

# RIVERBANK FILTRATION IN THE NETHERLANDS: WELL FIELDS, CLOGGING AND GEOCHEMICAL REACTIONS

Pieter J. Stuyfzand<sup>1,2</sup>, Maria H.A. Juhász-Holterman<sup>3</sup> and Willem J. de Lange<sup>4</sup>

<sup>1</sup>*Free University, Dept. Hydrology & Geo-Environmental Sciences, FALW, Boelelaan 1085, 1081 HV Amsterdam, Netherlands; Tel: +31. 20.444.7968 or +31.6.10945021; Email: [pieter.stuyfzand@falw.vu.nl](mailto:pieter.stuyfzand@falw.vu.nl)*

<sup>2</sup>*Kiwa Water Research, PO Box 1072, 3430 BB Nieuwegein, Netherlands; Tel: +31. 30.6069.552 or +31.6.10945021; Email: [pieter.stuyfzand@kiwa.nl](mailto:pieter.stuyfzand@kiwa.nl)*

<sup>3</sup>*Water Supply Limburg (WML), Limburglaan 25, 6229 GA Maastricht, Netherlands*

<sup>4</sup>*Institute for Inland Water Management and Waste Water Treatment (RIZA), PO Box 17, 8200 AA Lelystad, Netherlands*

**Abstract:** River Bank Filtration (RBF) contributes ca. 7% (80 Mm<sup>3</sup>/a) to the national drinking water supply in the Netherlands, through a total of 26 well fields. These RBF well fields are classified on the basis of (1) the main driving mechanism of flow (polder or pump driven); (2) RBF periodicity (flow direction temporarily reversing or not), (3) type of riverbed (sand or gravel), and (4) type of contact of river with aquifer (with or without intercalated aquitard(s)).

Three case studies are reviewed which demonstrate the water quality, geochemical reactions and clogging phenomena in differing systems. The mass balance approach, also called 'reverse modeling', is applied to identify and quantify the extent of the most important hydrogeochemical reactions at the three case study sites.

Clogging of the river bed seems to be a problem in the Netherlands only in RBF systems with a true gravel bed such as Roosteren along the Meuse River, and on sites where sludge is strongly accumulating due to structurally reduced river flows, as in the Hollandsch Diep estuary which was dammed in 1971 as part of the Delta Works.

Key words: river bank filtration, geoclogging, Netherlands, water supply

## **1. INTRODUCTION**

Riverbank filtration (RBF), in a broad sense, is the process of river water infiltrating its riverbed. RBF *sensu stricto* is defined as the process of river water infiltrating where pumping wells (mostly for drinking water supply) create induced recharge.

Induced recharge has demonstrated in Europe, in the past 100-150 years, to be a very vital technique of collecting high quality raw water for drinking water preparation, with many advantages (Van der Kooij et al., 1986; Sontheimer, 1991; Ray et al., 2002; Jülich & Schubert, 2001).

Advantages with respect to direct surface water treatment include: elimination of suspended fines with the attached pollutants, strong reduction of quality fluctuations, and strong quality improvement by elimination of heavy metals, organic micropollutants, bacteria and viruses. Disadvantages are: (a) the impossibility of completely impeding the river to infiltrate when this would be episodically desirable because of a bad quality; (b) geochemical reactions of the infiltrate with sludge and aquifer materials that raise the concentrations of notably  $\text{Fe}^{2+}$ ,  $\text{Mn}^{2+}$ , As,  $\text{NH}_4^+$ ,  $\text{CH}_4$ ,  $\text{Ca}^{2+}$  and  $\text{HCO}_3^-$ ; and (c) risks of a cumbersome clogging of the river bed.

In this contribution, experiences from the Netherlands are presented pertaining to RBF in general, also clogging and geochemical reactions (both positive and negative). Experiences with RBF along lake banks, canals and ditches are not considered here.

## **2. RIVER BANK FILTRATION IN THE NETHERLANDS**

### **2.1 Historical Evolution**

Various hydrological manipulations forced the water of the Rhine and Meuse Rivers to increasingly infiltrate, since about 1200 AD, along their

many water courses in the fluvial and coastal plain of the Western Netherlands, as well as in former seepage areas (Stuyfzand, 1989b). The first riverbank filtrate was pumped, for public drinking water supply in the Netherlands, probably in 1879 along the Rhine River at pumping station Nijmegen (Site 42 in Fig.1). In 1950 15 well fields pumped 11 Mm<sup>3</sup> and in 1995 25 pumping stations produced 79 Mm<sup>3</sup> of Rhine bank filtrate (including admixed autochthonous groundwater). In 1998 the first Meuse bank filtrate was pumped near Roosteren (site 80 in Fig.1). An annual total of 80 Mm<sup>3</sup> of riverbank filtrate in 2002 (Fig.2) constitutes nearly 7% of the total production of drinking water in the Netherlands (1200 Mm<sup>3</sup>/a).

The deterioration of the quality of the Rhine River, especially in the period 1920-1975, had at least three impacts on the preparation of drinking water from Rhine River water: (a) a switch from the direct intake of river water to the pumping of Rhine bank filtrate in the period 1928-1962, on 10 stations (Table 1); (b) closure of well fields pumping Rhine bank filtrate in the period 1944-2000, at 17 locations (Table 1); and (c) extension of the classical treatment (aimed at removal of iron, manganese, ammonia and methane), with processes removing organic contaminants.

RBF is both a desired and undesired mechanism of groundwater recharge. It is desired where groundwater tables should not be lowered in the neighborhood of pumping wells (for instance due to risks of land subsidence by compacting clay and peat). It is undesired where recent bank filtrate with its bad quality would displace old, autochthonous groundwater of unimpeachable composition, especially in ecologically susceptible areas.

## **2.2 Types of Well Fields**

In the Netherlands four types of well fields with RBF can be distinguished on the basis of the flow driving mechanism, RBF periodicity, type of river bed and type of contact of river with the aquifer:

1. pumping driven (and therefore receiving RBF from one side and autochthonous groundwater from the other side composed of uplands bordering the valley), periodical (flow reversing during low river stages or reduced pumping), with a fluvial gravel bed in direct contact with the aquifer;
2. as 1 but permanent (without flow reversal) and with a sandy river bed;
3. polder and pumping driven (and therefore receiving RBF from all sides), permanent, with a sandy river bed in direct contact with the aquifer; and

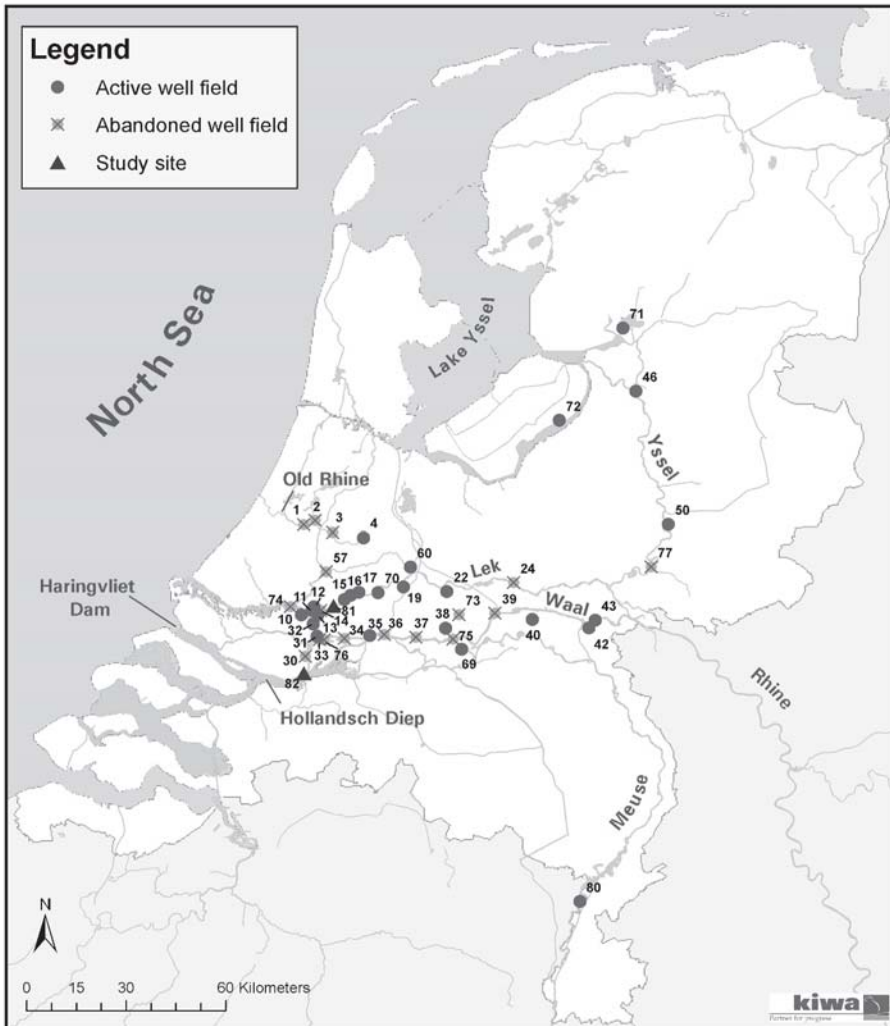


Figure 1. Location of all well fields pumping riverbank filtrate in the Netherlands, with distinction between active and abandoned sites. Numbering corresponding with Table 1.

4. as 3 but not in direct contact with the aquifer by intercalation of at least one aquitard (mostly of Holocene age).

The most frequent types in the Netherlands are 3 and 4, type 1 is unique (Table 1). Types 3 and 4 have been distinguished on the basis of the redox level of the water pumped: if anoxic (no sulphate reduction) then 3, if deep anoxic (with significant sulphate reduction and methane present) then 4.

Lake bank filtrate (type 5 in Table 1) is pumped on two sites in the Netherlands.

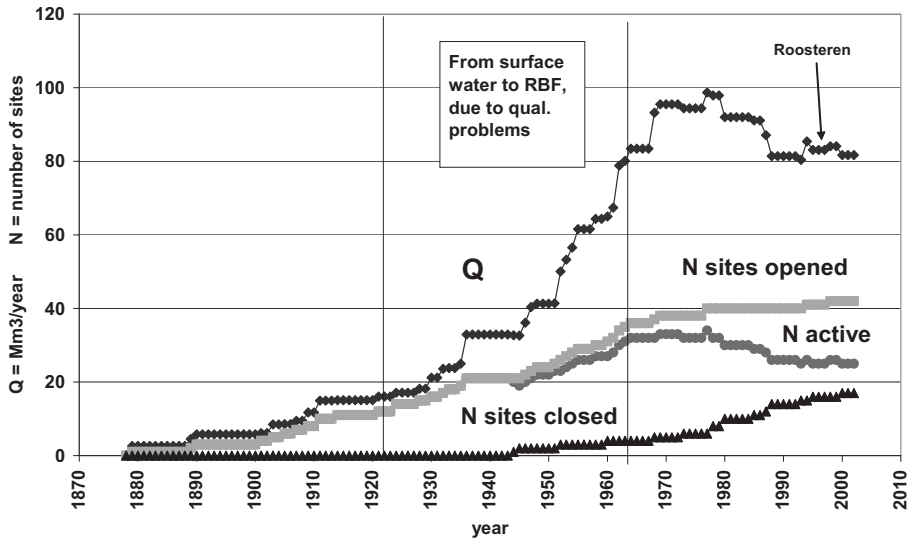


Figure 2. The total annual amount of riverbank filtrate pumped for drinking water preparation in the Netherlands (Q), and the number of well fields pumping this water (N active), from 1879 till 2002. Induced recharge along lake banks, canals and ditches excluded, admixed autochthonous groundwater included.

Table 1. Characteristics of all well fields in the Netherlands that pump(ed) river bank filtrate (type 5 = lake bank filtrate).

No.	PUMPING STATION NAME / SITE	RBF	START	END	END	ALTITUDE
		TYP	PS	Surf.wat	PS	m+MSL
		1	2	3	4	5
<b>LEK RIVER (RHINE)</b>						
10	RIDDERKERK (KIEVIETSWEG)	4	1906	1961		-1
11	LEKKERKERK (SCHUWACHT)	3	1910	1963		1
12	LEKKERKERK (TIENDWEG)	3	1969			-1.5
13	N. LEKKERLAND	3	1964			-1.5
14	N. LEKKERLAND (LEKDIJK)	3	1922	1960	1978	0
15	BERGAMBACHT (DIJKLAAN)	3	1936			-0.5
16	BERGAMBACHT	3	1968			1.5
17	SCHOONHOVEN	3	1901			-1
19	LEXMOND (DE LAAK)	3	1936			0.5
22	CULEMBORG	3	1911			2
24	REMMERDEN	2	1977		1988	6.8
60	IJSSELSTEIN	3	1911			4.5
70	LANGERAK (DE STEEG)	3	1994			
<b>MEUSE RIVER</b>						
80	ROOSTEREN (DE RUG)	1	1990			28.6
<b>OLD RHINE RIVER</b>						
1	HAZERSWOUDE	3-4	1909		1995	-0.5
2	ALPHEN a/d RIJN (HOORN)	3-4	1903		2000	-1
3	BODEGRAVEN (BUITENKERK)	3-4	1907		1985	0
4	KAMERIK (HOOG BOOM)	3	1931			-1
<b>OTHER WATER COURSES</b>						
57	GOUDA	4	1883	1921	1968	-1.5
71	ST. JANSKLOOSTER	5	1936			8.5
72	BREMERBERG	5	1962			-3
<b>WAAL RIVER (RHINE)</b>						
30	s-GRAVENDEEL (KIL)	4	1924		1988	-0.5
31	ZWIJNDRECHT (RINGDIJK)	4	1897	1954		-0.5
32	H-I-AMBACHT	4	1912	1948		-1
33	DORDRECHT (ORANJELAAN)	4	1893	1946	1987	-1
34	SLIEDRECHT	3	1886	1928	1973	-1
35	HARDINXVELD ('T KROMME	3	1924			-1
36	GORINCHEM (VISSERSLAAN)	3	1886	1932	1980	0
37	BRAKEL (VELP)	3	1951		1978	2
38	WAARDENBURG (KOLFF)	3	1958			1.5
39	TIEL	4	1890		1993	6.5
40	DRUTEN	3	1953			5.5
42	NIJMEGEN (NIEUWE	4	1879			14
43	LENT (ELST)	4	1935			8
69	AALST (VELDDRIEL, SELLIK)	3	1977			2.5
73	GELDERMALSEN	3	1924		1952	3
74	VEERDAM (IJSSELMONDE)	4	1903	1933	1944	-1
75	ZALTBOMMEL	3	1905		1960	3
76	DORDRECHT	3-4	1946		1980	-1
<b>YSSEL RIVER (RHINE)</b>						
46	ZWOLLE (ENGELSCH WERK)	2	1930			1
50	ZUTPHEN (VIERAKKER)	2	1889			6
77	DOESBURG	2	1914		1945	9

1 = see section 2.2 for types of well fields; 2 = start Pumping Station (well field); 3 = switch from surface water to RBF; 4 = well field abandoned; 5 = altitude of land surface in m above Mean Sea Level; 6 = depth of well screens pumping RBF (m below land surface); 7 = depth of well screens mainly pumping autochthonous groundwater (m below land surface); 8 = annual total of RBF (incl. autochthonous groundwater) pumped in 1992 or during last year prior to closure; 9 = percentage of RBF in Q as determined

Table 1(continued). Characteristics of all well fields in the Netherlands that pump(ed) river bank filtrate (type 5 = lake bank filtrate).

No.	PUMPING STATION NAME / SITE	SCREEN DEPTH		Q m <sup>3</sup> /a	%	%younger	DISTANCE m
		RBF 6	Autocht 7				
	<b>LEK RIVER (RHINE)</b>						
10	RIDDERKERK (KIEVIETSWEG)	15-30	40-110	2.8	18	34	940
11	LEKKERKERK (SCHUWACHT)	15-30		1.2	100	95	205
12	LEKKERKERK (TIENDWEG)	15-30		2.2	82	92.5	1250
13	N. LEKKERLAND (MIDDELWEG)	15-28		4.3	100	64	630
14	N. LEKKERLAND (LEKDIJK)	15-45		0.7			
15	BERGAMBACHT (DIJKLAAN )	22-42		0.7	100	94	580
16	BERGAMBACHT	17-39		11.	89	50	810
17	SCHOONHOVEN	29-39		0.5	100	95	310
19	LEXMOND (DE LAAK)	18-43	70-120	9.4	47	2	1300
22	CULEMBORG	15-45	70-120	1.2	65 <sup>A</sup>		2000
24	REMMERDEN	3-18		0.4	82	100	30
60	IJSSELSTEIN	70-110	70-110	2.5	21		2000
70	LANGERAK (DE STEEG)				?? <sup>B</sup>		
	<b>MEUSE RIVER</b>						
80	ROOSTEREN (DE RUG)	6-16	16-17 <sup>H</sup>	1.9	10 <sup>C</sup>		145
	<b>OLD RHINE RIVER</b>						
1	HAZERSWOUDE	25-40		2.3	53	70	50
2	ALPHEN a/d RIJN (HOORN)	25-40		1.1	62	70	170
3	BODEGRAVEN (BUITENKERK)	15-35		0.9			
4	KAMERIK (HOOG BOOM)	15-35	67-85	3.9	24	27	
	<b>OTHER WATER COURSES</b>						
57	GOUDA	13-30		0			
71	ST. JANSKLOOSTER	35	85	5.6	60 <sup>D</sup>		
72	BREMERBERG	60	80	7.3			
	<b>WAAL RIVER (RHINE)</b>						
30	s-GRAVENDEEL (KIL)	12-20	50-120	5.2	18	34	175
31	ZWIJNDRECHT (RINGDIJK )	18-25	105-125	3.9	69	61	130
32	H-I-AMBACHT	15-24	80-100	0.9	65	73	300
33	DORDRECHT (ORANJELAAN )	15-25	60-130	4			
34	SLIEDRECHT	10-30		1.1			
35	HARDINXVELD (T KROMME	12-35		1	95	89	350
36	GORINCHEM (VISSERSLAAN)	10-35		2.4			
37	BRAKEL (VELP)	40-45		0.1			
38	WAARDENBURG (KOLFF)	10-40		3.6	68	68	3110
39	TIEL	90-140	90-140	1	82	20	340
40	DRUTEN	10-30	30	1.7	29	35	2980
42	NIJMEGEN (NIEUWE	55	75	2.8	40 <sup>E</sup>	58	370
43	LENT (ELST)	60	100	1.3	66	40	350
69	AALST (VELDDRIEL, SELLIK)	22-72	22-72	3.6	49	2	3500
73	GELDERMALSEN	20-27		0.2			
74	VEERDAM (IJSSELMONDE)	19-24		0.2			
75	ZALTBOMMEL	29-39		0.0			
76	DORDRECHT (W.PARK/JEUGD)	13	70	3.5			
	<b>YSSEL RIVER (RHINE)</b>						
46	ZWOLLE (ENGELSCHER WERK)	30-95	130-170	12.	85	47	815
50	ZUTPHEN (VIERAKKER)	25-40	25-40	2	30	17	1500
77	DOESBURG	32-39		0.1			

by natural tracers; 10 = percentage of young water (age < 22 years) in total mix, as determined by tritium analysis; 11 = horizontal distance between central part of well field and river bank during summer.

A = after 199? <5%; B = ; C = since 1998; D = around 2000; E = in the 1980s ; H = Horizontal well

### 3. CASE 1: MEUSE RBF NEAR ROOSTEREN (SITE 80)

#### 3.1 Situation

Well field No 80 (in Fig.1 and Table 1) near Roosteren along the Meuse River, belongs to type 1 (section 2). It is situated in an area of Pleistocene fluvial terraces, 130 km upstream of tidal influences (28 m+Mean Sea Level). Originally this well field pumped groundwater exclusively recharged by rainwater on the hilly uplands and on the Meuse fluvial plain. In 1998 pumping wells X and XI were installed at 147 m distance from the steep river bank (Fig.3). Since 12 January 1998 they contribute to the supply of raw water for drinking water supply, and since then the river at that site changed from a predominantly effluent into a predominantly influent river.

Intensive hydrological and hydrochemical research was carried out in the period January 1998 – October 1999 (Stuyfzand & Juhász-Holterman, 2000; Juhász-Holterman, 2001).

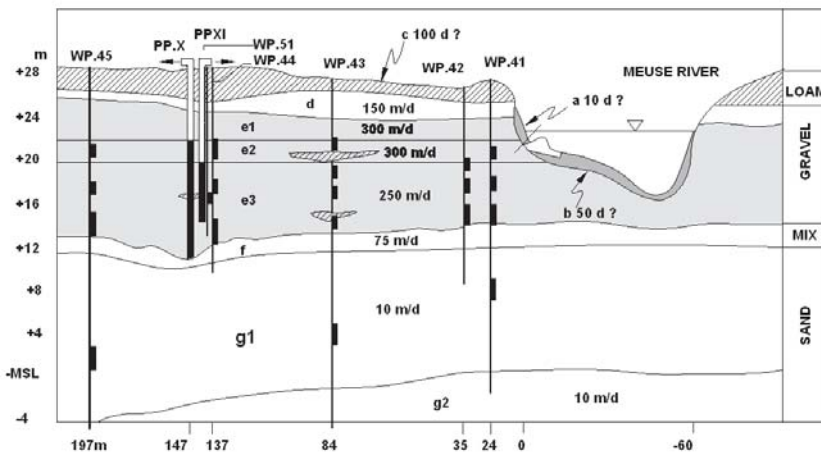


Figure 3. [Please provide a caption for this figure]



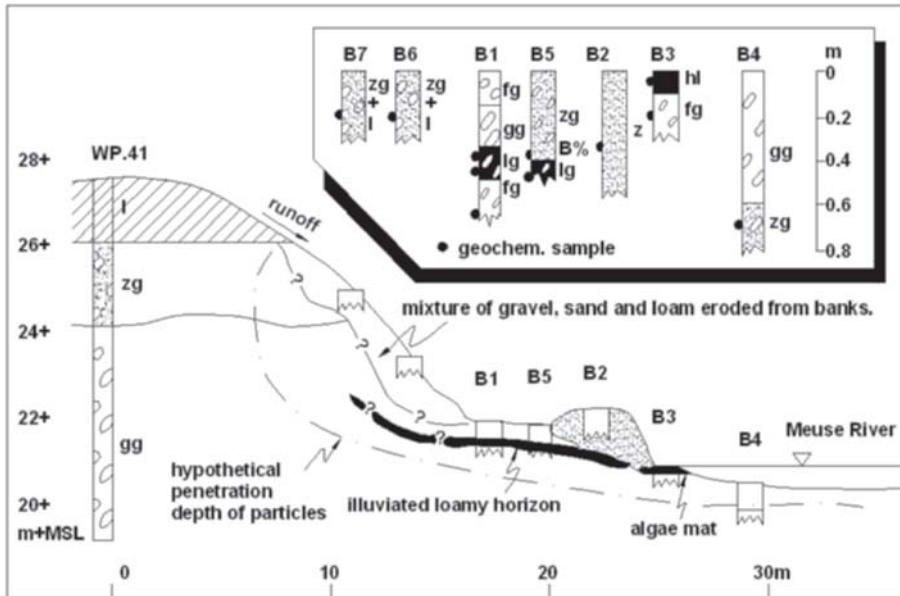


Figure 4. Detailed lithology of the southern bank and bed of the Meuse River as observed in autumn 1999 (after Stuyfzand & Juhász-Holterman, 2000). fg = fine grained gravel; gg = coarse grained gravel; hl = loam with algae; l = loam; zg = sandy gravel; lg = loamy gravel.

### 3.2 Hydrological Aspects (incl. Clogging)

Groundwater flow is directed towards the Meuse River during periods when the wells PP.X and PP.XI do not pump or pump at reduced capacity, and when there is no rapidly rising water level in the Meuse River. The median travel time of infiltrating Meuse River water to the pumping wells at ca. 147 m distance, as determined by natural tracer analysis, strongly depended on the river's water level (pumping rate constant at 140 m<sup>3</sup>/h): 18 days during the rising leg of a high flood wave reaching 25.5 m+MSL, 43 days during mean high water of 23.4 m+MSL and >>54 days during low flows (21.2 m+MSL).

The river bed is composed of gravel (incl. boulders up to 25 cm) with a high tendency to clog. This is due to deep bed filtration of clogging particles (lack of cake filtration) and a small chance of resuspension of the upper boulders that pave the clogging zone.

Piezometric data did not reveal, however, any significant clogging of the river banks and river bed during the 640 days of monitoring. There was indeed no clear increase in hydraulic resistance between the river and the piezometers in the first observation well (WP.41). This does not mean,

however, that clogging did not occur. On the one hand it was calculated that a partial clogging of the river bed would create only 2 cm of hydraulic resistance due to the very high permeability of the upper aquifer. And on the other hand, such small hydraulic changes could be easily caused by variations in pumping rate of the wells upgradient.

### 3.3 Geochemistry

Geochemical reactions of the infiltrate are strong with the recent clogging layer (a + b in Fig.3) and weak with the coarse grained aquifer (e + f + g in Fig.3).

The sedimentological/geochemical characteristics of the four most important aquifer layers are given in Table 2. The gravel aquifer is very inert indeed with extremely low organic carbon and cation exchange capacity (CEC), no calcite, no pyrite and no reduction capacity.

The aquifer sands underlying the gravels, are a bit more reactive especially regarding their reduction and exchange capacity.

The river's bank and bed, in its upper 0.5 m, are quite reactive in all respects, due to significant amounts of clay sized particles, calcite, pyrite, organic matter and a high CEC.

These highly reactive materials derive from suspended matter in the river, and chemical precipitation (calcite and pyrite).

*Table 2.* Composition of the 4 most important aquifer layers at RBF site Roosteren

	unit	River bank	River bed	Gravel aquifer	Sand aquifer
Layer in Fig.3		a	b	e1-e3	g1
No of samples		2	9	9	2
Grains < 2 µm	% d.w.	10.3	7.6	0.4	0.9
Grains > 2 mm	% d.w.	26	44	85	0
CaCO <sub>3</sub>	% d.w.	2.5	1.3	<0.007	<0.002
C-organic	% d.w.	1.7	2.0	0.02	0.1
CEC	meq/kg d.w.	107	103	2.8	7.5
S-FeS <sub>2</sub>	% d.w.	0.017	0.004	<0.0009	0.018
Fe-oxalate	% d.w.	0.47	0.28	0.055	0.027
Mn-oxalate	% d.w.	0.0148	0.0111	0.0044	0.0060

### 3.4 Hydrochemistry

Some data on water quality changes during aquifer passage are given in Table 3, for a period with a high river stage (23.7 m+MSL), normal for the winter period, and thus relatively high infiltration rates. Water quality changes depended mainly on the number of pore flushes with Meuse River water, infiltration intensity (water stage in river; Fig.5) and temperature. Passage of the anoxic clogging layers a (river bank) and b (river bed) clearly resulted in redox reactions and the dissolution of calcite (increase of  $\text{Ca}^{2+}$  by 10 mg/L and of  $\text{HCO}_3^-$  by 15 mg/L). These reactions were insignificant in the aquifer beyond the clogging layers. The redox reactions consisted of oxidation of organic matter and pyrite (or FeS). This was evidenced by a decrease of  $\text{O}_2$  (5 mg  $\text{O}_2$ /L) and  $\text{NO}_3^-$  (1-4 mg  $\text{NO}_3^-$ /L), a pH-decrease (from 7.9 to 6.7) and an increase of total inorganic carbon and  $\text{SO}_4^{2-}$  (with 10 mg/L). Iron and Mn hardly dissolved,  $\text{NH}_4^+$  was nearly completely nitrified.

Organic matter (DOC) was lowered by about 50%, probably by breakdown. Phosphate was nearly completely removed by adsorption while  $\text{SiO}_2$  increased by either desorption or dissolution of feldspars. Fluoride break-through was delayed (retardation factor ca. 4), Barium was clearly mobilized (30-50  $\mu\text{g/L}$ ). The concentrations of all other trace elements (incl. heavy metals) remained quite low, lower than in the Meuse River, probably by filtration and sorption. Concentrations of Organic Micropollutants (OMPs) were very low (<0.01-<0.1  $\mu\text{g/L}$ ) in wintertime (November – April), when the Meuse River is relatively clean and infiltrating well during normal high river flows. The only OMP detected during aquifer passage in winter was 1,2-dichloroethane, with most removal occurring close to the river (in the clogging layers). Although OMP concentrations were clearly much higher during summer (May-October), they could not be detected in Meuse RBF. The most important causes are: (1) the Meuse River hardly infiltrates in that season during the usual low river discharges; (2) the OMPs are delayed by sorption, and did not reach the observation wells during the research period due to a lack of sufficient pore flushes; and (3) decomposition.

### 3.5 Microbiology

Bacteria and viruses were effectively removed in the gravel aquifer by >4 log<sub>10</sub> units during the research period. During extreme flood peaks in winter, however, very low numbers of coliforms, SSRC and coliphage

Table 3. Mean composition (18 Nov. 1998 – 26 March 1999; days 300-479) of Meuse River water and its bank infiltrate along the row of monitoring wells near Roosteren.

	Unit	MEUSE	SHALLOW		MIDDLE DEEP	
		RIVER	WP.41-f1	WP.42-f1	WP41-f2	Wp.43-F2
Distance	m	0	24	35	24	84
Travel time	d	0	4.5	7.5	7	18
Pore flushes		0	27	20	20	6
Temp	°C	5.9	(6)	(7)	(7)	(8)
EC (20°C)	µS/cm	367	404	400	404	481
pH	-	7.91	7.01	6.83	6.93	6.75
O2	mg/L	12.1	8.3	8.3	5.3	4.2
DOC	mg/L	2.7	1.6	-	1.3	0.8
UV-Ext	E/m	8.7	3.6	2.3	3.0	1.4
SiO2	mg/L	6.7	9.5	-	9.0	8.5
TIC ##	mmol/L	2.57	3.49	3.78	3.62	4.87
SI-calcite	-	0.12	-0.66	-0.83	-0.73	-0.74
Cl	mg/L	26	25	23	26	31
SO4	mg/L	33	40	41	41	62
HCO3	mg/L	151	166	163	166	200
NO3	mg/L	15	16.2	16.8	13.9	10.8
F	mg/L	0.20	0.23	0.11	0.19	0.07
Na	mg/L	15.6	16	-	17	20
K	mg/L	2.8	1.9	-	2.4	2.5
Ca	mg/L	58.6	68.5	68.5	68.1	84.1
Mg	mg/L	6.2	4.9	-	5.6	6.3
Fe	mg/L	0.08	0.02	0.02	0.02	0.02
Mn	mg/L	0.04	0.01	0.01	0.01	0.01
NH4	mg/L	0.25	0.03	0.03	0.03	0.03
SiO2	mg/L	6.7	9.5	-	9.0	8.5
Al	µg/L	36	6	-	6	<5
As	µg/L	1.5	0.6		0.6	0.7
Ba	µg/L	20	61		61	56
Cd	µg/L	0.14	0.10		0.10	<0.1
Ni	µg/L	1.5	1.7		3	0.8
Zn	µg/L	40	20		<10	<10
AOX	µg/L	9.5			5	4
1,2-Dclea	µg/L	0.23	0.07		0.07	<0.05
Trichloroethylene	µg/L	0.25	<0.1		<0.1	<0.1
AMPA	µg/L	0.12			<0.05	<0.05
Atrazin	µg/L	0.02	<0.02		<0.02	<0.02
Diuron	µg/L	0.05			<0.05	<0.05

##: TIC = Total Inorganic Carbon =  $\text{H}_2\text{CO}_3 + \text{HCO}_3^- + \text{CO}_3^{2-}$

reached the pumping well (Medema et al., 2001). This is explained by low temperatures (depressing inactivation), and short travel times in the gravel aquifer, being 10-14 instead of 45-65 days. The latter is caused by a >20% shortening of the travel distance and a >400% steepening of the hydraulic

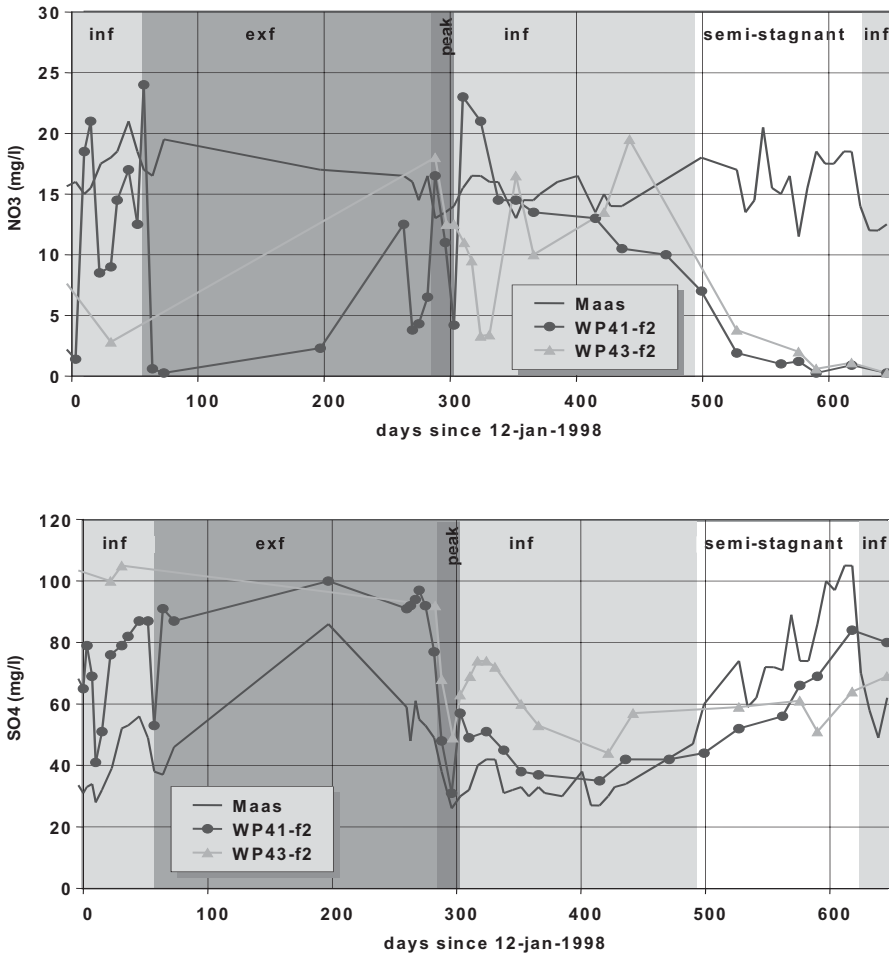


Figure 5. Changes in nitrate and sulphate concentrations in Meuse River water and its RBF at 24 and 84 m distance from the riverbank, in the period January 1998 – October 1999. inf = Meuse infiltrating; exf = Meuse draining; peak = high peak flow; semi-stagnant = hardly any or slight infiltration.

gradient. As elsewhere, the removal rates were highest during the first 7 meters of the aquifer.

## **4. CASE 2: RHINE RBF NEAR OPPERDUIT (SITE 81)**

### **4.1 General Description of Site**

The study site Opperduit is situated 3 km to the west of well field Dijklaan (No 15 in Fig.1 and Table 1), along the northern bank of the Lek River (a tributary of the Rhine River), in the Holocene flood plain, with minor tidal influences (land surface at 0 m+MSL). The river bed is composed of sand (deep fairway) and a complex of Holocene clays and peat (in between the groins and in the winter bed). The site is more or less representative for well fields of type 3 (section 2). Intensive hydrological and hydrochemical research was carried out in the period 1981 – 1994 (Stuyfzand & Lüers, 1996).

### **4.2 Hydrological Aspects (incl. Clogging)**

Groundwater flow is permanently directed towards the central parts of the deep polder (land surface at 1.6 m-MSL) bordering the northern river bank. The pumping of RBF at well fields in the neighborhood has a negligible impact on the local flow pattern. A cross section over the study site is given in Fig.6, showing the position of the sandy aquifer of Middle Pleistocene age (10-35 m-MSL), and the upper Holocene aquitard (clays and peat). The travel time towards the most distant observation well (294, 675 m from river bank) was about 8 years.

There is a moderate tendency to clog in between the groins, and no clogging at all in the fairway (deep summer bed). It seems likely that the winter bed (dry land in summer) is clogging to some extent, but data on this aspect have not yet been studied in detail.

Periodic flood waves appear capable of completely removing the superficial clogging layer (by cake filtration) on the sandy floor of the fairway, where the bulk of aquifer recharge is occurring. RBF sites along sandy river beds thus seem to be protected from the nuisance of river bed clogging.

### **4.3 Geochemistry**

The main aquifer (10-35 m-MSL) is composed of anoxic, calcareous (5%), coarse sands, with 0.1-0.2 % organic carbon, a CEC of 20-60 meq/kg and <0.1% ironsulfides. The top layer (2 m thick) of the winter bed in

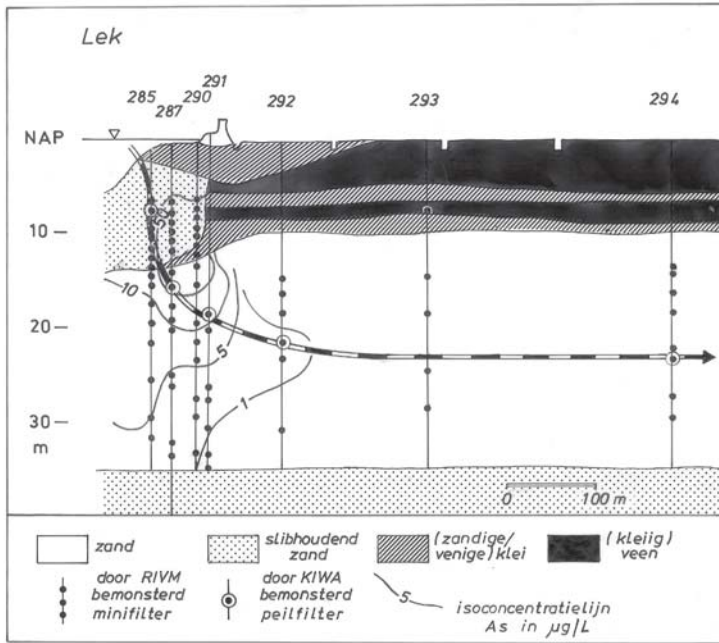


Figure 6. Hydrogeological cross section over the 'polder driven' Rhine bank filtration site Opperduit (site 81 in Fig.1), with isoconcentration lines for arsenic. Stuyfzand (1992).

between the groins is composed of anoxic calcareous (13%) sludge, with on average 8% organic carbon and a CEC of 200 meq/kg

#### 4.4 Hydrochemistry

Some data on mean water quality changes during aquifer passage are given in Table 4. The analyses refer to measurements in 1994-1995. The results indicate that the system is largely anoxic (oxygen and nitrate being completely reduced), and partly deep anoxic (monitoring wells 293-a and 294-a exhibiting sulphate reduction). The system is calcite saturated as expected.

The data reveal clear variations downgradient. Many are connected with trends in the pollution record of the Rhine River. Examples of this are Cl, K, Ca, Mg, NH<sub>4</sub>, B, Ba, F, and bentazone. The concentrations of many pollutants were lower in 1994-1995 lower than in surveys at the same site in the 1980s (Stuyfzand, 1998). These positive developments are due to strong quality improvements in the Rhine River since 1975. They are composed of strong concentration decreases for Na, Cl, Ca, SO<sub>4</sub>, DOC, NH<sub>4</sub>, PO<sub>4</sub> and

Table 4. Mean water quality of Rhine River water (Lek tributary) and its RBF near Opperduit (site 81) in 1994-1995 (based on Stuyfzand & Lüers, 1996). Lek = average 1994.

	Unit	Rhine (Lek)	291-b	292-b	293-a	294-a	Well field 15
Distance	m	0	10	100	220	675	581
Travel time	days	0	450	900	1800	2900	2000
Temp	°C	12.9	12.6	12.9	12.1	11.4	
EC (20°C)	µS/cm	690	797	863	871	837	828
pH	-	7.95	7.58	7.55	7.33	7.54	7.38
O2	mg/L	9.1	<1	<1	<1	<1	<1
DOC	mg/L	3.8	1.6	1.5	2.1	2.0	1.4
TIC	mmol/L	2.76	3.15	3.47	5.82	4.16	3.52
SI-calcite	-	0.33	0.04	0.11	0.11	0.03	*0.11
Cl	mg/L	111	145	166	144	154	145
SO4	mg/L	60	74	69	18	46	77
HCO3	mg/L	163	180	198	318	233	194
NO3	mg/L	15.5	<0.4	<0.4	<0.4	<0.4	<0.2
PO4-ortho	mg/L	0.32	0.35	0.37	0.34	0.73	
F	mg/L	0.15	0.28	0.30	0.14	0.08	0.18
Na	mg/L	61	86	82.9	81.1	84.8	80.5
K	mg/L	5.1	6.2	5.5	9.0	4.6	5.2
Ca	mg/L	72	77	89	92	82	84
Mg	mg/L	10.8	10.8	12.3	12.5	12.5	11.5
Fe	mg/L	0.98##	1.7	1.8	2.65	1.6	2.1
Mn	mg/L	0.13##	1.57	0.84	1.04	0.21	0.65
NH4	mg/L	0.18	0.34	1.21	5.04	2.9	1.19
SiO2	mg/L	4.6	7.3	7.7	9.2	11.1	10.7
Al	µg/L	225##	35	7	8	9	2
As	µg/L	2##	2	<1	<1	1.5	0.5
B	µg/L	100	96	113	158	110	120
Ba	µg/L	118	96	113	158	110	96
Cd	µg/L	0.03	0.04	0.02	<0.01	0.01	<0.05
Cr	µg/L	4##	<0.5	<0.5	<0.5	<0.5	<0.5
Cu	µg/L	5	<0.5	<0.5	<0.5	<0.5	4
Hg	µg/L	<0.01	<0.1	<0.1	<0.1	<0.1	<0.02
Ni	µg/L	3.3	0.5	0.4	0.6	<0.5	<1
Pb	µg/L	0.3	1.3	1.3	0.4	0.8	<1
Se	µg/L	0.2	0.02	0.01	0.01	0.02	<1
Zn	µg/L	26##	<5	<5	<5	<5	5
Tetrachloroethene	µg/L	0.13	<0.01	<0.01	<0.01	<0.01	
Trichloroethylene	µg/L	<0.1	0.21	0.17	<0.01	<0.01	-
AMPA	µg/L	0.54	<0.03	0.05	0.06	0.08	-
Atrazin	µg/L	0.07	<0.01	<0.01	<0.01	<0.02	-
Diuron	µg/L	-	<0.01	<0.01	<0.01	<0.01	-
Bentazone	µg/L	<0.1	<0.07	<0.07	0.33	1.96	0.45
Mecoprop (MCP)	µg/L	0.03	<0.03	<0.03	0.04	0.13	-
Trichloroethylphosphate	µg/L	0.13	0.05	0.05	0.06	0.02	

## = in unfiltered samples, thus including suspended species



the more hydrophylic organics like bentazone, mecoprop (MCP), 1,3,3-trimethyloxindole and chlorobenzenes (see Fig.7 and Table 5).

Table 5. Comparison of the concentration levels of 16 organic pollutants from 1995 with those from 1983 (Stuyfzand & Lüers, 1996).

Compound	log KOW	1983	1995	1995/ 1983
		µg/L	µg/L	
N-acetyl-N-ethylaniline	?	0.03-0.04	<0.01-0.05	<0.3-1.3
p,p'-bis(dimethylamino) benzofenone	?	0.1-0.3	<0.03	0.1
bis-(2-methoxyethyl)ether	?	1	<0.03	<0.03
o-chloroaniline	1.85	0.1-0.5	<0.02-0.27	<0.2-0.5
Sum chloroalkylethers	ca. 2	0.1-3.1	0.07-0.24	0.1-0.7
chlorotoluidine	2.25	0.02-0.04	<0.02-0.06	<1-1.5
diacetonglucose	-0.6	1-1.4	<0.01-0.05	0.02
dichloroaniline	2.7	0.1	<0.01-0.03	0.2
Sum dichlorobenzenes	3.38	0.4-1.4	<0.01-0.02	0.02
difenylsulfone	2.4	0.1-0.3	<0.02-0.07	0.2
dimethylaniline	1.8	0.1-0.4	0.01-0.03	0.1
N-ethyl-N-fenyl-acetamide	?	0.5	<0.03	<0.06
lactones	?	0.02-0.7	0.02-0.19	0.3-1
tributylphosphate	2.5-4	0.04-0.06	0.01-0.14	0.25-1.5
triisobutylphosphate	?	0.03-0.3	0.01-0.09	0.3
1,3,3-trimethyloxindole	?	0.1-0.35	0.02-0.06	0.2

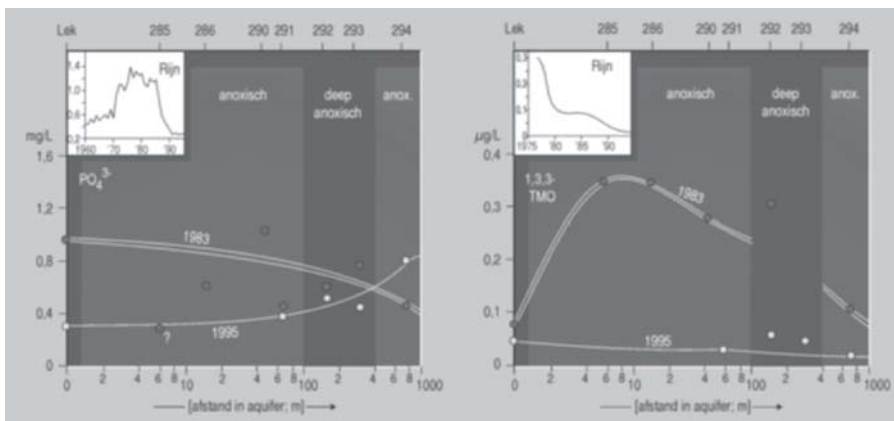


Figure 7. Quality evolution down gradient in the aquifer recharged by the Rhine River at site 81 (Opperduit), in 1983 and 1995 (Stuyfzand & Lüers, 1996). The inset shows concentration trends in the Rhine. 1,3,3-TMO = 1,3,3-trimethyloxindole (a constituent of paint).

A detailed study in the 1980s revealed the behavior of the trace elements As, Co, Cu, Ni and Pb (Fig.8). These showed breakthrough fronts in 1983 at relatively close distance to the supposed infiltration point (<40 m), with Ni advancing most rapidly. Arsenic showed an anomalous peak at 10 m, suggesting that it is mobilized from the aquifer (or rather the river bed; Fig.6) and retarded by sorption downgradient from the source area.

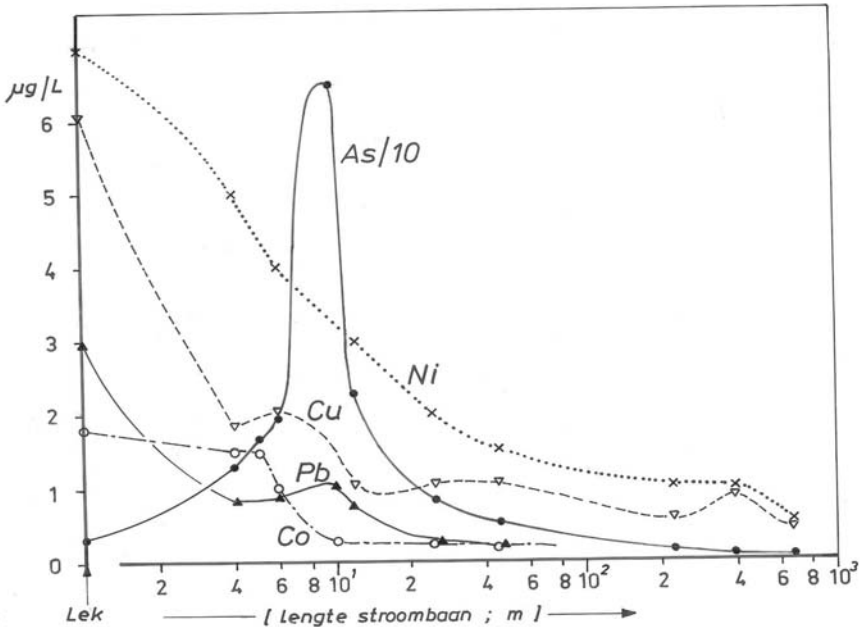


Figure 8. Survey of trace element concentrations in RBF along the Lek at site Opperduit (site 81 in Fig.1). Stuyfzand (1992)

## 5. CASE 3: ESTUARINE RBF (HOLLANDSCH DIEP, SITE 82)

### 5.1 Situation

The last case study presented here, involves a polder area 4 km to the south of well field No 30 (in Fig.1 and Table 1), near Gravendeel, where estuarine water from the Hollandsch Diep (composed of a mix of mainly

Rhine and Meuse River water) is infiltrating. The site is situated in the compound Rhine-Meuse delta, where tidal influences were operating before closure in 1971 of the Haringvliet outlet by a dam with sluices, as part of the famous ‘Delta Works’.

The sandy river bed seriously clogged on this site due to reduced river flows. This clogging layer therefore was not primarily the result of RBF. However, its presence has a tremendous impact on water quality, and serves therefore as an interesting example. The site is somehow representative for well fields of type 4 (section 2), although it exhibits exceptional characteristics. Intensive hydrological and hydrochemical research was carried out in the period 1997 – 1999 (Stuyfzand & Zindler, 1998; Stuyfzand et al., 1999, 2004).

## **5.2 Hydrological Aspects (including Clogging)**

Groundwater flow is permanently directed towards the central parts of the deep polder (land surface at 1.6 m-MSL) bordering the northern estuary bank. The pumping of RBF at well field 30 had no impact on the local flow pattern, because it was abandoned in 1988.

A cross section over the study site is given in Fig.9, showing the position of the recent sludge layer at the bottom of the Hollandsch Diep estuary in 1990-2000, an upper sandy aquifer of Holocene age (0-15 m-MSL) with an intercalated, discontinuous peat aquitard (5-8 m-MSL), a continuous aquitard of Middle Pleistocene age (15-20 m-MSL) and a sandy aquifer of Middle to Lower Pleistocene age (20->35 m-MSL).

It follows from diagnostic concentrations of Cl and <sup>18</sup>O (Stuyfzand et al., 2004) that water which infiltrated the Hollandsch Diep in the study area during the period 1972-1996 consisted of a mixture of Rhine River water (72%), Meuse River water (28%) and North Sea water (0.1%). The clogging sludge layer of 0.2-4 m thickness in 1997 thus formed in 26 years. Along the estuary banks the deposition is limited by wave action, and in the fairway it is curtailed by shipping and frequent dredging.

## **5.3 Geochemistry**

Only the upper aquifer system and the sludge layer have been studied in detail. Analyses of cores of the upper, recent sludge layer (formed after 1971), the underlying old sandy sludge layer (formed before 1971) and Holocene sands are presented in Table 6. All sediment samples are deep anoxic and calcareous. The recent sludge layer contains most fine particles,

calcium carbonate, bulk organic material and iron sulphides, the sand the least.

## **5.4 Hydrochemistry**

Water quality data are presented in Table 8, for four generic groundwater types whose spatial distribution is shown in Fig.9: anoxic water type A (along the estuary banks) with little sludge contact; water type C (in the deeper parts of the sludge layer and just beneath thick sludge) with the most intense sludge contact; water type B (downgradient of type C), with strong sludge contact; and water type F (in the deeper aquifer), without sludge contact but with strong imprints of the aquitard at 15-20 m-MSL.

All watertypes are fresh ( $Cl < 300$  mg/l; E and F excluded which are slightly brackish), (deep) anoxic mainly by reaction with abundant labile organic material in sludge and aquitards, and calcite saturated.

Contact with the recent sludge at the bottom of the estuary yields extremely high concentrations of Ca,  $HCO_3$ , DOC, Br,  $NH_4$ , Mg, K, Fe,  $CH_4$ , As and B. The high Br concentrations, which probably derive from organic material, result in anomalously low Cl/Br ratios in watertype C. Pore water samples taken from and under the recent sludge layer also reveal extreme concentration levels, even higher than groundwater type C (Table 7). Calculations show that the extreme  $NH_4$  and Mg concentrations approximate equilibrium with the mineral struvite ( $MgNH_4PO_4 \cdot 6H_2O$ ) which is known to form in sewage treatment plants (Schellekens, 2004). This could imply that parts of the sludge layer are composed of sewage treatment waste.

Passage of the thick aquitard of Middle Pleistocene age at 15-20 m depth does not result in the extreme concentration increases as the passage of the recent sludge layer does. This is due to a prolonged leaching of this old fluvial layer and a far better water quality during its deposition. The raised concentrations of Na, K and Mg in water type F are mainly due to cation exchange as a result of an earlier fresh water intrusion in this formerly brackish aquifer.

For a further discussion of the hydrochemistry, including all watertypes, geochemical reactions and a discussion of the potential effects of pressure filtration in compacting sludge layers, refer to Stuyfzand & Zindler (1998) and Stuyfzand et al. (1999).

Table 6. Composition of recent sludge (formed after 1971), old sandy sludge (formed before 1971) and Holocene sands (upper aquifer) on site 82 in the Hollandsch Diep.

Description	Unit	Recent sludge			Old sandy sludge		Holocene sand	
depth top	m-LS	0.1	1.25	1.4	2	2.75	2.95	4.7
depth bottom	m-LS	0.25	1.4	1.55	2.15	2.9	3.45	4.9
Fraction<2 um	%	11	29.1	23.2	5.6	10.3	0.6	2
Fraction<16 um	% dw	19.7	48.8	39.7	11.2	18.3	2.4	2.6
C-organic	%	2.1	6.8	7.0	2.3	3.4	1.2	0.1
Bulk Org Matter	%	8.7	13.9	14.9	7.6	9.2	2.9	0.1
CaCO <sub>3</sub>	%	10.0	10.8	12.5	10.0	7.5	4.2	4.2
CEC	meq/kg	121	362	335	103	164	43	13
<b>TOTAL ELEMENT CONTENT (ANALYSIS BY XRF)</b>								
Al	%	4.07	5.95	4.89	2.65	3.11	2.39	1.94
As	mg/kg	10.2	42.2	72.9	58.0	115.0	18.1	1.0
Ca	%	4.36	4.86	5.02	3.96	3.92	1.67	2.12
Fe	%	2.09	4.18	3.56	1.64	2.15	0.73	0.53
K	%	1.64	1.97	1.69	1.31	1.31	1.16	1.11
Mg	%	0.70	1.14	0.91	0.44	0.50	0.17	0.08
Mn	%	0.070	0.108	0.083	.040	0.054	0.017	.015
Na	%	0.56	0.42	0.47	0.56	0.52	0.58	0.53
P	%	0.17	0.51	0.51	0.12	0.16	0.04	0.02
S	mg/kg	1100	4700	4900	2000	3100	1000	-
Sorg (calc)	mg/kg	168	544	560	184	272	96	7
Si	%	32.4	24.6	26.0	35.3	33.2	40.2	42.1
Ti	%	0.25	0.44	0.43	0.20	0.26	0.08	0.05

## 5.5 Hydrochemical Evolution

The hydrochemical situation in the area is complicated by pollution related changes in river water quality, variations in the contribution of intruding sea water, an intricate hydrogeological structure with many discontinuous aquitards, changes in the mixing ratio of Rhine and Meuse River water and man-made changes by the construction of weirs, dams, storm surge barriers and fairways.

Nevertheless, a clear picture of the hydrochemistry, origin and ages of the groundwaters in the Hollandsch Diep estuary could be made (Fig.9). The

Table 7. Chemical composition of pore water and groundwater from recent sludge (formed after 1971), old sandy sludge (formed before 1971) and Holocene sands (upper aquifer) on site 82 in the Hollandsch Diep.

Water sample type		Pore water						Groundwater	
Sediment type		Recent sludge			Old sandy sludge		Sand	sludge	sand
Depth	cm-LS	10-25	125-140	140-155	200-215	275-290	295-345	250	350
pH		7.49	7.6	7.46	7.77	7.9	7.92	7.00	7.22
EGV	uS/cm	630	690	640	920	1140	960	3060	1710
<sup>3</sup> H	TU	66.4	139	81.5	65.2	79.6	73.6	68.1	55.5
Cl <sup>-</sup>	mg/l	93	141	149	148	179	161	200	160
HCO <sub>3</sub> <sup>-</sup>	mg/l	1066	2821	2598	1340	1326	601	2180	970
SO <sub>4</sub> <sup>2-</sup>	mg/l	2.2	3.4	5.9	10	14	114	<1	<1
NO <sub>3</sub> <sup>-</sup>	mg/l	<2.2	<2.2	<2.2	<2.7	3.5	-	<0.1	<0.1
PO <sub>4</sub> <sup>3-</sup>	mg/l	<1.5	<1.5	<1.5	<1.8	<1.8	<1.5	0.28	0.28
Na <sup>+</sup>	mg/l	64	109	118	134	138	114	130	110
K <sup>+</sup>	mg/l	15	44	47	34	40	38	29	22
Ca <sup>2+</sup>	mg/l	151	212	203	547	130	120	310	83
Mg <sup>2+</sup>	mg/l	29	78	85	19	39	35	84	42
Fe <sup>2+</sup>	mg/l	11	2.3	1.5	<0.2	<0.3	<0.2	48	24
Mn <sup>2+</sup>	mg/l	6	0.6	0.4	0.1	0.2	0.2	0.8	0.6
NH <sub>4</sub> <sup>+</sup>	mg/l	36.1	857	868	415	384	255	168	112
DOC	mg/l	27	67	62	30	56	50	48	24
Σ anions	meq/l	20.14	50.3	46.9	26.34	27.1	16.76	41.37	20.4
Σ cations	meq/l	15.70	70.4	71.6	33.96	38.0	28.94	39.84	20.0

Table 8. Quality survey of 5 water types: the estuarine water in the Hollandsch Diep, and river bank filtrates A, B, C and F. Spatial distribution of RBF water types in Fig.9.

Water type		Holl Diep	A	C	B	F
Sludge contact		none	weak	very strong	strong	weak
Depth #	m-LS	0	6	9	14	22
Recharge	period	1996	1960-1998	1972-1998	1972-1992	1920-1952
Watertype @			F3Ca HCO3	f6NH4 HCO3+	F5Ca HCO3+	B4Na HCO3+
Temp	oC	10.5	10.5	10.5	10.5	10.5
EC (20oC)	uS/cm	820	720	3490	1690	1780
pH	-	8.2	7.45	6.8	6.99	6.99
O2	mg/l	9.1	<0.1	<0.1	<0.1	<0.1
CH4	mg/l	0	0.1	19	16	20
3H	TU	49	56	144	54	<0.2
δ18O	‰V-SMOW	-9.2	-8.8	-9.4	-9.1	-9.3
DOC	mg/l	2.9	6	33	23	9
TIC	mmol/l	2.72	4.74	54.1	23.0	13.4
SI-calcite	-	0.55	0.13	0.65	0.53	*0.05
Cl/Br	(mg/l)	-	423	93	141	254
Cl	mg/l	146	110	260	130	330
SO4	mg/l	65	18	<1	<1	<1
HCO3	mg/l	162	265	2400	1120	650
NO3	mg/l	16.1	<0.1	<0.1	<0.1	<0.1
PO4-ortho	mg/l	0.31	5.51	0.46	0.49	0.21
F	ug/l	210	270	320	120	230
Na	mg/l	80.5	59	200	94	220
K	mg/l	6.4	6.3	36	19	10
Ca	mg/l	75	88	230	190	87
Mg	mg/l	12.9	11	110	40	59
NH4	mg/l	0.19	3	230	53	15
Fe	mg/l		2	36	39	4
Mn	mg/l		0.6	0.5	3	0.2
SiO2	mg/l	4.7	27	16	18	24
As	ug/l	<2	6	81	413	0.6
B	ug/l	120	110	580	190	130
Br	ug/l		260	2800	920	1300
Cu	ug/l	<5	3	<1	<1	<1
Ni	ug/l	3.4	2	3	<1	<1
Pb	ug/l	<2	15	<0.5	<0.5	<0.5
Zn	ug/l		56	7	<5	7
phenanthrene	ug/l	0.011	5.2	0.15	0.02	<0.02

# below the bottom of the estuary; @ = using classification of Stuyfzand (1989).

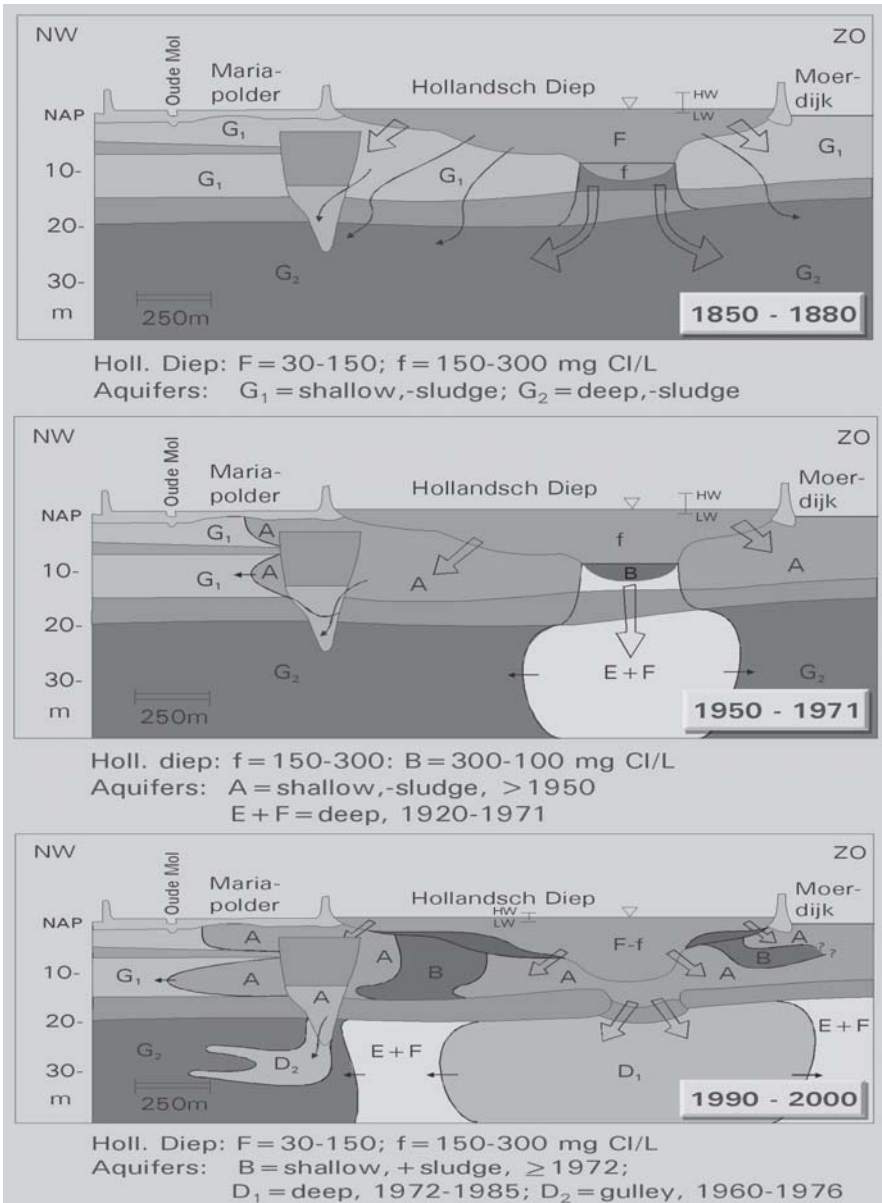


Figure 9. Changes in the spatial distribution of generic groundwater types (some explained in Table 8) along section AA' in the study area, in the period 1850-2000 (after Stuyfzand et al., 2004). Codes for surface water in Hollandsch Diep: F = 30-150 mg Cl<sup>-1</sup>/l; f = 150-300 mg Cl<sup>-1</sup>/l; B = 300-1000 mg Cl<sup>-1</sup>/l; HW = mean High Water level; LW = mean Low Water level.



study area shows a largescale fresh water intrusion of a mix of mainly Rhine and Meuse river waters into an aquifer system that salinized probably during the Holocene transgression and during and after the so-called St. Elisabeth's storm surge in 1421 AD. The fresh water intrusion probably started after 1600 when many dikes were completed in the area, and drainage of the diked land intensified. Remnants of the old saline groundwaters were not observed, because maximum age of the groundwaters studied is about 200 years.

Prior to the construction of the dam in the Haringvliet outlet, there was a period of about 50 years (1920-1970) with reduced river flows and a somewhat increased seawater intrusion. This is evidenced by the limited occurrence of brackish groundwater (water types E+F), dated 30-80 years old by various tracers, in the deep aquifer.

The dam in the Haringvliet outlet, completed in 1971, strongly reduced the seawater intrusion in the study area. The resulting deposition of a 0.1-4 metres thick sludge layer generated, however, a strongly polluted groundwater plume (watertypes B and C in Fig.9). Urgent problems with exfiltrating water from this plume are not expected. The first reason is that this plume does not flow, in the study area, in an upward direction but in a subhorizontal direction towards a remote deep polder 4-5 km to the northwest. The second reason is that accretion and compaction of this layer over time results in an ever increasing hydraulic resistance and thus in a shrinking recharge of the plume.

## **6. CLOGGING**

Experiences with RBF in the Netherlands indicate that there are serious risks of clogging in case of a river bed in gravel, and hardly any such risks where the river bed is sandy and periodically cleaned by flood waves. This means there is an interesting paradox (Fig.10): sand is less permeable than gravel, but can sustain a higher recharge rate (on average) for a longer time in (semi)natural systems. The reason is that cake filtrated material on sand can be more easily removed by river flows than deep bed filtrated material in gravel.

When surface water flow reduces to low velocities, sedimentation of suspended material may become a serious enemy of RBF, as experienced in the Hollandsch Diep.

CHARACTERISTICS	SAND	GRAVEL
mm	0,064-2	> 2
K(h (m/d)	1-100	100-1000
porosity	0,3-0,4	0,2-0,3
CEC (meq/kg)	5-50	< 5
clogging	cake	deeper

Figure 10. The clogging paradox: less permeable sand sustains, in (semi)natural systems, higher recharge rates for a longer time than gravel, because of another clogging mechanism.

## 7. GEOCHEMICAL INTERACTIONS

### 7.1 Overview of Compartments and Processes

RBF systems are subject to the following processes in the four compartments depicted in Fig.11:

1. The surface water compartment: the admixing of rain water (+ dry deposition), evaporation, nutrient uptake, biogenic hardness reduction, volatilisation, photolysis and (bio)degradation;
2. The water sediment interface: filtration, additional  $O_2$  and  $CO_2$  inputs (unsaturated zone or root respiration), mineralisation of organic matter, dissolution of  $CaCO_3$  and iron(hydr)oxides, precipitation of sulphides, nitrification and DOC oxidation;
3. During aquifer passage: displacement of native groundwater, cation exchange, oxidation of pyrite and organic matter, dissolution of various mineral phases (like  $CaCO_3$ ,  $FeCO_3$ ,  $MnO_2$ , opal, silicate minerals), sorption of trace elements, Organic MicroPollutants (OMPs) and micro-organisms,

radioactive decay, (bio)degradation of OMPs and inactivation of micro-organisms;

4. The recovery system: the mixing of various RBF water qualities, and the admixing with native groundwater.

In this section the focus is on the geochemical reactions in compartments 2-3. The most important reactions demonstrated by river bank filtrate in the Netherlands are listed in Table 9.

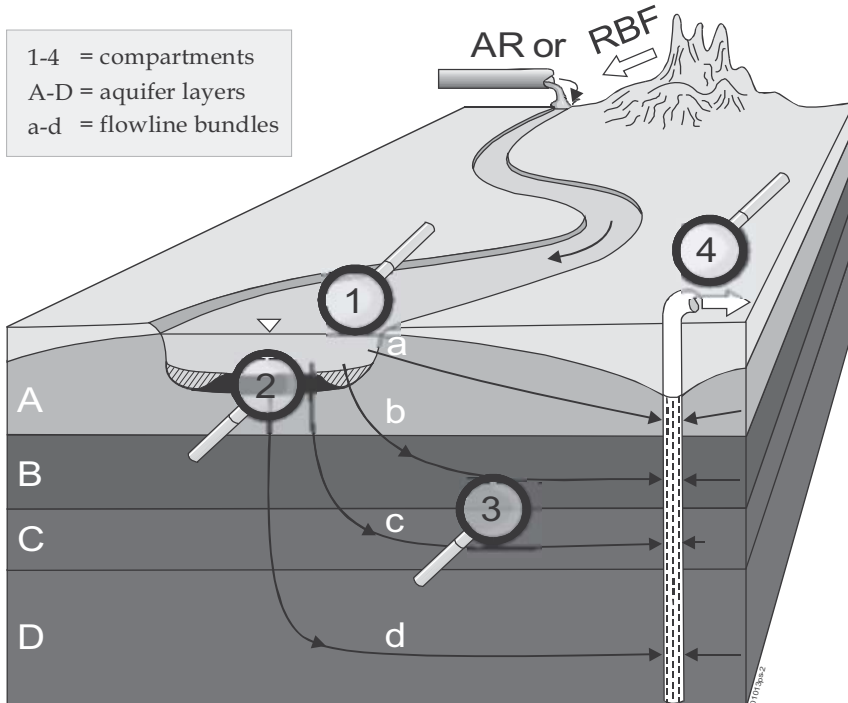


Figure 11. The 4 compartments in RBF systems, with schematisation of the aquifer system, flow, sludge interaction and recovery. Numbering of compartments same as in text above.

## 7.2 Quantifying Geochemical Reactions by the Mass Balance Approach

The extent of all reactions at the water sediment interface and in the aquifer can be quantified by applying the mass balance approach, a technique also referred to as 'inverse modelling'. Chemical mass balances are drawn up by using a set of reaction equations in appropriate order and

Table 9. Listing of the most important hydrogeomical reactions during RBF in the Netherlands, excluding organic micropollutants and micro-organisms.

REACTION EQUATION	Occurrence @	Comments
<b>IN UNSATURATED ZONE</b>		
$2.25\text{O}_2 + \text{FeS} + \text{CaCO}_3 + 1.5\text{H}_2\text{O} \rightarrow \text{Fe}(\text{OH})_3 + \text{SO}_4 + \text{Ca} + \text{CO}_2$	RRR (80)	Oxidation of deep bed filtrated matter especially during winter
Atmospheric O <sub>2</sub> -addition	R (80)	Especially during quickly rising river levels
OX of BOM + RESPIRATION: $\text{O}_2 + \text{CH}_2\text{O} \rightarrow \text{CO}_2 + \text{H}_2\text{O}$	R (80)	Additional CO <sub>2</sub> -inputs along strongly vegetated river banks
<b>IN SATURATED ZONE</b>		
FILTRATION	CCC	Relevant for all silt-bound compounds
<b>Normal redox sequence due to oxidizing NOM #</b>		
$\text{O}_2 + \text{CH}_2\text{O} \rightarrow \text{CO}_2 + \text{H}_2\text{O}$	CCC	Oxidation NOM in aquifer and/or sludge
$0.5\text{O}_2 + 0.4\text{NO}_3 + \text{CH}_2\text{O} - \text{DOC} \rightarrow 0.6\text{CO}_2 + 0.4\text{HCO}_3 + 0.2\text{N}_2 + 0.8\text{H}_2\text{O}$	CC	Oxidation DOC-input, by assumed mix of O <sub>2</sub> and NO <sub>3</sub>
$4\text{NO}_3 + 5\text{CH}_2\text{O} \rightarrow 2\text{N}_2 + \text{CO}_2 + 4\text{HCO}_3 + 3\text{H}_2\text{O}$	CC	Normal denitrification
$\text{NO}_3 + 0.5\text{CH}_2\text{O} \rightarrow \text{NO}_2 + 0.5\text{CO}_2 + 0.5\text{H}_2\text{O}$	R	Partial denitrification
$4\text{NO}_2 + 3\text{CH}_2\text{O} + \text{CO}_2 \rightarrow 2\text{N}_2 + 4\text{HCO}_3 + \text{H}_2\text{O}$	CC	Nitrite removal by reduction
$\text{MnO}_2 + 0.5\text{CH}_2\text{O} + 1.5\text{CO}_2 + 0.5\text{H}_2\text{O} \rightarrow \text{Mn} + 2\text{HCO}_3$	CCC	Typical reaction for RBF, raising Mn/Fe-ratio above normal
$\text{Fe}(\text{OH})_3 + 0.25\text{CH}_2\text{O} + 1.75\text{CO}_2 \rightarrow \text{Fe} + 2\text{HCO}_3 + 0.75\text{H}_2\text{O}$	CCC	Iron dissolution from iron(hydr)oxide (often coatings)
$\text{SO}_4 + 2\text{CH}_2\text{O} + \text{Fe} \rightarrow \text{FeS} + 2\text{CO}_2 + 2\text{H}_2\text{O}$	CC	S-reduction with FeS formation (lack of SO <sub>4</sub> )
$2\text{SO}_4 + 3.5\text{CH}_2\text{O} + \text{Fe} \rightarrow \text{FeS}_2 + 2\text{HCO}_3 + 1.5\text{CO}_2 + 2.5\text{H}_2\text{O}$	CC	S-reduction with pyrite formation (enough SO <sub>4</sub> )
$\text{CO}_2 + 2\text{CH}_2\text{O} \rightarrow \text{CH}_4 + 2\text{CO}_2$	CC	Methanogenesis
<b>Redox reactions with pyrite and siderite</b>		
$3.75\text{O}_2 + \text{FeS}_2 + 4\text{HCO}_3 \rightarrow \text{Fe}(\text{OH})_3 + 2\text{SO}_4 + 4\text{CO}_2 + 0.5\text{H}_2\text{O}$	R	Pyrite oxidation with maximum O <sub>2</sub> availability
$3.5\text{O}_2 + \text{FeS}_2 + 2\text{HCO}_3 \rightarrow \text{Fe} + 2\text{SO}_4 + 2\text{CO}_2 + \text{H}_2\text{O}$	R	Pyrite oxidation with limited O <sub>2</sub> availability
$\text{O}_2 + 4\text{FeCO}_3 + 6\text{H}_2\text{O} \rightarrow 4\text{Fe}(\text{OH})_3 + 4\text{CO}_2$	RR	Siderite oxidation by O <sub>2</sub> with maximum O <sub>2</sub> availability
$\text{NO}_3 + 5\text{FeCO}_3 + 8\text{H}_2\text{O} \rightarrow 5\text{Fe}(\text{OH})_3 + 0.5\text{N}_2 + 4\text{CO}_2 + \text{HCO}_3$	RRR?	Siderite oxidation by NO <sub>3</sub> with maximum NO <sub>3</sub> availability
$3\text{NO}_3 + \text{FeS}_2 + \text{HCO}_3 + \text{H}_2\text{O} \rightarrow \text{Fe}(\text{OH})_3 + 2\text{SO}_4 + \text{CO}_2 + 1.5\text{N}_2$	RRR	Pyrite oxidation with maximum NO <sub>3</sub> availability
$2.8\text{NO}_3 + \text{FeS}_2 + 0.8\text{CO}_2 + 0.4\text{H}_2\text{O} \rightarrow \text{Fe} + 2\text{SO}_4 + 1.4\text{N}_2 + 0.8\text{HCO}_3$	RR	Pyrite oxidation with limited NO <sub>3</sub> availability

Table 9 (continued). Listing of the most important hydrogeological reactions during RBF in the Netherlands, excluding organic micropollutants and micro-organisms.

REACTION EQUATION	Occurrence @	Comments
<b>Exchange reactions</b>		
[Cations-X]-EXCH+Cations-Y ↔ [Cations-Y]-EXCH+Cations-X	CCC	Especially Na <sup>+</sup> , K <sup>+</sup> , Ca <sup>2+</sup> , Mg <sup>2+</sup> , NH <sub>4</sub> <sup>+</sup> , Fe <sup>2+</sup> and Mn <sup>2+</sup> (any combination)
[Anions-X]-EXCH+Anions-Y ↔ [Anions-Y]-EXCH+Anions-X	CCC	Especially F, H <sub>4</sub> SiO <sub>4</sub> , PO <sub>4</sub> , DOC and HCO <sub>3</sub> (any combination)
<b>Dissolution/precipitation and pH-buffering</b>		
CaCO <sub>3</sub> +CO <sub>2</sub> +H <sub>2</sub> O ↔ Ca+2HCO <sub>3</sub>	CCC	Calcite/aragonite dissolution, more rarely also precipitation
2CH <sub>2</sub> O + CaCO <sub>3</sub> → H <sub>2</sub> O + CO <sub>2</sub> +Ca(Hum) <sub>2</sub>	C	Calcite/aragonite dissolution, relevant in high DOC-waters
Fe <sub>1-x</sub> Mn <sub>x</sub> CO <sub>3</sub> + CO <sub>2</sub> + H <sub>2</sub> O ↔ 1- X Fe + XMn + 2HCO <sub>3</sub>	R	Manganous siderite dissolution/precipitation
Fe <sub>3</sub> (PO <sub>4</sub> ) <sub>2</sub> .8H <sub>2</sub> O + 4CO <sub>2</sub> ↔ 3Fe+2H <sub>2</sub> PO <sub>4</sub> +4HCO <sub>3</sub> +4H <sub>2</sub> O	R	Vivianite dissolution/precipitation
Ca <sub>5</sub> (PO <sub>4</sub> ) <sub>3</sub> OH+7CO <sub>2</sub> +6H <sub>2</sub> O ↔ 5Ca+3H <sub>2</sub> PO <sub>4</sub> +7HCO <sub>3</sub>	R	Apatite dissolution/precipitation (strong supersaturation)
beta-Ca <sub>3</sub> (PO <sub>4</sub> ) <sub>2</sub> ↔ 3Ca+2PO <sub>4</sub>	R	Metastable phase, forming often in surface water
MgNH <sub>4</sub> PO <sub>4</sub> .6H <sub>2</sub> O ↔ Mg + NH <sub>4</sub> + PO <sub>4</sub>	RRR? (82)	Struvite dissolution/precipitation
SiO <sub>2</sub> (.nH <sub>2</sub> O)+2H <sub>2</sub> O → H <sub>4</sub> SiO <sub>4</sub> (+nH <sub>2</sub> O)	CC	Quartz and Opal dissolution; opal (diatoms etc.) more soluble
Fe(OH) <sub>3</sub> +xH <sub>4</sub> SiO <sub>4</sub> +H <sub>2</sub> O → SixFe(OH)[3+4x].H <sub>2</sub> O	C/R	Binding of Si in freshly precipitating iron(hydr)oxides
CaAl <sub>2</sub> Si <sub>2</sub> O <sub>8</sub> +2CO <sub>2</sub> +8H <sub>2</sub> O ↔ Ca+2HCO <sub>3</sub> +2H <sub>4</sub> SiO <sub>4</sub> +2Al(OH) <sub>3</sub>	R	Dissolution of anorthite
CaSO <sub>4</sub> .2H <sub>2</sub> O ↔ Ca + SO <sub>4</sub>	RRR?	Not relevant in the Netherlands
<b>REACTIONS UPON MIXING (IN WELL)</b>		
O <sub>2</sub> +4Fe+8HCO <sub>3</sub> +2H <sub>2</sub> O → 4Fe(OH) <sub>3</sub> +8CO <sub>2</sub>	CC	Most important clogging reaction for pumping wells
2O <sub>2</sub> +NH <sub>4</sub> +2HCO <sub>3</sub> → NO <sub>3</sub> +2CO <sub>2</sub> +3H <sub>2</sub> O	CC	Nitrification, also during first meters of aquifer passage
0.5 O <sub>2</sub> + Mn <sup>2+</sup> + 2HCO <sub>3</sub> <sup>-</sup> → MnO <sub>2</sub> + 2CO <sub>2</sub>	C	Black staining
NO <sub>3</sub> +5Fe+9HCO <sub>3</sub> +3H <sub>2</sub> O → 5Fe(OH) <sub>3</sub> +0.5N <sub>2</sub> +9CO <sub>2</sub>	R	Requires enough time and microbiological mediation
NO <sub>2</sub> +3Fe+5HCO <sub>3</sub> +2H <sub>2</sub> O → 3Fe(OH) <sub>3</sub> +0.5N <sub>2</sub> +5CO <sub>2</sub>	R	Requires enough time and microbiological mediation

@: RRR, RR, R = resp. extremely, very and rather rare in the Netherlands; CCC, CC, C = resp. extremely, very and rather common in the Netherlands. (80) = observed on site 80 (see Table 1);

# NOM simplified as CH<sub>2</sub>O, to be replaced by CH<sub>2</sub>O(NH<sub>3</sub>)<sub>Y</sub>(H<sub>3</sub>PO<sub>4</sub>)<sub>Z</sub> with Y = 0.075-0.151, and Z = 0.0045-0.0094

summing up all resulting mass transfers between an arbitrary starting and ending point in a flow system. In the present case the observed composition of the river bank filtrate (during aquifer passage, in an observation well) is simulated by adding to its initial composition (prior to infiltration) the losses and gains by all relevant reactions with the aquifer matrix (Fig.11). If such a simulated transformation of water prior to infiltration into the observed groundwater succeeds well, we may have confidence in the mass balance and we have quantified the relative contribution of each reaction.

Such a simulation approach is, however, never unequivocal and requires further independent evidence from geochemical inspection and laboratory experiments

Mass balances were drawn up with the spread-sheet model REACTIONS+ (Fig.11; for further details see Stuyfzand & Timmer, 1999). Each reaction requires the input of either the reactant or one reaction product, after which the total mass balance is directly recalculated. The distribution of a reactant or reaction product over various reactions may need various trials. The best distribution and the best mass balance are obtained when the result, the simulated water quality after interaction with the aquifer, closely approximates, in this case, the observed water quality as sampled from a monitoring well. The most important calibration terms in the balance are  $\text{CO}_2$  and  $\text{HCO}_3^-$ , because they are not balanced themselves but result from balancing the other parameters.

### **7.3 Results**

Mass balances were drawn up for five observation wells in different hydrochemical RBF environments. As an example part of the results are presented for Meuse River water near Roosteren in Fig.11, with omission of the reactions that did not contribute. A very concise form of presenting the results for all five observation wells is to (1) add up all reactions pertaining to each mineral phase, Natural Organic Matter (NOM), cation and anion exchange, and oxidation of  $\text{NH}_4 + \text{DOC}$  dissolved in the river water prior to infiltration, and (2) separately express all dissolved/precipitated minerals, all oxidized NOM, all exchanged cations and anions, and the sum of  $\text{NH}_4$  and DOC oxidized, in  $\mu\text{mol/L}$ .

REACTIONS+ : version 4.5 by Pieter J. Stuyfzand, Kiwa NV	O2	CO2	SO4	NO3	HCO3	Cl	
03-sep-04 03.05 PM = date print				Figures in bold = autobalanced (rest term)		YELLOW = FILL	
CHARACTERISTICS OBSERVATION POINT of OUTPUT -->	i50 (d) = 7		X (m) = 13				
MEAN INPUT = Meuse Water, sampling point grindschip, WINTER PERIOD 1998-1999	O2	CO2	SO4	NO3	HCO3	Cl	
MGL: MEASURED	12.1	3.9	33	15	151	26	
UMOLL: CALCULATED	378	89	344	242	2475	733	
UMOLL: CORRECTED FOR IONIC INBALANCE	378	89	346	244	2491	738	
CONVECTIVE TRANSPORT	0	0	0	25	0	5	
REACTIONS UNSATURATED ZONE	REAC.No						
2.25O2 + FeS + CaCO3 + 1.5H2O --> Fe(OH)3 + SO4 + Ca + CO2	3C	50	50				
atmospheric O2-addition		100					
Ox of BOM + RESPIRATION: O2+CH2O --> CO2 + H2O	5C	603.60					
REACTIONS SATURATED ZONE	REAC.No	O2	CO2	SO4	NO3	HCO3	
FILTRATION	-						
2O2+NH4+2HCO3 --> NO3+2CO2+3H2O	2	-8.00	8.00		4.00	-8.00	
OXIDATION BOM-aquifer/sludge: O2+CH2O --> CO2+H2O	5A	-177.38	156.81		20.57	-20.57	
OXIDATION DOC input 0.5O2+0.4NO3+CH2O --> 0.6CO2+0.4HCO3+0.2N	5B	-58.33	70.00		-46.67	46.67	
3.75O2+FeS2+4HCO3 --> Fe(OH)3+2SO4+4CO2+0.5H2O	3A	-68.79	73.37	36.69		-73.37	
4NO3+5CH2O --> 2N2+CO2+4HCO3+3H2O	9A		2.37		-19.27	19.27	
4NO2+3CH2O+CO2 --> 2N2 + 4HCO3 + H2O	9C		-0.77			3.06	
[Fe,Mn,NH4,Mg]-EXCH+Ca+Na+K <-> [Ca,Na,K]-EXCH+Fe+Mn+NH4+Mg	14						
[OH]-EXCH+H+CO2 <-> [F]-EXCH+HCO3	15		-0.46			0.46	
[HCO3]-EXCH+H2PO4 <-> [H2PO4]-EXCH+HCO3	17					4.35	
SiO2+2H2O --> H4SiO4	21						
Fe(OH)3+H4SiO4+H2O <-> SixFe(OH)[3+4x]H2O	22						
CaCO3+CO2+H2O --> Ca+2HCO3	18		-164.46			328.91	
SUM BALANCE TERMS + INPUT		166	887	433	227	2792	
MEASURED OUTPUT = wp41-f2 (period F)		O2	CO2	SO4	NO3	HCO3	
UMOLL: CORRECTED FOR IONIC INBALANCE		166	887	433	227	2757	
MGL: MEASURED		5.3	39.1	41	13.9	165	
UMOLL: CALCULATED		166	887	427	224	2720	
ALL ZONES INCLUDED:							
SUMMATION of CHANGE in MINERAL CONTENT (phases in red)	SiO2-mineral	CH2O	FeS(2)	CaCO3	Fe(OH)3	EXCH.Cat	MnO2
TOTAL CHANGE IN SOIL CONTENT (UMOLL water)	-30	-117	-68	-214	68	53	0

Figure 12. Fragment of the spread sheet model REACTIONS+ (in EXCEL), showing Meuse River water as input, the RBF water in monitoring well WP41-f2 (from Table 3) as output, and the required reactions to transform the first into the latter. Yellow cells = fill in; Blue cells = use default or change; Roze = check that value >=0; Green = compare results.

The results of this (shown in Table 10), allow a comparison of the extent of the principal reactions diagnosed. It can be concluded from Table 10, that the principal hydrogeochemical reactions are, in general decreasing order: (a) oxidation of NOM from the aquifer (by successively O<sub>2</sub>, NO<sub>3</sub>, MnO<sub>2</sub>, Fe(OH)<sub>3</sub>, SO<sub>4</sub> and CO<sub>2</sub>); (b) calcite dissolution; (c) cation exchange; (d) reductive dissolution of ironhydroxides (by NOM and methane); (e) pyrite formation; (f) dissolution of SiO<sub>2</sub> containing phases; (g) anion exchange, (h) the oxidation of imported NH<sub>4</sub> + DOC, and (i) the reductive dissolution of Mn-phases.

The dissolution or precipitation of other phases like siderite and the various phosphate minerals listed in Table 9 could not be identified with the mass balance approach, although their presence is not contradicted by mineral equilibrium calculations.

Table 10. Comparison of the total hydrogeochemical reaction scheme for riverbank filtrate on 5 locations: reactions with water sediment interface plus aquifer in  $\mu\text{mol/L}$  (+ = formation; - = loss of phase).

Location	Well/ water type	CH <sub>2</sub> O	FeS(2)	CaCO <sub>3</sub>	Fe(OH) <sub>3</sub>	C-exc	A- exc	SiO <sub>2</sub> - minerals	NH <sub>4</sub> + DOC oxid	MnO <sub>2</sub>
Roosteren	WP. 41-f2	-117	-68	-214	68	53	5	-30	-121	0
Opperduit	292-b	-185	67	-1	-100	313	11	-52	-196	-13
Holl Diep	Type A	-1238	478	-184	-512	225	43	-371	0	-8
Holl Diep	Type B	-27163	737	-4254	-1439	236 1	249	-221	0	-53
Holl Diep	Type C	-72691	1171	-10550	-1833	859 1	323	-188	0	-7

CH<sub>2</sub>O = oxidation of bulk organic matter from aquifer;

FeS(2) = hydrotroillite (FeS.nH<sub>2</sub>O) or pyrite (FeS<sub>2</sub>) formation (if +, otherwise oxidation);

CaCO<sub>3</sub> = calcite/aragonite dissolution;

Fe(OH)<sub>3</sub> = reduction of iron(hydr)oxides (if -, otherwise weathering product of pyrite);

C-EXC = cation exchange ( $\mu\text{mol/L}$  sum of all cations involved divided by 2);

A-EXC = anion exchange ( $\mu\text{mol/L}$  sum of all anions involved divided by 2);

SiO<sub>2</sub> minerals = dissolution mainly of opal (biogenic SiO<sub>2</sub>.nH<sub>2</sub>O), also of various silicate minerals;

NH<sub>4</sub> + DOC oxidation = oxidation of species dissolved in river water during infiltration (external load)

MnO<sub>2</sub> = dissolution of MnO<sub>2</sub>-like phases

Deviations of the above-mentioned sequence are mainly due to differences in redox environment:

- (sub)oxic near Roosteren, where O<sub>2</sub> and NO<sub>3</sub> are still present in the aquifer (Table 3)
- anoxic to slightly deep anoxic near Opperduit, where O<sub>2</sub> and NO<sub>3</sub> are absent and some sulfate reduction occurs without much methane forming (Table 4); and
- (very) deep anoxic at the Hollandsch Diep study site, where – especially in watertypes B and C) -- sulfate totally disappeared and high methane concentrations evolved (Table 8).

The mass balance approach also reveals where specific processes have to be taken into account, which are otherwise easily overlooked. At the Roosteren site this was the additional input of O<sub>2</sub> and CO<sub>2</sub> in the unsaturated



zone, because there was otherwise a strong deficit in  $O_2$  to sustain NOM and pyrite oxidation, and in  $CO_2$  to yield the increased TIC levels observed. At the Hollandsch Diep site, for watertypes B and C, additional  $CO_2$  inputs by methanogenesis were needed to explain the very high Ca and  $HCO_3$  concentrations. In fact the measured  $CH_4$  and  $CO_2$  concentrations were probably far too low due to the escape of pressurized gas. In addition, the permanent upward escape of gas bubbles through the calcareous sludge layer probably resulted in a progressive reaction with  $CaCO_3$  ( $CH_4$  escaping to the atmosphere,  $CO_2$  being partly consumed by reaction).

#### **7.4 Physico-Chemical Indications of River Bed Clogging**

For adequate monitoring, prevention and curative measures it is important to recognize any clogging of the river bed at an early stage. One way is to use hydraulic head measurements with due attention to effects of temperature and, only on specific sites, salinity changes or differences.

The other way is to use hydrochemistry as an indicator. Experiences with artificial recharge in the coastal dunes of the Western Netherlands indicate that the clogging of recharge basins is hydrochemically manifested by a.o. concentration decreases for oxidants ( $O_2$  and  $NO_3$  or even  $SO_4$ ), and concentration increases for TIC ( $=CO_2 + HCO_3 + CO_3^{2-}$ , largely as  $HCO_3$ ),  $NH_4$ , Fe and Mn and sometimes also  $SiO_2$  (Stuyfzand, 1985). However, seasonal fluctuations in river water quality and in interaction of the water with the aquifer need to be subtracted from trends in the hydrochemical monitoring network when aiming at an early detection of river bed clogging.

On-line measurements by electrical conductivity (EC) probes buried or drilled into the river bed at selected points could help as well, because clogging will lead to increased concentrations of dissolved solids, evidenced by EC increases. Even more potential is offered by comparing the frequent EC fluctuations in the river with those in the sensors. Changes in lag time between identical peaks reveal changes in travel time, which may be related to clogging (temperature changes need to be taken into account).

In a similar way, temperature fluctuations as measured by thermistor probes can be used, as is currently under investigation in the artificial recharge area in the Amsterdam dune water catchment area (T.N. Olsthoorn, pers. comm.).

## **8. CONCLUDING REMARKS**

River bed clogging on RBF sites in the Netherlands is not a frequent problem. This does not exclude, however, its existence and importance in the long term!

An exception is observed near Roosteren, where the river bed is made up of coarse gravel, and clogging seems to occur. In the Hollandsch Diep estuary, where RBF was abandoned in 1988, clogging was caused by reduced river flows in consequence of damming the Haringvliet outlet as part of the Delta Works. Passage of this clogging layer led to an extreme water quality, with unusually high concentrations of a.o. Ca, HCO<sub>3</sub>, DOC, Br, NH<sub>4</sub>, Mg, K, Fe, CH<sub>4</sub>, As and B.

Use of the mass balance approach is strongly recommended for quickly identifying and quantifying the most probable hydrogeochemical reactions, including those which are easily overlooked otherwise. Examples of the latter are additional O<sub>2</sub> and CO<sub>2</sub> inputs in systems with periodically unsaturated zones and dense vegetation in or along the river banks, and additional CH<sub>4</sub> and CO<sub>2</sub> inputs in systems with thick compacting sludge layers on the river bed.

It is a challenge to further develop and apply physico-chemical methods to detect clogging at an early stage. On-line measurements with thermistor and electrical conductivity probes seem most promising. An important question to be answered is: When or where is riverbed clogging to be prevented and restored? There are situations where clogging is easily circumvented by the system itself, and where clogging thus is beneficial to the RBF process by incrementing both travel times and (too) short flow distances in the aquifer.

## **9. ACKNOWLEDGEMENTS**

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