aq5, Aq6	Lognormal[2.5×10^{-4} ; 0.4]	2.5×10^{-4}	1.2×10^{-4} – 5.6×10^{-4}
pH		Aq1	Normal[7.2; 0.20]	7.2	6.8–7.6
		Aq2	Normal[7.4; 0.10]	7.4	7.2–7.6
		Aq3	Normal[7.1; 0.15]	7.1	6.8–7.4
		Aq4	Normal[7.1; 0.13]	7.1	6.8–7.4
		Aq5	Normal[7.2; 0.15]	7.2	6.9–7.5
		Aq6	Normal[7.2; 0.15]	7.2	6.9–7.5
Collision efficiency	α	Aq1	Equation (12)	1.0×10^{-5}	6.5×10^{-6} – 1.5×10^{-5}
		Aq2	Equation (12)	8.0×10^{-6}	6.5×10^{-6} – 9.8×10^{-6}
		Aq3	Equation (12)	1.1×10^{-5}	8.0×10^{-6} – 1.5×10^{-5}
		Aq4	Equation (12)	1.1×10^{-5}	8.4×10^{-6} – 1.4×10^{-5}
		Aq5	Equation (12)	1.0×10^{-5}	7.2×10^{-6} – 1.3×10^{-5}
		Aq6	Equation (12)	1.0×10^{-5}	7.2×10^{-6} – 1.3×10^{-5}

allowing presence/absence type of detection (Lodder 1999). By means of tissue culture, virus particles are detected that were able to infect a cell, but by RT-PCR non-infectious virus particles are also detected. Norovirus concentrations (expressed in PCR-detectable units per litre) appear several orders in magnitude higher than enterovirus concentrations (Hoogenboezem *et al.* 2000), however, the fraction of infectious norovirus particles is unknown. The data for noroviruses were obtained from measurements over a period of one year at two STP's in St. Maartensdijk and Tholen (van den Berg *et al.* 2004). A lognormal distribution of the concentrations was assumed. According to the likelihood ratio test, no significant differences existed between the two data sets and therefore they were pooled.

Recovery efficiency, E

No data for the recovery efficiency of the virus enumeration method, E , are available. For enteroviruses enumeration was assumed to be without any loss ($E = 1$).

Consumption of unboiled drinking water V [$l\ p^{-1}d^{-1}$]

Data on consumption of unboiled drinking water were taken from Teunis *et al.* (1997). These data are also lognormally distributed. But for the Monte Carlo simulations only values less than $3l\ p^{-1}d^{-1}$, were used, because it was considered unlikely that a person drinks more.

Leakage rate from a sewage pipe, q [$m^3\ day^{-1}$]

In the present study, it was assumed that the leakage rate at a single leakage point, q , is $1\ m^3\ day^{-1}$. At such a rate, a leakage may probably remain unnoticed. It is equivalent to the sewage production from about 8 persons (Voorhoeve & van de Kerk 2003). A lognormal distribution was assumed, because uncertainty about leakage rates may extend two orders in magnitude

Only leakage from a single point source was considered in the calculations. Contributions of additional nearby leakages were accounted for in the sensitivity analysis by varying the leakage rate.

Leakage is caused by subsidence of pipes, where joints open. If the sewers are located below the groundwater table, infiltration of groundwater occurs and if the sewers are located above the groundwater table, exfiltration of sewage

may occur. The amount of sewage that can enter the aquifer depends on the size and location of the leakage in the pipe, the flow rate of the sewage and the permeability of the aquifer. Exfiltration is more likely to occur in sandy aquifers. In clay soils, low permeability may prevent exfiltration. Peat is very sensitive to subsidence, but here sewage pipes are commonly below the groundwater table.

Voorhoeve and van de Kerk (2003) reported that an excess of water of 25% was found to arrive at a STP. In one area it was more than 55%. More infiltration occurred during winter, when groundwater tables are 0.2-1.0 m higher than during summer. Therefore, during summer, there could be more exfiltration.

Darwinkel (1995) studied effects of leaking sewage pipes and overflow on soil and groundwater quality in the province of Utrecht and showed that 90% of the studied sewage pipes that were located above the groundwater table and installed before 1960 were leaking. This would imply that about 5% of the Dutch sewerage system is leaking as, in the Netherlands, 40% of sewers are located above the groundwater table and 14% were installed before 1960.

Of the pipes that were installed before 1960, leakage rates under dry weather conditions were $8.6-130\ m^3\ day^{-1}\ km^{-1}$ and under wet weather conditions $17-470\ m^3\ day^{-1}\ km^{-1}$. The report of Darwinkel (1995) also refers to a study in the province of Overijssel, where leakage rates of 69 to $190\ m^3\ day^{-1}\ km^{-1}$ were observed. Similarly, in Hannover, Germany, it was found that sewage pipes located above the groundwater table were leaking sewage at a rate of $17-30\ m^3\ day^{-1}\ km^{-1}$ (Härig & Mull 1992).

By means of video inspection, Darwinkel (1995) found that the number of damages per km was 120 for pipes installed before 1950 and 40 for pipes installed between 1950 and 1960. Such damages were axial and angular displacements, in-growth of roots, etc. This implies that a leakage could occur every 8 to 25 m. From these observations it follows that leakage rates may vary from $0.07-4\ m^3\ day^{-1}$ at a single leakage point, if leakages occur every 8 m and $0.2-12\ m^3\ day^{-1}$ if leakages occur every 25 m.

Size of virus particles, d_p [m]

Bacteriophage MS2 was chosen as a model virus. MS2 attaches to sand grains as much as or less than the

pathogenic enteroviruses (Schijven 2001). The size of MS2 is 26 nm (Penrod *et al.* 1996). Most enteroviruses and noroviruses have a similar size as MS2. Therefore, a uniform distribution between 20 and 30 nm was assumed.

Virus inactivation rate coefficient, μ_1 [day^{-1}]

Viruses differ in stability. A major factor determining the rate of virus inactivation is temperature, but this temperature dependence is virus type specific (Schijven & Hassanizadeh 2000). Pedley *et al.* (2004) have compiled all available estimates of inactivation rate coefficients for bacteriophages and enteroviruses in groundwater from literature. This is the most recent overview on virus inactivation in groundwater. From these data, values of μ_1 were selected for the temperature range of 5–12°C. At these temperatures, the inactivation rate coefficient of many viruses lies within the range of 0.01 day^{-1} to 0.1 day^{-1} with more often lower than higher values. Thus, an average value of 0.024 day^{-1} was chosen. This value was also observed value at 12°C under anoxic conditions for MS2 (Schijven *et al.* 2000). Thus, a lognormal distribution with average 0.024 and variation 0.5 was assumed. This reflects variability in the inactivation rate of viruses, but was considered as uncertainty, because the inactivation rate of a certain virus in sewage is unknown.

Virus infectivity, p_m

Data on the infectivity of enteroviruses were obtained from dose response data for rotavirus (Teunis & Havelaar 2000). An approximation of the Beta Poisson dose response model for low doses was chosen to reflect variability in virus infectivity, where $p_m = \frac{a}{a+b}$ with a and b being the parameters of a beta distribution. Their Monte Carlo sample defines its uncertainty given the available (volunteer) data.

Aquifer-specific model parameters

The same selection of unconfined sandy aquifers was used as in Schijven and Hassanizadeh (2002) and also numbered Aq1 to Aq6. These aquifers were considered as being vulnerable to virus contamination because they are shallow

and therefore have the highest probability of being near a leaking sewage pipe and because they are highly permeable.

Porosity, n

The porosity n may vary spatially. The range of 0.25–0.50 for sand given by Freeze (1979) was applied and n was assumed to be uniformly distributed within this range (worst case assumption). This was applied to all the selected aquifers.

Aquifer thickness, h [m]

The aquifer thickness is an estimate of the mean value. An uncertainty of ± 5 m was assumed with a uniform distribution due to the lack of more detailed information.

Abstraction rate, Q [$\text{m}^3 \text{day}^{-1}$]

The abstraction rate is commonly specified and, thus, there is no uncertainty. However, it varies temporally. Effects of daily variation on virus transport over more than 60 days were assumed to be negligible. Maximum abstraction rates can be twice the monthly average. This variation was included in the sensitivity analysis.

Groundwater temperature, t [°C]

In The Netherlands, the groundwater temperature in the saturated zone is generally assumed to be around 10°C, but in the top two meters of soils there is some variation due to weather changes (Tiktak *et al.* 1994). Data on groundwater temperature were obtained from the REWAB-database (REWAB 2000). Assuming that seasonal changes can be approximated by a triangular wave, so that each temperature within a range occurs at the same frequency, it was assumed that the temperature was uniformly distributed. The temperature variation is very narrow.

Grain size, d_c [m]

The grain size was assumed to be lognormally distributed based on measurements in dune sand (Schijven *et al.* 1999). For the selected aquifers, average grain sizes were either 0.25 or 0.50 mm. The sand of the selected aquifers was assumed to be uniform and well sorted. According to the

Unified Soil Classification System (Freeze 1979), sand is uniform if the uniformity coefficient, $d_{c,60}/d_{c,10}$, is smaller than 4 and well sorted if the coefficient of curvature $(d_{c,30})^2/(d_{c,10}d_{c,60})$ lies between 1 and 3. Therefore, lognormal distributions were drawn that represent uniform well sorted sands with average values of either 0.25 or 0.50 mm.

pH and collision efficiency α (attachment)

Data for groundwater pH were obtained from the REWAB-database (REWAB 2000) and were normally distributed. The collision efficiency α depends on pH. At higher pH, electrostatic repulsion between the surfaces of the virus particles and the sand grains is stronger, which is represented by a lower value of the collision efficiency. Using data from column experiments of Bales *et al.* (1991, 1993), Kinoshita *et al.* (1993) and Penrod *et al.* (1996) an empirical relation was derived by Schijven and Hassanizadeh (2002). Within the pH range of 3.5 to 7, α decreases by a factor of 0.9 for each 0.1 increase in pH:

$$\alpha = \alpha_0 0.9^{\left(\frac{\text{pH} - \text{pH}_0}{0.1}\right)} \quad (12)$$

here $\alpha_0 = 1.5 \times 10^{-5}$, which is the reference value of the collision efficiency at the reference $\text{pH}_0 = 6.8$. These reference values were found for MS2 in a deep well injection study (Schijven *et al.* 2000). The value of α_0 may be considered as a very low, conservative value for attachment of a virus. This was the case in the anaerobic part of the aquifer in the study by Schijven *et al.* (2000). In the anaerobic part of the aquifer, no iron oxyhydroxides were present, and therefore few sites for attachment were present. Similarly low values of α_0 may apply when high concentrations of dissolved organic matter are present that block sites for attachment (Schijven & Hassanizadeh 2002).

Sensitivity analysis

Sensitivity analysis was carried out for the aquifer type Aq1 to determine the effect of the model parameters on the risk of infection and the size of the protection zone. It was assumed that the model parameters were independent. A number of parameters are known to be related to each other, but this relationship was considered not to be important. For example, inactivation and diffusion of virus particles are

temperature dependent. However, in our case, groundwater temperature does not change more than $\pm 0.5^\circ\text{C}$, so an effect of temperature on the inactivation rate could be neglected. The effect of the value of the collision efficiency was evaluated over a large range, independent of pH.

The effect of the value of a model parameter was determined at the 95 percentile of the distance from the source of contamination to the abstraction well, R_{95} , where the 10^{-4} -risk of infection is not reached. This was calculated by varying a model parameter (within a certain range), including the uncertainties of the other parameters. The range wherein each parameter was varied was the 95% uncertainty interval, except for α , Q and E . For these latter ones, specific values were chosen. In addition, for the most sensitive parameters, 95-percentiles of distance R_{95} and travel time T_{95} of the protection zone at $p_{inf} = 10^{-4} \text{p}^{-1} \text{y}^{-1}$ were calculated for a number of discrete values of that parameter.

Protection zones based on enterovirus concentrations were compared with those based on norovirus concentrations, assuming that noroviruses attach as poorly and are as stable as enteroviruses. This is a conservative estimate for attachment. Noroviruses attach more than bacteriophage MS2 (Redman *et al.* 1997). Also, noroviruses were assumed to be as infective as rotaviruses, which was close to what was described (Lindesmith *et al.* 2003).

RESULTS

Model parameters

Tables 1 and 2 list average values and 95%-intervals (uncertainties) for the distributions of the model parameters from the Monte Carlo simulations.

Norovirus concentrations are on average 3500 times higher than enterovirus concentrations in raw sewage. Outliers higher than 2000 enteroviruses per litre were deleted, because such high values may put too much weight on the estimated risk of infection, whereas such high concentrations probably do not occur. A better monitoring programme would be needed to know whether such high outliers really occur.

The thicknesses of all aquifers lie within 15 and 35 m and all temperatures are approximately 10°C . Also, the

variation in pH is small, therefore all collision efficiencies are within the range of 8.0×10^{-6} and 1.1×10^{-5} .

Protection zone of shallow unconfined aquifers

Figure 1 shows attachment, inactivation and total removal of enteroviruses between the source of contamination and abstraction well for Aq1. Uncertainty increases with the distance. Removal by inactivation and dilution are the most important removal processes under the current assumption of little attachment.

Figure 2 shows the virus concentration at the well and the risk of infection, both as a function of distance and travel time. From this it follows that, for 60 days of travel time from a leaking sewage pipe, the risk of infection would almost be equal to one.

Regli *et al.* (1991) defined maximum allowable concentrations in drinking water on the basis of a risk of infection of $10^{-4} \text{p}^{-1} \text{y}^{-1}$ and a consumption of 2 litres of unboiled drinking water per person per day. In combination with the infectivity of the rotavirus, this amounts to a maximum allowable concentration of 1.8×10^{-7} per litre. However, the estimation of drinking water consumption in The Netherlands (Teunis & Havelaar 2000) is almost a factor 10 lower, therefore the virus concentration may approximately be a factor 10 higher. The current estimate is on average 2.6×10^{-6} per litre. For all six aquifers, average C_A lies between 1.8×10^{-6} and 3.3×10^{-6} .

Table 3 presents the protection zones expressed in distance and travel time, and the removal for aquifers Aq1 to Aq6. The 95-percentiles of the distances R_{95} are 12%–20% larger than the distances estimated on the basis of point estimates in the study by Schijven and Hassanizadeh (2002). The 95-percentiles of the travel times T_{95} are 40% to 60% longer than those from the point estimates by Schijven and Hassanizadeh (2002). At T_{95} an average $7.5\text{--}7.9 \log_{10}$ removal appeared to be needed instead of $9 \log_{10}$ (Schijven & Hassanizadeh 2002). So, now by including uncertainties, 95 percentiles of travel time could be estimated and these were found to be 334–676 days (6 to 12 times longer than 60 days) in order to comply with $p_{inf} = 10^4 \text{p}^{-1} \text{y}^{-1}$ with 95% certainty.

Dilution provides a large contribution to the total removal (41%–51%) and its effect becomes larger if the abstraction rate increases. Inactivation contributes

25%–43% of the total removal and is relatively higher at low abstraction rates. In that case, travel time is longer, allowing more time for inactivation. The smallest contribution to removal is due to attachment (14%–23%). The relatively higher contribution by attachment for Aq5 and Aq6 is due to the smaller grain size of the sand, as colloid-filtration theory predicts (Yao *et al.* 1971).

Sensitivity analysis

Table 4 gives the sensitivity of p_{inf} for the model parameters. The change in p_{inf} due to the change of model parameter x from x_{min} to x_{max} is expressed as $\text{Log}_{10} \left(\frac{p_{inf}(x_{max})}{p_{inf}(x_{min})} \right)$. In all cases p_{inf} was either ascending or descending monotonically between x_{min} and x_{max} . Table 5 shows R_{95} and T_{95} (at $p_{inf} = 10^{-4} \text{p}^{-1} \text{y}^{-1}$) for a number of discrete values of the most sensitive model parameters and Table 6 shows this for a combination of aquifer thickness and abstraction rate values.

The most sensitive parameter is attachment (α). In the present study, a very conservative value for α was assumed. This was the very low value for bacteriophage MS2 under field conditions where little attachment took place (Schijven *et al.* 2000). The presence of more sites for attachment could lead to a strong reduction of the risk of infection. Even in the case of a low value of α of 10^{-4} , the risk of infection would be 3.5 orders in magnitude smaller and the protection zone would be almost three times smaller compared to the case of a value of α of 10^{-5} .

Schijven (2001) calculated α values from a number of published field studies. In almost all cases, virus removal was higher during the first meters of soil passage than in the following meters. Schijven *et al.* (2000, 2002) demonstrated that in this part of the aquifer more sites for attachment were presented in the form of ferric oxyhydroxides. Therefore, two α values were estimated: High α values for the transport within the first meters, in the order of 10^{-3} – 10^{-2} , and low α values, for the following meters of transport, lying between 10^{-5} and 10^{-3} . Table 5 shows that at $\alpha = 10^{-3}$ the required travel time becomes 29 days.

The second most sensitive model parameter is the virus inactivation rate coefficient. When the inactivation rate approximates 0.1 day^{-1} , the risk of infection is about 12 orders in magnitude smaller than with an inactivation rate of 0.024 day^{-1} . From Table 5 it can also be seen that 29 days

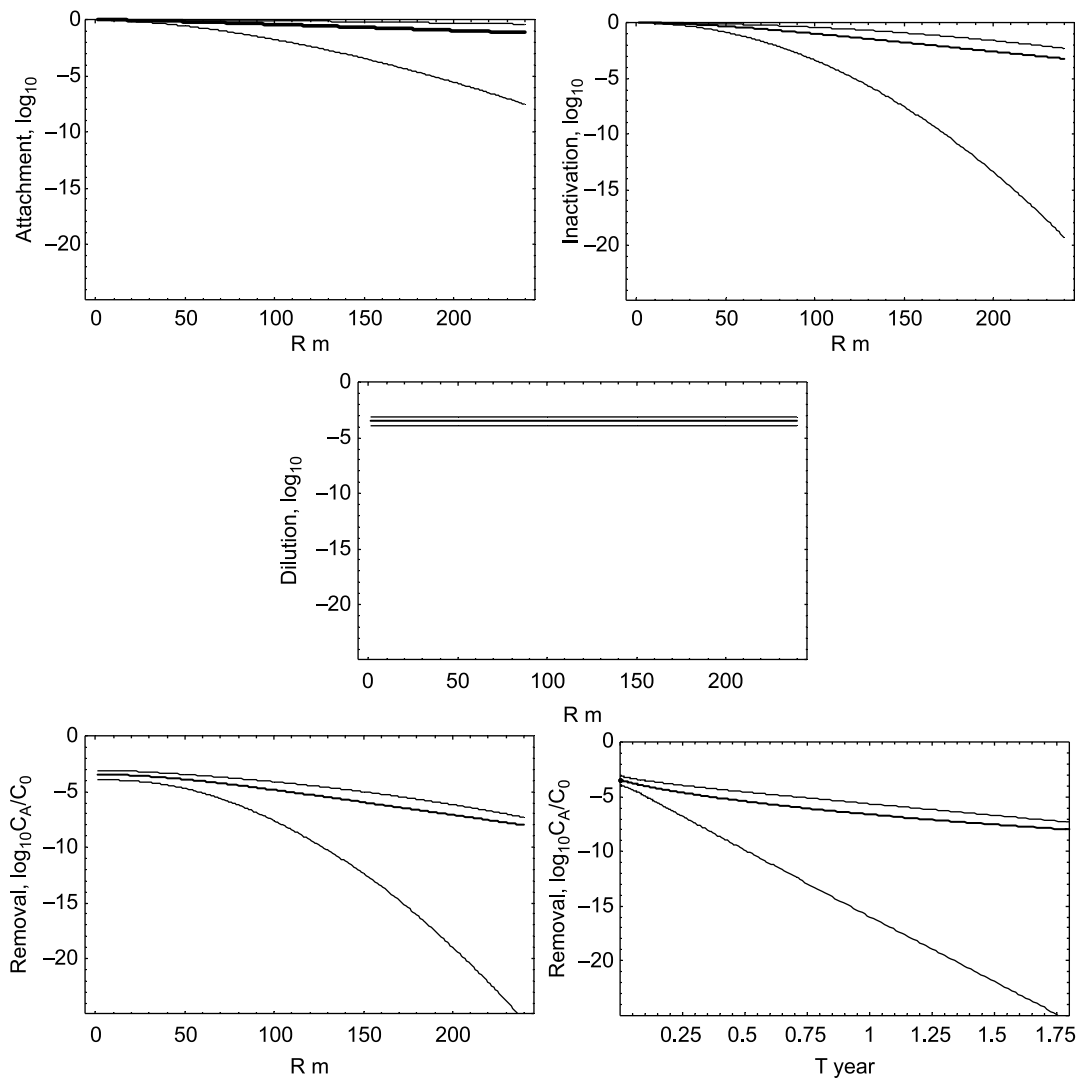


Figure 1 | Attachment, inactivation and dilution of enteroviruses as a function of distance. Total removal as a function of distance and travel time. The thick line is the average and the thin lines indicate the 95%-interval (uncertainty).

of travel time would be sufficient only if the inactivation rate coefficient was as high as 0.4 day^{-1} . However, most viruses are much more stable.

Grain size d_c is also a sensitive parameter. Its effect on attachment was already clear for Aq5 and Aq6 with the finer sand in comparison the other selected aquifers. In very fine sand (0.1 mm), the risk of infection is four orders in magnitude smaller than in coarse sand (1 mm). The difference in size of the protection zone is about a factor 1.5 and the difference in travel time almost a factor two.

Also the virus concentration in sewage strongly affects the size of the protection zone. Table 5 includes the estimates

based on noroviruses. Because these concentrations in sewage are three orders in magnitude higher than the enterovirus concentrations, the protection zone increases 1.5 times in distance and 2 times in travel time.

The sensitivity to the consumed volume of unboiled drinking water is about the same as for the virus concentration in sewage. This is because the ranges of these parameters extend over similar orders in magnitude and are similarly proportional to the risk of infection.

Leakage rate, pH, recovery efficiency, virus infectivity, porosity, virus size and temperature have a limited effect on the dimensions of the protection zone within the ranges in

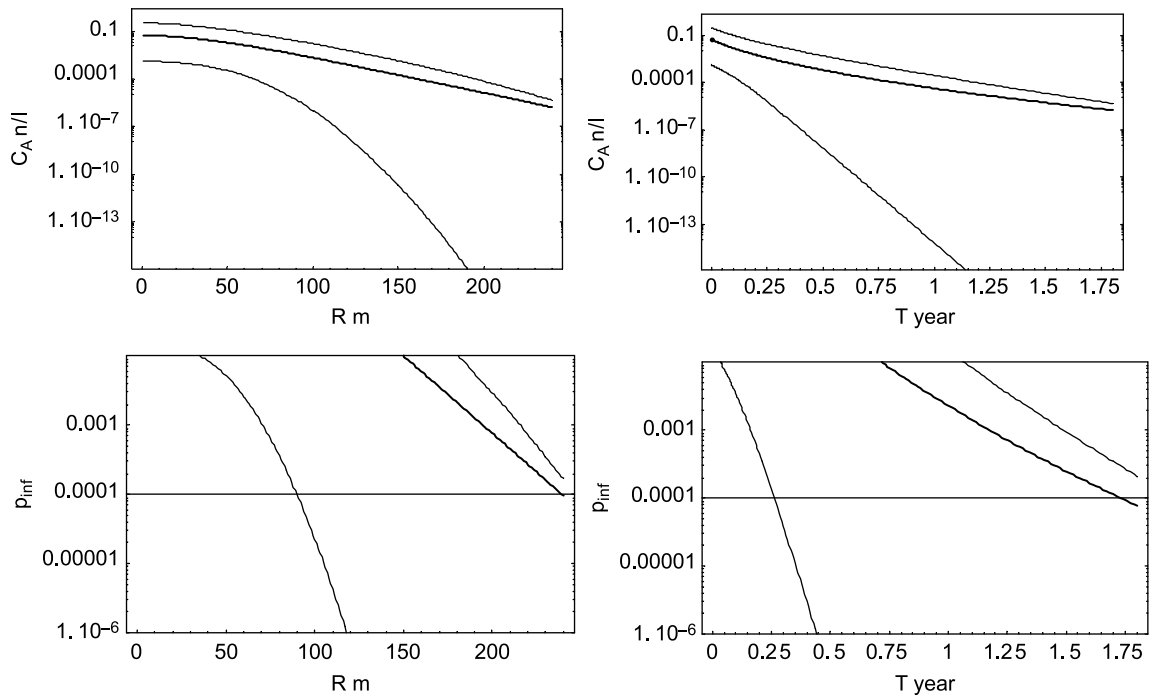


Figure 2 | Virus concentration C_A at the abstraction well and risk of infection p_{inf} as a function of distance and travel time for Aq1. The thick lines are the averages and the thin lines the 95%-intervals.

which they could vary. It must be noted that the leakage rate is very uncertain and could therefore vary over a larger range than applied here. Temperature and pH are known to strongly determine virus inactivation rate and attachment, respectively. However, here, temperature and pH changes are very small, and are therefore relatively unimportant. The effect of these small temperature changes is negligible.

The sensitivity to changes in aquifer thickness h and abstraction rate Q are shown in Table 6. For the smallest aquifer thickness, the required distance and travel time increase the strongest with increasing abstraction rate. For the combinations of aquifer thickness and abstraction rate that can occur in actual situations, the required distance and travel time of the protection zone lie between 151 m and 457 m, respectively 0.7 year and 7 years, which is a large range.

CONCLUSIONS AND DISCUSSION

Protection zones of shallow unconfined aquifers in The Netherlands were calculated that allow protection against virus contamination to the level that the infection risk of 10^{-4} per person per year is not exceeded with a 95% certainty.

An uncertainty and a sensitivity analysis of the calculated protection zones were included.

The following conclusions could be drawn:

- Including all parameter uncertainties, it can be concluded that, for shallow unconfined sandy aquifers, protection zones are needed with travel times of 1 to 2 years (206–418 m). This is 6 to 12 times 60 days. In that case the risk of infection of $10^{-4} \text{ p}^{-1} \text{ y}^{-1}$ will not be exceeded with 95% certainty.
- Under the condition of little attachment, as was assumed here, dilution contributes the most to the reduction of virus concentrations, followed by inactivation and then attachment.
- According to the sensitivity analysis, virus attachment and virus inactivation are the most sensitive parameters. Next most sensitive parameters, from high to low, are grain size of the sand, the abstraction rate at the well, virus concentrations in raw sewage, and unboiled drinking water consumption.
- Also, a conclusion can be drawn on the vulnerability of aquifers below confining layers or aquitards. In these cases, vertical flow from the leaking sewage pipe to these

Table 3 | Estimated distance (R) and travel time (T) at $p_{inf} = 10^{-4} \text{ p}^{-1} \text{ y}^{-1}$, attachment, inactivation, dilution and total removal at R_{95} for aquifers Aq1 - Aq6. Removal by attachment, dilution and inactivation are also expressed as percentages of total removal

			Aq1	Aq2	Aq3	Aq4	Aq5	Aq6
Distance	[m]	R_{avg}	157	140	125	280	213	179
		R_{95}	232	206	183	418	324	271
		95%-interval	91–252	81–221	73–198	158–454	107–352	47–145
Travel time	[days]	T_{avg}	295	336	322	234	151	173
		[year]	0.8	0.9	0.9	0.65	0.41	0.48
	[days]	T_{95}	605	676	639	482	333	373
		[year]	1.7	1.9	1.8	1.3	0.9	1.0
	[days]	95%-interval	90–702	108–776	102–744	68–568	31–391	39–439
		[year]	0.25–1.9	0.30–2.1	0.28–2.1	0.19–1.6	0.085–1.1	0.11–1.2
Attachment	[$-\log_{10}$]	Average	1.1 (14%)	0.99 (13%)	1.2 (16%)	1.2 (15%)	1.8 (23%)	1.8 (23%)
		95%-interval	0.36–7.0	0.31–5.6	0.39–7.6	0.44–8.1	0.81–16	0.86–16
Inactivation	[$-\log_{10}$]	Average	3.1 (40%)	3.3 (43%)	3.2 (43%)	2.7 (34%)	2.0 (25%)	2.2 (28%)
		95%-interval	2.1–18	2.4–21	2.2–20	1.7–16	1.1–9.7	1.3–11
Dilution	[$-\log_{10}$]	Average	3.5 (45%)	3.3 (43%)	3.1 (41%)	3.9 (49%)	4.0 (51%)	3.7 (47%)
		95%-interval	3.1–3.9	2.8–3.7	2.7–3.6	3.5–4.3	3.6–4.4	3.2–4.1
Removal	[$-\log_{10}$]	Average	7.7 (100%)	7.7 (100%)	7.5 (100%)	7.9 (100%)	7.9 (100%)	7.8 (100%)
		95%-interval	7.0–24	7.0–26	6.8–25	7.1–22	7.1–24	7.1–24

aquifers needs to be considered. Vertical groundwater flow under these conditions has been studied by Meinardi (1994). The long-term average precipitation in a sandy area amounts to 0.80 m per year and the long-term average evapotranspiration 0.55 m per year, implying a net recharge of 0.25 m per year. At a porosity of 0.35 this implies vertical flow of 0.7 m per year. As was shown before, 2 years of travel time (considering dilution and inactivation) would provide adequate protection of the groundwater against virus contamination. Therefore, this type of aquifers is adequately protected.

It is recommended that drinking water companies calculate protection zones for shallow unconfined sandy aquifers

according to the method and data given in this paper. This will imply a considerable enlargement of protection zones from 60 days of travel time to 1.0 to 1.9 years. This encompasses about 110 unconfined groundwater well systems that produce $3 \times 10^8 \text{ m}^3 \text{ y}^{-1}$ of drinking water, which is 38% of the total drinking water production from groundwater in The Netherlands.

However, a smaller protection zone could be applied if it can be demonstrated, that the aquifer has such properties that the risk of virus contamination is reduced more than according to the method presented in this paper. This may be the case if there are more attachment sites present or, possibly, due to enhanced removal during transport through an unsaturated zone.

Table 4 | Sensitivity of p_{inf} for the model parameters

Model parameter, x^a	x_{min}	x_{max}	$p_{inf}(x_{min})$	$p_{inf}(x_{max})$	$Log_{10}\left(\frac{p_{inf}(x_{max})}{p_{inf}(x_{min})}\right)$
α	1.0×10^{-5}	1.0×10^{-3}	1.5×10^{-4}	1.1×10^{-20}	-16
α	1.0×10^{-5}	1.0×10^{-4}	1.5×10^{-4}	5.0×10^{-8}	-3.5
μ_t [day ⁻¹]	0.0089	0.064	1.7×10^{-3}	2.1×10^{-13}	-9.9
d_c [m]	2.3×10^{-4}	1.1×10^{-3}	7.8×10^{-8}	8.8×10^{-4}	4.1
Q [m ³ day ⁻¹]	1.0×10^3	1.0×10^4	2.6×10^{-6}	2.3×10^{-3}	2.9
C_0 [n l ⁻¹]	5.3	810	5.6×10^{-6}	8.6×10^{-4}	2.2
V [l p ⁻¹ y ⁻¹]	6.3	480	8.2×10^{-6}	6.1×10^{-4}	1.9
h [m]	25	35	3.1×10^{-4}	4.4×10^{-5}	-0.84
q [m ³ day ⁻¹]	0.38	2.6	5.7×10^{-5}	3.4×10^{-4}	0.78
pH	6.8	7.6	5.8×10^{-5}	3.3×10^{-4}	0.75
E	0.1	1.0	1.6×10^{-3}	3.8×10^{-4}	-0.63
p_m	0.26	0.87	7.0×10^{-5}	2.3×10^{-4}	0.52
n	0.26	0.49	2.7×10^{-4}	8.6×10^{-5}	-0.50
d_p [m]	2.0×10^{-8}	3.0×10^{-8}	1.2×10^{-4}	2.1×10^{-4}	0.23
t °C	10	11	1.6×10^{-4}	1.6×10^{-4}	-0.019

^aThe model parameter range x_{min} to x_{max} corresponds to the 95% uncertainty interval (see Tables 1 and 2), except for α , Q and E .

It was assumed that few sites for attachment were available under the commonly anoxic conditions (Schijven *et al.* 2000). The uncertainty about the presence of attachment sites is large and has a large effect on the size of the protection zone. When there are more attachment sites, like ferric oxyhydroxides, a much smaller protection zone would be needed. Values of α in the order of 10^{-3} were found for the first meters of dune recharge (Schijven *et al.* 1999) and of deep well injection (Schijven *et al.* 2000). At such values, a travel time of 60 days could provide enough protection. Although virus removal from artificially recharged groundwater, as in dune recharge and deep well injection has been studied extensively, removal of viruses in natural groundwater in a sandy aquifer in The Netherlands has not been studied yet. Therefore, the presence of attachment sites in that kind of situation is not actually

known. Probably, because commonly the abstracted water is anaerobic, conditions are reducing and only a few sites for attachment are expected to be present, as was the case in the anoxic part of the aquifer of the deep well injection study (Schijven *et al.*, 2000). However, because of the strong effect that attachment sites can have on the size of the protection zone it is worthwhile investigating their presence. This could be done at a number of representative sites by investigating geochemical conditions, including heterogeneity.

An important factor that affects the attachment of viruses is the presence of high concentrations of bonded and dissolved organic matter, which is the case when sewage is present. Commonly, organic matter prevents attachment of viruses to the sand grains because organic matter may occupy the potential attachment sites. In the

Table 5 | Distance R_{95} and travel time T_{95} at $p_{inf} = 10^{-4} \text{ p}^{-1} \text{ y}^{-1}$ for a number of values of the most sensitive model parameters

Parameter	Value	R_{95} [m]	T_{95} [day]	T_{95} [year]
α	10^{-5}	231	603	1.7
	10^{-4}	132	215	0.6
	10^{-3}	47	29	0.30
μ_l [day $^{-1}$]	0.01	280	859	2.4
	0.1	105	109	0.30
	0.4	55	29	0.08
d_c [mm]	0.1	181	392	1.1
	0.2	214	537	1.5
	0.5	227	590	1.6
	1.0	244	657	1.8
	2.0	258	729	2.0
C_o [n/l]	10^2	237	594	1.7
	10^3	266	746	2.1
	10^4	292	880	2.4
	10^5	316	1049	2.9
	10^6	338	1199	3.3
C_o [n/l], norovirus	5.3×10^5 ($5.6 \times 10^3 - 3.6 \times 10^6$)	313	1098	3.0
V [l p $^{-1}$ y $^{-1}$]	100	234	1246	1.7
	200	243	1338	1.9
	500	254	1465	2.0
	1000	262	1546	2.1

applied model this was represented by the low α of 10^{-5} . However, depending on the nature of organic matter, it may also cause viruses to attach through hydrophobic interaction (Schijven & Hassanizadeh 2000).

After attachment, the inactivation rate coefficient was found to be the most sensitive parameter. Variability between inactivation rates of viruses may be large, and for a given sample of sewage it will be uncertain what type of

virus is actually present. The applied distribution for the inactivation rate coefficient was, therefore, relatively conservative.

Norovirus concentrations in sewage were found to be 3500 times higher than enterovirus concentrations. However, it is unknown what part of the RT-PCR determined noroviruses are infectious virus particles. If noroviruses indeed are so much higher in concentration and as

Table 6 | Distance R_{95} and travel time T_{95} at $p_{inf} = 10^{-4} \text{ p}^{-1} \text{ y}^{-1}$ for a number of values of aquifer thickness h and abstraction rate Q

h [m]		Q [$\text{m}^3 \text{day}^{-1}$]			
		1000	2000	5000	10000
20	R_{95} [m]	174	235	343	457
	T_{95} [day]	332	610	1321	2327
	T_{95} [year]	0.9	1.7	3.6	7
50	R_{95} [m]		151	222	149
	T_{95} [day]		252	552	978
	T_{95} [year]		0.7	1.5	2.7
100	R_{95} [m]			160	108
	T_{95} [day]			283	507
	T_{95} [year]			0.8	1.4

infectious as enteroviruses, and attach just as poorly and survive equally well, then the required travel time would need to be a factor of two longer. Thus, further research should be focussed on assessing the fraction of infectious noroviruses under a range of environmental conditions.

The leakage rate is very uncertain. Data is lacking for the rate at a given location, on the number and location of leakages as well as the location relative to the groundwater table. Clogging of the pores in the aquifer near a leakage by solids from the sewage will also have a strong effect on the leakage rate.

Diffuse contamination sources like manure on agricultural land are important too. Manure may contain viruses that are zoonotic, in other words are infectious to both humans and animals, such as hepatitis E viruses (Van der Poel *et al.* 2001).

In the cases of leaking sewage pipes above the groundwater table and diffuse contamination sources like manure, transport of viruses through the unsaturated zone comes into play (in The Netherlands commonly 1–2 m). In that case, attachment and inactivation may possibly be enhanced (Schijven & Hassanizadeh 2000), providing additional protection. The presence of high concentrations

of organic matter from sewage or manure is expected to play an important role too. Under unsaturated conditions bacteria and parasites, like the persistent oocysts of *Cryptosporidium* may also be of relevance.

Grain size appeared to be of significance too. For a specific location, uncertainties are easily reduced by taking sand samples for analysis of the grain size distribution, but physical heterogeneity needs to be taken into account and a representative number of soil samples will be needed.

The present study has increased the insight into the factors that determine the size of protection zones against virus contamination and what relevant data are to be collected to reduce uncertainties of the most important factors.

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