

A strategy for sampling reactive aquifer sediments in drinking water well fields

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Abstract The modelling of solute transport around drinking water well fields requires information on the hydrogeochemical reactivity of the aquifer sediments. This paper describes a sampling strategy that was developed to collect reactivity data for transport modelling in phreatic well fields (PWF) and deep-well recharge systems (DWR). The scheme accounts for the hydraulic and chemical heterogeneity of the aquifer sediments. Different sampling objectives were stated for phreatic well fields and deep-well recharge systems to account for the propagation of reaction fronts relative to the main direction of geochemical variation. The sampling strategy includes two sampling stages using systematic and stratified random sampling. Sampling volumes and the use of geochemical analysis methods are discussed briefly. The approach is illustrated, calculating the breakthrough of solutes for a simple DWR case in the geochemically layered deposits of the Oostrum aquifer (The Netherlands).

Key words aquifer reactivity; geochemical and hydraulic heterogeneity; sampling strategy; solute transport modelling

INTRODUCTION

Solute transport models are becoming widely used for the prediction of the evolution of groundwater composition in aquifers used for drinking water production (Griffioen *et al.*, 1998). The models require input on the hydrogeochemical reactivity of the aquifer. A sampling strategy should yield a selection of aquifer samples that represents the reactive subsurface properties at the relevant spatial scale of a drinking water production site. It should account for the hydraulic and the chemical heterogeneity of the aquifer sediments, since they may have serious implications for the predicted breakthrough of solutes. This paper describes a sampling strategy that was developed to collect reactivity data for use in transport modelling in phreatic well fields and deep-well recharge systems in The Netherlands.

EFFECTS OF GEOCHEMICAL HETEROGENEITY ON SOLUTE BREAKTHROUGH

The solute breakthrough in a geochemical heterogeneous system depends on the direction of solute transport relative to the main direction of reactive geochemical variation. This is illustrated for an aquifer volume containing an average content of 100 mmol l^{-1} reactive sediment phase which is flushed with 1 mmol l^{-1} reactive solute

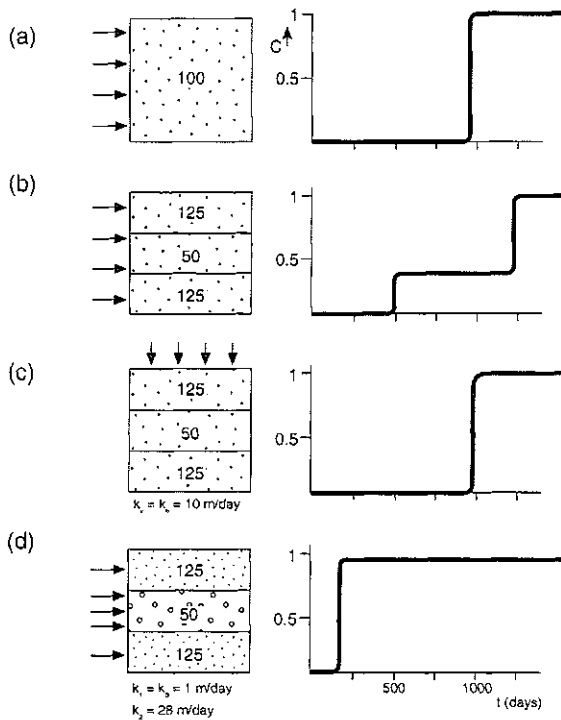


Fig. 1 Solute breakthrough in layered reactive systems: (a) homogeneous distribution of reactive phase; (b) geochemically layered, parallel flow; (c) geochemically layered, perpendicular flow; (d) geochemically and hydraulically layered, parallel flow. Dimensions: $\Delta x = \Delta y = 3$ m, hydraulic gradient 0.01, porosity 0.33, initial solute concentration 1 mmol l^{-1} . Reactive solid phase concentrations are expressed in mmol per litre pore water.

(Fig. 1). For a homogeneously distributed reactive phase the solute will break through after 100 pore volumes, assuming instantaneous equilibrium and reaction stoichiometry of 1:1 (Fig. 1(a)). The breakthrough in a geochemically stratified aquifer volume depends on the flow direction. If groundwater flow is perpendicular to the geochemical stratification, the breakthrough is similar to the homogeneous case (Fig. 1(c)). For parallel flow relative to the geochemical stratification, the reactive phase is depleted earlier in the more permeable middle section than in the less permeable upper and lower parts of the aquifer volume (Fig. 1(b)). Thus, for perpendicular flow an estimate of the average reactive solid phase content satisfies the information needs. For parallel flow the vertical variation of reactive properties should be quantified to predict the solute breakthrough. For parallel flow the hydraulic heterogeneity becomes important as well (Fig. 1(d)). Preferential flow through the less reactive parts of the aquifer volume results in earlier breakthrough compared with the hydraulic homogeneous cases.

MODEL CONCEPTS AND SAMPLING OBJECTIVES

The reactivity data to be sampled will be used for the transport modelling in the unconsolidated aquifers of The Netherlands. At the scale of well fields we normally

assume a vertical variation of reactive properties, which is related to the main horizontal layering of the sedimentological facies. Lateral variations in hydraulic and geochemical properties within the layers are neglected in the model schematization. The proposed sampling strategy should yield input data for these kinds of models. Therefore, a concept of geochemical and hydraulic layering was used to define the sampling objectives. Different objectives were adopted for phreatic well fields and deep-well recharge systems, to account for the different directions of groundwater transport relative to the geochemical and hydraulic stratification.

Phreatic well fields

For a PWF the intention is to predict the propagation of reaction fronts from diffuse pollution sources, such as pesticides and nutrients. The groundwater recharge, the aquifer thickness and the porosity determine the groundwater travel times and the vertical position of an advective pollution front (Raats, 1981). The pollution fronts tend to move downward, perpendicular to the reactive geochemical layering (following Fig. 1(c)). The shifts of the pollution fronts can be measured in the vertical, assuming homogeneous land use in the upstream area (Appelo & Postma, 1993). Therefore, the chosen sampling objective is to determine the depth and thickness of the reactive subsurface layers, and the average content of the reactive components in those layers.

Deep-well recharge systems

In a DWR or Aquifer Storage and Recovery system (ASR) groundwater transport is merely horizontal, parallel to the assumed reactive geochemical layering. The breakthrough of solutes could proceed differently at different depths, due to vertical variations in the reactive aquifer properties (Fig. 1(b)). Vertical variations in hydraulic properties could even have a larger impact on the breakthrough (Fig. 1(d)). For these systems, the sampling should yield quantitative information on the vertical variation of hydraulic and geochemical properties. Average values are not sufficient to predict solute breakthrough.

SAMPLING STAGES

Two sampling stages are proposed for both systems studied. The first reconnaissance stage consists of systematic sampling of selected boreholes. Systematic sampling, using a fixed vertical sampling distance, is preferred over random sampling because the depth of the reaction zone and the thickness of the recharge aquifer are not yet known at this stage. For PWF, the sampling focuses on the depth of the expected reactive zone. The sampling yields information on the sedimentological facies, the position of the reactive layers and a first indication of the average content (\bar{x}) of the major reactive layer. A first indication of the frequency distribution (estimated percentiles) is also acquired for DWR. Figure 2 illustrates the two subsequent stages of the sampling strategy for a DWR system. The second sampling phase is proposed to

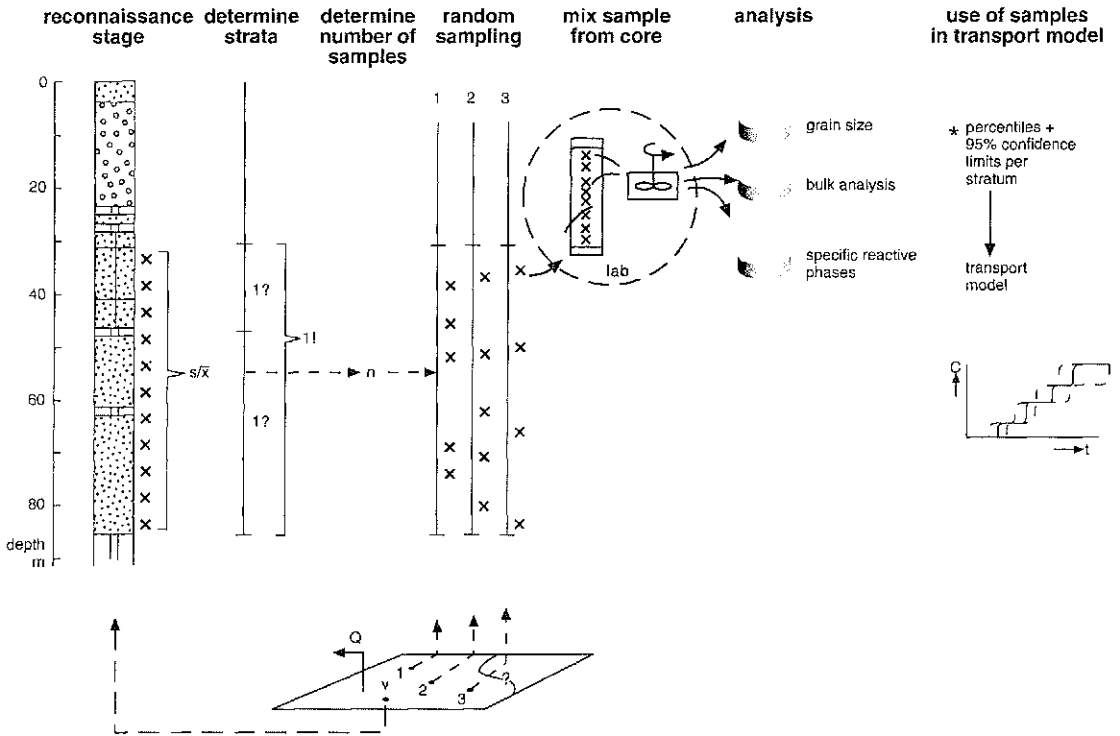


Fig. 2 The subsequent stages of the proposed sampling strategy for a DWR system.

quantify the uncertainty of the estimates of average values (PWF) or percentiles (DWR). Stratified random sampling is recommended for the second stage to collect transport model input data from the main lithological strata. The stratified random sampling is preferably done from a number of boreholes to ensure that the estimates are spatially representative (Fig. 2). The number of samples is adapted to the level of variation (s/\bar{x} , where s = standard deviation) measured during the reconnaissance stage. In general, a smaller sample size is required to attain accurate estimates of \bar{x} than to attain accurate estimates of the percentiles (for details see Broers, 1999).

SAMPLING METHODS AND SAMPLE VOLUMES

Cable tool drilling is preferred for geochemical sampling since no drilling fluid is applied and water use is minimized. Thin wall tube samplers of about 0.5 m length are favoured to acquire minimally disturbed samples of the unconsolidated deposits. The sample volumes should conform to the vertical discretization scale of the solute transport model. A choice is made to acquire average values for a 0.5 m depth interval of the aquifer. These can be obtained by mixing the whole sampling device or by mixing of a number of random samples from the core (Fig. 2).

GEOCHEMICAL ANALYSIS AND GRAIN SIZE DISTRIBUTION

The sampling strategy focuses on the specific reactive properties required for the transport properties of the sediments (e.g. Griffioen & Broers, 1994). However, in the reconnaissance stage, measurements on the geochemical bulk properties of the sediments are advisable in order to determine if the specific analyses can be replaced by less expensive alternatives (Fig. 2). Low-cost bulk analysis can be obtained from X-ray spectrometry (XRF), total destruction analysis or differential heating (Huisman, 1998). Grain size distribution is measured in each sample to relate hydraulic and geochemical variations at the sample scale.

USE OF THE SAMPLES IN TRANSPORT MODELLING

Sample data from the Oostrum aquifer (Limburg, The Netherlands) were used to show how the acquired sample statistics could be employed in the transport model. The Oostrum aquifer was studied to trace the origin of heavy metals and arsenic in pumped groundwater (Broers, 1998). The sediments were sampled using thin wall tube samplers and bulk sediments and specific reactive phases were analysed. The reconnaissance stage showed two reactive strata of about 5 and 10 m thickness: Venlo Top and Venlo Sand. The strata consist of coarse fluvial sands. Sediment samples were taken from nine boreholes. Table 1 shows summary statistics for the pyrite content measured in the two strata. The Venlo Top stratum has larger pyrite contents, and larger variation and skewness.

Table 1 Summary statistics for pyrite content (g kg^{-1}) in two strata.

	<i>n</i>	Average \bar{x}	95% conf. interval of \bar{x}	coeff. of variation s/\bar{x}	P25	P50 (median)	P75
Venlo Sand	39	2.5	2.5 ± 0.6	0.75	1.0	2.1	3.0
Venlo Top	18	8.0	8.0 ± 4.5	1.16	1.3	3.9	13.1

For a PWF case, the average content of the reactive strata can be used directly as input for the transport model. The parametric 95% confidence interval boundaries (Table 1) could be used as worst-case and best-case scenarios in a sensitivity analysis. For a DWR case, we need to include the observed vertical variations in pyrite content in the transport model. Since grain size and pyrite content did not appear to be correlated in the Oostrum case, a hydraulically homogeneous subsoil was assumed. The observed variation of the pyrite content was modelled in a four-layer schematization. The model layers do not describe the physical structure of the subsurface, but represent the statistical characteristics of the aquifer. Each model layer represents one-quarter of the total variation in pyrite content (P0–P25, P25–P50, P50–P75 and P75–P100) and is characterized by the typical value of the pyrite content for that quarter. The estimated 12.5th percentile (P12.5), which is the median of the lowest 25% of the frequency distribution, characterizes the least reactive parts of the aquifer (Fig. 3(a)). Each model layer receives 25% of the horizontal flux to achieve a hydraulically homogeneous model. In this way, the four-layer model assumes that the

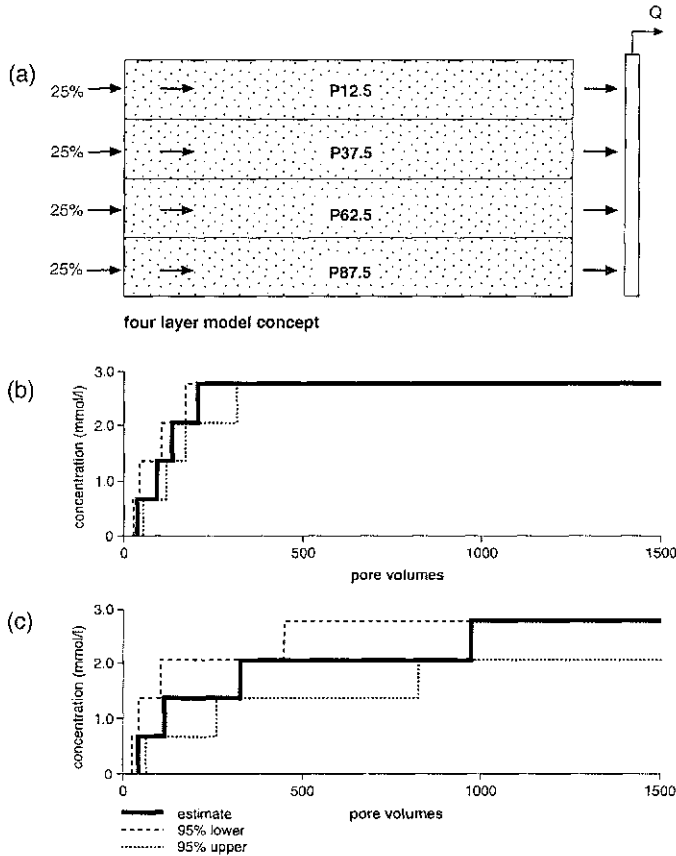


Fig. 3 Estimated breakthrough (solid line) and 95% confidence limits around the estimate (dashed lines) for a DWR system in a geochemically layered aquifer for the Venlo Sand (b) and Venlo Top (c) strata.

Table 2 Percentiles and 95% confidence limits used for four-layer transport model (contents in $g\ kg^{-1}$).

	<i>n</i>	P12.5 (95% interval)	P37.5 (95% interval)	P62.5 (95% interval)	P87.5 (95% interval)
Venlo Sand	39	0.8 (0.5↔0.9)	1.8 (0.9↔2.2)	2.5 (2.0↔3.2)	3.9 (3.2↔5.9)
Venlo Top	18	0.8 (0.4↔1.2)	2.2 (0.8↔4.9)	6.2 (2.0↔15.7)	18.4 (8.5↔31.7)

least reactive zones of the sampled sedimentary stratum are mutually connected. This represents the extreme condition where the geochemical reactivity has the largest effects on solute breakthrough. The other extreme condition is where the geochemical variation is randomly distributed within the flow domain. The estimated percentiles for both strata are listed in Table 2. The 95% confidence interval for the estimated percentiles was computed non-parametrically following Helsel & Hirsch (1992).

The four-layer model was used to calculate the breakthrough of a continuous input of $2.8\ mmol\ l^{-1}$ nitrate in the vertically heterogeneous DWR system for Venlo Sand (Fig. 3(b)) and Venlo Top (Fig. 3(c)). The velocity of the nitrate front is retarded by

the redox reaction with pyrite. The first step of the breakthrough is determined by the least reactive sublayer of the aquifer, where pyrite is exhausted after about 40 pore volumes. The first breakthrough occurs simultaneously for both strata, despite the larger overall pyrite content of Venlo Top. Ultimately, the larger pyrite content and variability of Venlo Top results in a retarded breakthrough relative to Venlo Sand. The confidence interval boundaries were used to determine upper and lower limits for the calculated breakthrough in the sensitivity analysis (Fig. 3(b, c)). The heterogeneous Venlo Top stratum exhibits a larger uncertainty on the breakthrough curve compared to the relatively homogeneous Venlo Sand stratum.

In the four-layer model a connection was assumed between the least reactive parts of the aquifer, which results in early breakthrough and tailing in a later stage. If a random distribution of pyrite in the aquifer is assumed, the average content can be used to calculate the breakthrough after 130 ± 30 and 430 ± 240 pore volumes for Venlo Sand and Venlo Top, respectively. Which model is most appropriate should be decided on the basis of the observed sedimentary structures in the sampled stratum.

A choice was made to model the Oostrum DWR case using a four-layer model. Developing models consisting of more than four layers is only feasible when accurate estimates of the percentiles can be obtained from the data. This requires a large number of samples or a stratum with little variation in reactivity.

CONCLUSIONS

The study shows that different quantitative sampling objectives are necessary for PWF and DWR systems. The examples reveal that the uncertainty of the estimated sample statistics directly effects the uncertainty of the calculated breakthrough. Hence, a proper design of the sampling strategy helps to reduce the uncertainty of the transport model results.

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