

Supporting information for:

Predicting the denitrification capacity of sandy aquifers from shorter-term incubation experiments and sediment properties

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Other possible electron donors

During incubations Fe and Mn concentrations in the batch solution were always (mostly far) below 1 mg Fe L^{-1} and $0,5 \text{ mg Mn L}^{-1}$. Only some transition zone samples showed Fe concentrations between 4 and 7 mg Fe L^{-1} at the beginning of incubation. The measured concentrations of Fe(II) and Mn(II) in the groundwater at the origin of the samples are below $<0.5 \text{ mg Fe L}^{-1}$ and $<0.1 \text{ mg Mn L}^{-1}$ in the oxidized zone of both aquifers. Only in the reduced NO_3^- free zone of both aquifers concentrations of Fe(II) and Mn(II) are higher (1 to 7 mg Fe L^{-1} and $<0,1 \text{ mg Mn L}^{-1}$ in the GKA and 4 to 16 mg Fe L^{-1} and 0.1 to 1 mg Mn L^{-1} in the FFA). Therefore, only solids like e.g. pyrite are possible sources for the electron donors for NO_3^- reduction in both aquifers and it is assumed that pyrite is the major source for Fe(II). Recently Korom et al. (2012) indicated that non-pyritic ferrous iron might play a more important role for denitrification than considered up to now. They assume that ferrous iron from amphiboles contributed to denitrification with 2–43% in a glaciofluvial shallow aquifer in North Dakota.

The NH_4^+ concentrations in the groundwater at the sample origins are below detection limit in the GKA and below 0.5 mg N L^{-1} at multilevel well N10 in the FFA, it is assumed that NH_4^+ is not a significant electron donor during NO_3^- reduction in both aquifers (see also section 4.5.1 of the manuscript and below).

Limitations of the $^{15}\text{NO}_3^-$ labelling approach

For the quantification of denitrification ^{15}N labelled NO_3^- was used during the conducted anaerobic incubations. ^{15}N labelling of nitrate can not completely exclude the possible contribution of dissimilatory nitrate reduction to ammonium (DNRA) followed by anaerobic ammonium oxidation (anammox) to the formation of ^{15}N labelled N_2 from the labelled NO_3^- during anaerobic incubations.

Under strict anaerobic conditions, DNRA is an alternative pathway for the reduction of NO_3^- . But DNRA is seldom reported to be the dominant process of NO_3^- reduction in groundwater systems (Rivett et al., 2008) and chemical modelling by van de Leemput et al. (2011) suggested that DNRA is rather of importance under low NO_3^- concentrations and high C: NO_3^- ratios. But denitrification was presumably not NO_3^- limited since NO_3^- concentrations were always above 1 mg N L^{-1} (Wall et al., 2005) during the incubations. DNRA is presumably not an important process during this investigation because the batch

solutions contained only small amounts of NH_4^+ ($<0,5 \text{ mg N L}^{-1}$, samples from B2 in depth 8-10 m $\approx 1 \text{ mg N L}^{-1}$). Also NH_4^+ accumulation was generally not observed during the conducted experiments. Since the incubations were anaerobic NH_4^+ accumulation should be expected if DNRA was a significant contributing process, except anammox consumed the possibly produced NH_4^+ immediately. If significant N_2 production via anammox occurred, this would have been difficult to observe since NH_4^+ and NO_2^- , the educts of this process, came from the same ^{15}N labelled NO_3^- pool in the batch solution. (At the beginning of incubation NO_2^- concentrations were below detection and NH_4^+ concentrations $<0,5 \text{ mg N L}^{-1}$, respectively.) If anammox contributed significantly to N_2 production, than also DNRA must have been a significant process with half the turnover rate of anammox.

Contrary to marine environments, where high rates of anammox are reported (Canfield et al., 2010), in freshwater systems there is not much evidence for anammox (van de Leemput et al., 2011; Burgin and Hamilton, 2007). To our knowledge, there are no studies about anammox in fresh water aquifers, whereas it is reported to exist in wastewater treatment systems, marine sediments and lakes (Jetten et al., 1998; Schubert et al., 2006; Dalsgaard et al., 2005). To distinguish anammox from denitrification during anaerobic incubation experiments ^{15}N labelled NO_2^- might be used.

NH_4^+ concentrations in the groundwater are mostly below detection limit in the GKA and in the reduced zone at multilevel well N10 in the FFA between $0,3$ and $0,5 \text{ mg NH}_4^+ \text{ L}^{-1}$ (own measurements). Therefore, the possible occurrence of DNRA or DNRA-anammox can not strictly be excluded in both aquifers.

Quantification of total $\text{N}_2+\text{N}_2\text{O}$ production

The molecular ion masses 28 and 29 ($^{28}\text{N}_2$, $^{29}\text{N}_2$) were recorded for IRMS analysis of denitrification derived ^{15}N labelled N_2 and N_2O . The N_2O in the headspace samples was reduced to N_2 in a reduction column prior to the mass spectrometer entrance. The headspace samples were a mixture of unlabeled N_2 und denitrification denitrified ^{15}N labelled N_2 and N_2O . On condition that (i) the ^{15}N abundance of the denitrified NO_3^- is known, (ii) denitrification is the sole gaseous nitrogen forming process and (iii) the amount of N_2 evolved from the ^{15}N labelled NO_3^- pool is small compared with the unlabelled N_2 in the sample, the fraction of denitrified N_2 in a given mixture can be determined by measuring only $^{29}\text{N}_2/^{28}\text{N}_2$ ratios using the equations provided by Mulvaney (1984) (see also discussion in: (Mulvaney, 1984) and (Eschenbach and Well, 2011)). For the measurement of the ^{15}N abundance of the

denitrified NO_3^- and to check for the conditions mentioned above, replicate samples were measured as described in detail in (Well et al., 1998).

The headspace samples represented a mixture of two binomial N_2 isotopologue distributions according to the ^{15}N abundances of the unlabelled N_2 and the ^{15}N labelled denitrification derived ($\text{N}_2+\text{N}_2\text{O}$), respectively. A high frequency discharge unit was then used for online equilibration of N_2 molecules prior to isotope analyses. After equilibration the measured samples consisted of one binomial distribution of N_2 isotopologues according to the total ^{15}N abundance of the mixture. The ^{15}N abundance of the denitrified NO_3^- can then be calculated from the measurement of the $^{29}\text{N}_2/^{28}\text{N}_2$ ratios of unequilibrated and equilibrated replicate samples (Well et al., 1998).

Fit between NO_3^- consumption and ($\text{N}_2+\text{N}_2\text{O}$) production

The NO_3^- decrease during incubations showed the same pattern as the measured production of ($\text{N}_2+\text{N}_2\text{O}$) by GC-IRMS. The measurement of ($\text{N}_2+\text{N}_2\text{O}$) production by GC-IRMS was more precise and had a lower detection limit compared to the measurement of NO_3^- consumption (compare Fig. 1a and Fig. S3a).

The N balance between the NO_3^- content at the start of incubations and the sum of NO_3^- and denitrification derived ($\text{N}_2+\text{N}_2\text{O}$) concentrations during incubation was for most of the incubated samples $< 1 \text{ mg N / batch assay}$. The samples with the highest measured production of ($\text{N}_2+\text{N}_2\text{O}$) showed also the highest deviation between the amount of NO_3^- consumed and the measured production of ($\text{N}_2+\text{N}_2\text{O}$) (compare Fig. 1c and Fig. S3c).

Recommendations for future anaerobic incubations

Control of air contamination during incubation experiments

Canfield et al. (2010) recommended to de-aerate rubber septa by boiling them for 24 hour in water and store them in a He atmosphere before use. An elegant way to check for possible air contamination is the measurement of Ar in the headspace of the transfusion bottles during incubation. Increasing Ar concentrations are an indicator of air contaminations during incubation. Unfortunately we were not able to measure Ar during the incubations, due to instrumental restrictions.

Table S1. Sediment parameters and basic properties of all incubated samples

Sample location	Depth interval [m]	SG ^a	SO ₄ ^{2-b}	DOC ^c	C _{hws} ^d	C _l ^e	C _{org}	total-S	total-N	Sand	Silt
								mg S kg ⁻¹	mg C kg		
FFA B1	6.0-7.0	s ⁿ	3.3	7.2	30.3	82.2	643	86	33	95.0	5.0
FFA B1	7.0-8.0	s	3.3	5.7	32.3	887.0	5955	603	94	94.8	5.2
FFA B2	2.0-3.0	n s ⁿ	10.2	11.5	20.0	2.7	237	29	26	98.9	0.2
FFA B2	3.0-4.0	n s ⁿ	25.3	10.2	17.2	2.7	203	38	23	98.9	0.2
FFA B2	4.0-5.0	n s ⁿ	19.5	8.9	21.6	228.6	545	46	54	96.4	1.3
FFA B2	8.0-9.0	s ⁿ	0.0	6.9	33.8	93.9	1625	176	31	40.4	59.6
FFA B2	9.0-10.0	s ⁿ	0.9	6.2	40.0	116.9	538	156	28	94.7	5.3
FFA B4	7.0-8.0	s	n.d. ¹	n.d. ¹	n.d. ¹	n.d. ¹	483	220	21	97.3	2.7
FFA B4	8.0-9.0	s	n.d. ¹	n.d. ¹	n.d. ¹	n.d. ¹	1114	359	39	95.4	4.7
FFA B6	2.0-3.0	n s ⁿ	17.7	11.6	22.1	259.6	695	56	41	97.8	0.6
FFA B6	3.0-4.0	n s ⁿ	23.3	10.3	21.6	172.5	1047	59	46	97.8	0.4
FFA N10	4.5-5.0	s ⁿ	5.4	9.2	22.2	462.7	1291	50	87	94.9	1.0
FFA N10	5.0-5.5	s ⁿ	3.8	9.6	27.6	206.9	737	49	55	98.0	0.3
FFA N10	5.5-6.0	s ⁿ	12.8	10.8	28.4	160.6	687	49	36	97.4	0.4
FFA N10	7.7-8.3	s ⁿ	n.d. ¹	n.d. ¹	41.2	n.d. ¹	311	57	10	96.3	3.8
FFA N10	8.3-8.6	s ⁿ	n.d. ¹	n.d. ¹	42.5	n.d. ¹	320	47	11	97.9	2.2
FFA N10	10.0-10.4	s	n.d. ¹	n.d. ¹	n.d. ¹	n.d. ¹	310	45	18	96.3	3.7
FFA N10	10.4-10.7	s	n.d. ¹	n.d. ¹	n.d. ¹	n.d. ¹	5627	464	113	96.4	3.6
FFA N10	12.0-13.0	s	n.d. ¹	n.d. ¹	0.0	n.d. ¹	2554	558	64	96.7	3.3
FFA N10	13.0-14.0	s	n.d. ¹	n.d. ¹	39.7	n.d. ¹	1848	588	53	95.1	4.9
FFA N10	16.0-17.0	s	1.1	5.7	42.6	241.0	2608	448	51	97.2	2.8
FFA N10	17.0-18.0	s	n.d. ¹	n.d. ¹	41.1	n.d. ¹	2504	441	48	96.9	3.1
GKA	8.0-9.0	n s ⁿ	14.5	8.1	18.3	1.8	102	54	9	96.8	1.4
GKA	9.0-10.0	n s ⁿ	14.5	9.0	14.9	0.9	76	38	6	97.3	0.9
GKA	22.0-23.0	n s ⁿ	11.1	8.6	43.8	221.3	176	42	15	95.4	1.2
GKA	23.0-24.0	n s ⁿ	10.8	9.4	33.7	50.3	192	36	23	96.0	0.9
GKA	25.9-27.0	s	8.2	6.1	31.1	1021.2	2553	682	69	87.6	12.4
GKA	27.0-28.3	s	4.8	5.8	39.0	1531.1	6373	989	127	79.6	20.4
GKA	28.3-29.3	s	10.3	8.1	27.4	2504.9	4159	883	114	76.8	21.3
GKA	29.3-30.3	s	12.7	6.6	26.2	2205.8	4543	760	96	83.9	14.2
GKA	30.3-31.2	s	13.6	5.2	28.9	347.7	784	509	14	97.6	2.2
GKA	31.3-32.0	s	18.1	9.9	42.6	192.0	834	494	27	96.5	3.2
GKA	32.9-33.7	s	20.2	5.1	20.8	377.4	821	630	23	96.9	2.8
GKA	33.7-34.7	s	15.6	5.3	29.2	150.5	752	510	17	98.5	1.4
GKA	35.7-36.7	s	2.2	5.4	32.0	2391.1	8972	708	120	96.9	3.1
GKA	36.7-37.7	s	5.1	5.5	22.4	37.7	232	677	3	98.8	1.2
GKA	37.7-38.7	s	0.5	4.7	23.2	447.4	1162	379	30	97.8	2.3
GKA	65.1-65.4	s	1.8	6.2	23.7	239.8	1009	716	39	89.4	10.7
GKA	67.1-67.5	n s	0.3	6.9	56.5	132.1	358	196	21	92.1	7.9
GKA	67.5-68.0	n s	3.5	5.2	58.5	n.d. ¹	377	194	44	94.7	5.3

^a sediment group; ^b extractable sulfate-S; ^c extractable dissolved organic carbon; ^d extractable hot-water soluble carbon; ^e KMnO₄ labile organic carbon; ¹ n.d.: not determined; n s non-sulphidic; s sulphidic aquifer material, n s and s with the subscript n indicates NO₃⁻-bearing samples.

Table S2. Denitrification rates, long-term denitrification capacity, stock of reduced compounds, sulphate formation capacity and estimated minimal lifetime of denitrification of all incubated samples.

Sample location	Depth interval [m]	SG ^a	D _r (7) ^b	D _{cum} (365) _d	SRC ^e	SRC _C ^f	SRC _S ^g	aF _{SRC} ^h	SFC ⁱ	em LoD ^j
				$\mu\text{g N kg}^{-1}\text{d}^{-1}$						
FFA B1	6.0-7.0	s ⁿ	51.66	17.18	659.6	599.5	60.1	2.60	6.1	5.0
FFA B1	7.0-8.0	s	33.89	56.24	5974.2	5552.7	421.5	0.94	39.4	44.8
FFA B2	2.0-3.0	n s ⁿ	1.27	0.19	240.8	220.7	20.1	0.08	0.1	1.8
FFA B2	3.0-4.0	n s ⁿ	2.12	0.37	215.4	189.2	26.3	0.17	-0.1	1.6
FFA B2	4.0-5.0	n s ⁿ	35.27	4.34	540.2	508.0	32.2	0.80	1.0	4.1
FFA B2	8.0-9.0	s ⁿ	21.05	10.53	1638.2	1515.5	122.7	0.64 ^(10.0)	3.5	12.3
FFA B2	9.0-10.0	s ⁿ	41.41	12.68	610.7	502.0	108.7	2.08 ^(26.4)	2.2	4.6
FFA B4	7.0-8.0	s	45.67	20.16	603.6	450.2	153.4	3.34	9.6	4.5
FFA B4	8.0-9.0	s	25.24	34.09	1289.5	1038.9	250.7	2.64	22.0	9.7
FFA B6	2.0-3.0	n s ⁿ	11.53	2.64	687.0	648.9	39.1	0.38	0.3	5.2
FFA B6	3.0-4.0	n s ⁿ	6.93	1.46	1017.4	976.5	40.9	0.14	0.1	7.6
FFA N10	4.5-5.0	s ⁿ	35.97	8.69	1239.0	1204.1	34.8	0.70	1.5	9.3
FFA N10	5.0-5.5	s ⁿ	61.03	8.75	721.6	687.1	34.5	1.21	2.1	5.4
FFA N10	5.5-6.0	s ⁿ	36.99	7.82	674.6	640.3	34.3	1.16	5.2	5.1
FFA N10	7.7-8.3	s ⁿ	33.71	15.04	329.5	290.0	39.5	4.56	1.5	2.5
FFA N10	8.3-8.6	s ⁿ	20.25	15.17	331.5	298.7	32.9	4.58	6.9	2.5
FFA N10	10.0-10.4	s	12.34	17.45	320.6	289.3	31.3	5.44	5.4	2.4
FFA N10	10.4-10.7	s	23.75	50.07	5571.6	5247.7	323.9	0.90	9.4	41.8
FFA N10	12.0-13.0	s	26.47	52.84	2771.3	2381.7	389.6	1.91	37.9	20.8
FFA N10	13.0-14.0	s	35.58	38.04	2134.1	1723.3	410.8	1.78	18.2	16.0
FFA N10	16.0-17.0	s	69.90	46.65	2744.7	2431.5	313.2	1.70 ^(6.3)	23.6	20.6
FFA N10	17.0-18.0	s	34.48	46.55	2642.7	2335.0	307.8	1.76 ^(6.3)	36.8	19.8
GKA	8.0-9.0	n s ⁿ	0.81	0.63	132.6	95.0	37.6	0.47	0.9	1.0
GKA	9.0-10.0	n s ⁿ	0.71	0.34	97.1	70.7	26.4	0.35	0.4	0.7
GKA	22.0-23.0	n s ⁿ	14.68	1.57	193.3	164.2	29.1	0.81	0.2	1.5
GKA	23.0-24.0	n s ⁿ	31.77	2.83	204.5	179.2	25.3	1.38	0.0	1.5
GKA	25.9-27.0	s	26.36	15.63	2857.4	2381.0	476.4	0.55	1.2	21.4
GKA	27.0-28.3	s	29.43	41.82	6634.0	5943.2	690.8	0.63 ^(4.9)	8.3	49.8
GKA	28.3-29.3	s	46.38	37.82	4495.6	3878.5	617.2	0.84 ^(7.3)	13.8	33.7
GKA	29.3-30.3	s	57.08	35.49	4766.8	4236.0	530.8	0.74 ^(6.4)	8.1	35.8
GKA	30.3-31.2	s	26.07	6.54	1086.9	731.4	355.4	0.60	3.8	8.2
GKA	31.3-32.0	s	14.06	4.09	1122.4	777.7	344.7	0.36	5.0	8.4
GKA	32.9-33.7	s	38.39	7.28	1206.0	765.6	440.4	0.60	10.2	9.1
GKA	33.7-34.7	s	62.14	12.25	1057.4	700.9	356.6	1.16	17.7	7.9
GKA	35.7-36.7	s	64.30	52.46	8861.3	8366.7	494.6	0.59 ^(4.6)	30.0	66.5
GKA	36.7-37.7	s	87.51	11.07	689.6	216.7	472.8	1.60	9.2	5.2
GKA	37.7-38.7	s	109.2	12.06	1347.7	1083.1	264.7	0.89 ^(15.3)	4.6	10.1
GKA	65.1-65.4	s	33.12	13.22	1441.2	941.3	499.9	0.92	1.3	10.8
GKA	67.1-67.5	n s	30.54	8.18	471.0	333.8	137.2	1.74	1.3	3.5
GKA	67.5-68.0	n s	23.62	8.11	487.1	351.5	135.6	1.67	0.7	3.7

^a sediment group; ^b initial denitrification rate; ^c average denitrification rate after one year; ^d cumulative denitrification after one year; ^e depot of reactive compounds (SRC); ^f concentration of reduced compounds derived from measured C_{org} ; ^g concentration of reduced compounds derived from total-S values; ^h fraction of SRC available for denitrification during one year of incubation, in parenthesis aF_{SRC} from the intensive treatment; ⁱ sulphate formation capacity (SFC); ^j estimated minimal lifetime of denitrification; n s non-sulphidic; s sulphidic aquifer material, n s and s with the subscript n indicates NO_3^- -bearing samples.

Table S3. Simple regression between $D_{cum}(365)$ and sediment parameters (X), $f^{B-C}(D_{cum}(365)) = A + B \times f^{B-C}(X)$. Regressions with C_{org} , total-S are listed for each partial data set. Regression with a third independent sediment variable are only given, if correlation coefficient were better compared to correlations with C_{org} or total-S.

Data set	X^a	N^b	R^c	A	B
Whole data set	C_{org}	151	0.80	-11.022	2.654
whole data set	total-S	151	0.71	-2.397	0.805
whole data set	C_1	111	0.83	-1.028	0.492
FFA	C_{org}	86	0.72	-26.950	8.017
FFA	total-S	86	0.83	-14.879	6.312
FFA	DOC_{extr}	46	0.84	10.503	-0.495
GKA	C_{org}	65	0.93	-9.525	2.457
GKA	total-S	65	0.86	-0.252	0.026
GKA	C_1	65	0.93	-0.730	0.416
non-sulphidic	C_{org}	44	0.52	-5.434	1.205
non-sulphidic	total-S	44	0.77	-231.440	284.854
non-sulphidic	C_{hws}	44	0.77	-164.600	233.898
sulphidic	C_{org}	107	0.66	-3.097	1.293
sulphidic	total-S	107	0.40	2.747	0.001
sulphidic	C_1	67	0.60	-0.119	0.638
NO_3^- -bearing	C_{org}	64	0.58	-4.946	0.661
NO_3^- -bearing	total-S	64	0.67	-268.670	312.977
NO_3^- -bearing	C_1	56	0.73	-0.737	0.267
NO_3^- -free	C_{org}	87	0.77	-5.862	1.623
NO_3^- -free	total-S	87	0.32	3.741	0.004
transition zone	C_{org}	28	0.58	18.117	-4.020
transition zone	total-S	28	0.20	-178.180	277.350
transition zone	C_1	20	0.73	192.880	-190.340

^a Independent sediment parameter

^b Sample number

^c Correlation coefficient

Table: S4. Ratios of modelled $D_{\text{cum}}(365)$ vs measured $D_{\text{cum}}(365)$ (group means with standard deviation, ranges in parentheses) for samples with high ($> 20 \text{ mg N kg}^{-1}$) and low $D_{\text{cum}}(365)$ ($< 20 \text{ mg N kg}^{-1}$).

Data set	Modelled $D_{\text{cum}}(365)$ / Measured $D_{\text{cum}}(365)$					
	Multiple regressions			Simple regressions		
	Selection I ^a	Selection II ^b	Selection III ^c	C_{org}	Total-S	Best ^d
	$D_{\text{cum}}(365) \geq 20 \text{ mg N kg}^{-1} \text{ yr}^{-1}$					
Whole data set	0.88 ±0.33 (0.33 – 1.67)	0.89 ±0.28 (0.39 – 1.26)	0.87 ±0.24 (0.55 – 1.30)	0.86 ±0.32 (0.29 – 1.53)	0.68 ±0.25 (0.42 – 1.54)	0.83 ±0.38 (0.22 – 1.35)
FFA	0.86 ±0.12 (0.71 – 1.26)	0.86 ±0.50 (0.79 – 0.93)	0.84 ±0.07 (0.74 – 0.94)	0.71 ±0.17 (0.30 – 1.08)	0.86 ±0.15 (0.68 – 1.29)	0.57 ±0.06 (0.49 – 0.66)
GKA	0.89 ±0.33 (0.41 – 1.47)	1.14 ±0.18 (0.78 – 1.38)	1.08 ±0.19 (0.79 – 1.34)	1.14 ±0.19 (0.88 – 1.46)	0.84 ±0.30 (0.39 – 1.38)	1.13 ±0.26 (0.67 – 1.51)
sulphidic	0.73 ±0.22 (0.44 – 1.35)	0.78 ±0.16 (0.57 – 1.13)	1.15 ±0.38 (0.81 – 2.05)	0.74 ±0.22 (0.43 – 1.36)	0.33 ±0.09 (0.23 – 0.68)	0.66 ±0.25 (0.28 – 1.19)
	$D_{\text{cum}}(365) < 20 \text{ mg N kg}^{-1} \text{ yr}^{-1}$					
Whole data set	2.29 ±3.06 (0.20 – 18.28)	1.90 ±2.27 (0.17 – 11.08)	1.38 ±1.02 (0.34 – 6.23)	2.69 ±4.40 (0.23 – 26.07)	3.03 ±3.85 (0.20 – 18.32)	1.72 ±1.49 (0.23 – 8.79)
FFA	2.52 ±3.03 (0.23 – 12.41)	1.77 ±1.44 (0.34 – 5.69)	1.14 ±0.66 (0.26 – 3.41)	3.56 ±4.90 (0.24 – 20.27)	2.63 ±3.39 (0.25 – 13.64)	2.19 ±2.53 (0.18 – 11.82)
GKA	1.73 ±1.29 (0.31 – 5.51)	1.35 ±0.71 (0.23 – 3.10)	1.19 ±0.43 (0.30 – 2.16)	1.39 ±0.82 (0.23 – 3.99)	1.76 ±1.38 (0.34 – 6.02)	1.35 ±0.68 (0.23 – 3.02)
non-sulphidic	1.36 ±1.04 (0.18 – 5.23)	1.36 ±1.04 (0.18 – 5.23)	1.09 ±0.45 (0.52 – 0.45)	1.94 ±2.39 (0.21 – 10.45)	1.47 ±1.00 (0.18 – 8.25)	1.55 ±0.94 (0.24 – 7.26)
sulphidic	1.49 ±0.84 (0.51 – 4.33)	1.29 ±0.66 (0.33 – 3.13)	1.39 ±0.60 (0.43 – 3.19)	1.48 ±0.84 (0.50 – 4.36)	1.27 ±0.61 (0.69 – 3.69)	1.46 ±0.76 (0.44 – 3.49)
transition zone	1.03 ±0.22 (0.71 – 1.52)	1.03 ±0.22 (0.67 – 1.56)	1.01 ±0.13 (0.84 – 1.27)	1.05 ±0.27 (0.64 – 1.77)	1.07 ±0.32 (0.67 – 1.73)	1.03 ±0.24 (0.72 – 1.58)

^a C_{org} and total-S;

^b two sediment parameters giving highest correlation coefficient;

^c stepwise multiple regression;

^d simple regression with the sediment parameter giving the best correlations with $D_{\text{cum}}(365)$;

Table S5. Lambda values of the Box-Cox transformed sediment parameters

Data set	Lambda values											
	$D_r(7)$	$D_r(84)$	$D_r(168)$	$D_{\text{cum}}(365)$	silt	C_{org}	total-S	$\text{SO}_4^{2-}\text{-extr}$	DOC_{extr}	C_{hws}	C_l	SRC
Whole data set	0.512	0.346	0.341	0.294	0.021	-0.056	0.132	0.700	-0.213	0.040	0.171	-0.024
FFA	0.626	0.441	0.428	0.370	0.007	-0.176	-0.196	0.347	1.426	0.811	0.364	-0.185
GKA	0.503	0.345	0.259	0.208	-0.206	-0.080	0.750	0.670	-0.789	-0.133	0.170	0.039
non-sulphidic	0.220	0.100	0.172	0.106	-0.069	-0.050	-1.217	0.784	0.732	-1.400	0.758	1.492
sulphidic	0.219	0.209	0.305	0.059	-0.067	-0.111	1.100	0.358	-2.02	0.635	-0.059	0.229
NO_3^- -bearing	0.408	0.134	0.221	0.235	-0.210	0.108	-1.145	0.650	1.401	-0.039	0.261	0.797
NO_3^- -free	0.160	0.103	0.313	0.144	-0.337	-0.017	0.950	0.214	-2.422	-0.335	0.230	0.492

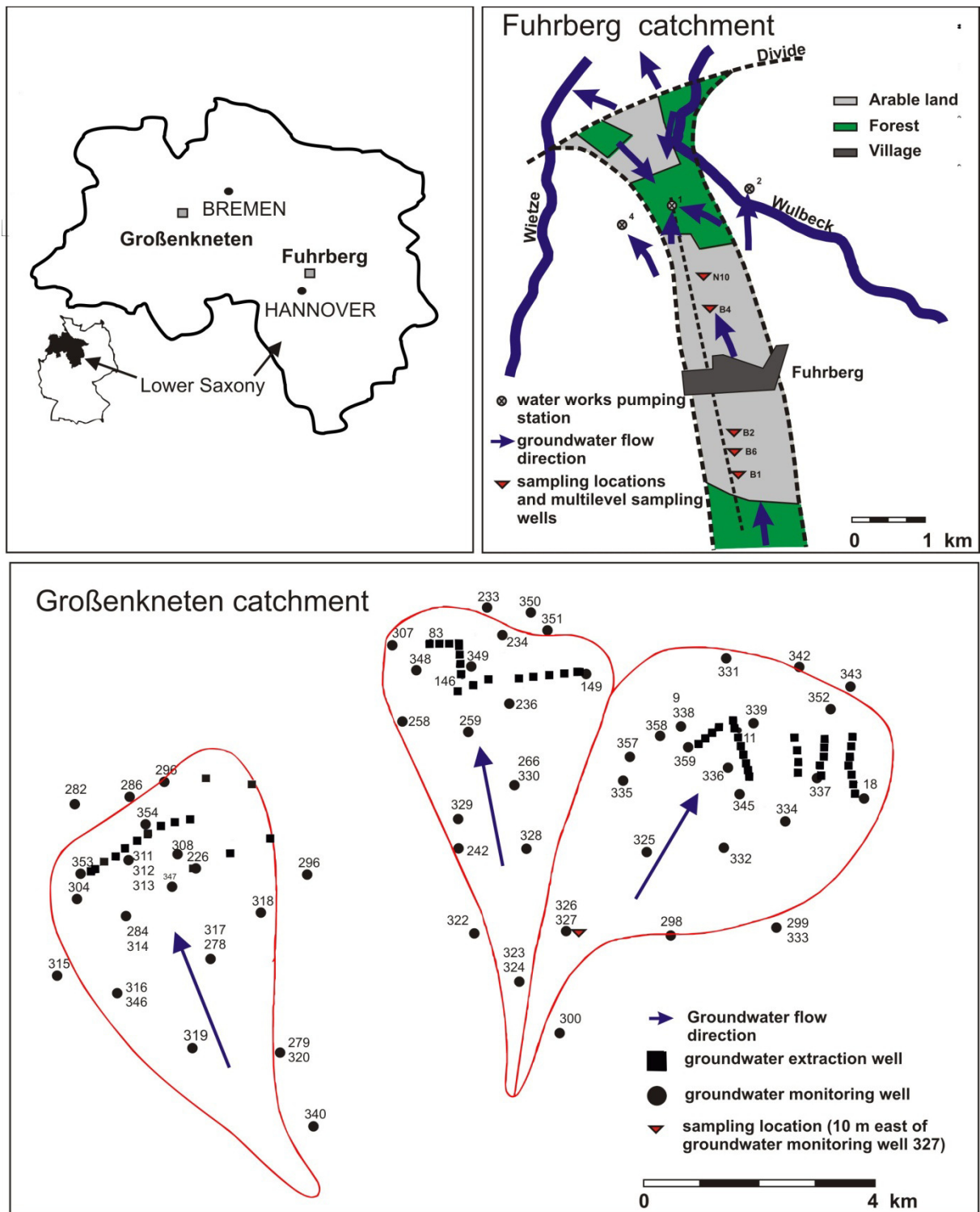


Fig. S1. Sampling locations within the Fuhrberger Feld and Großenkneten catchment in Lower Saxony (Germany).

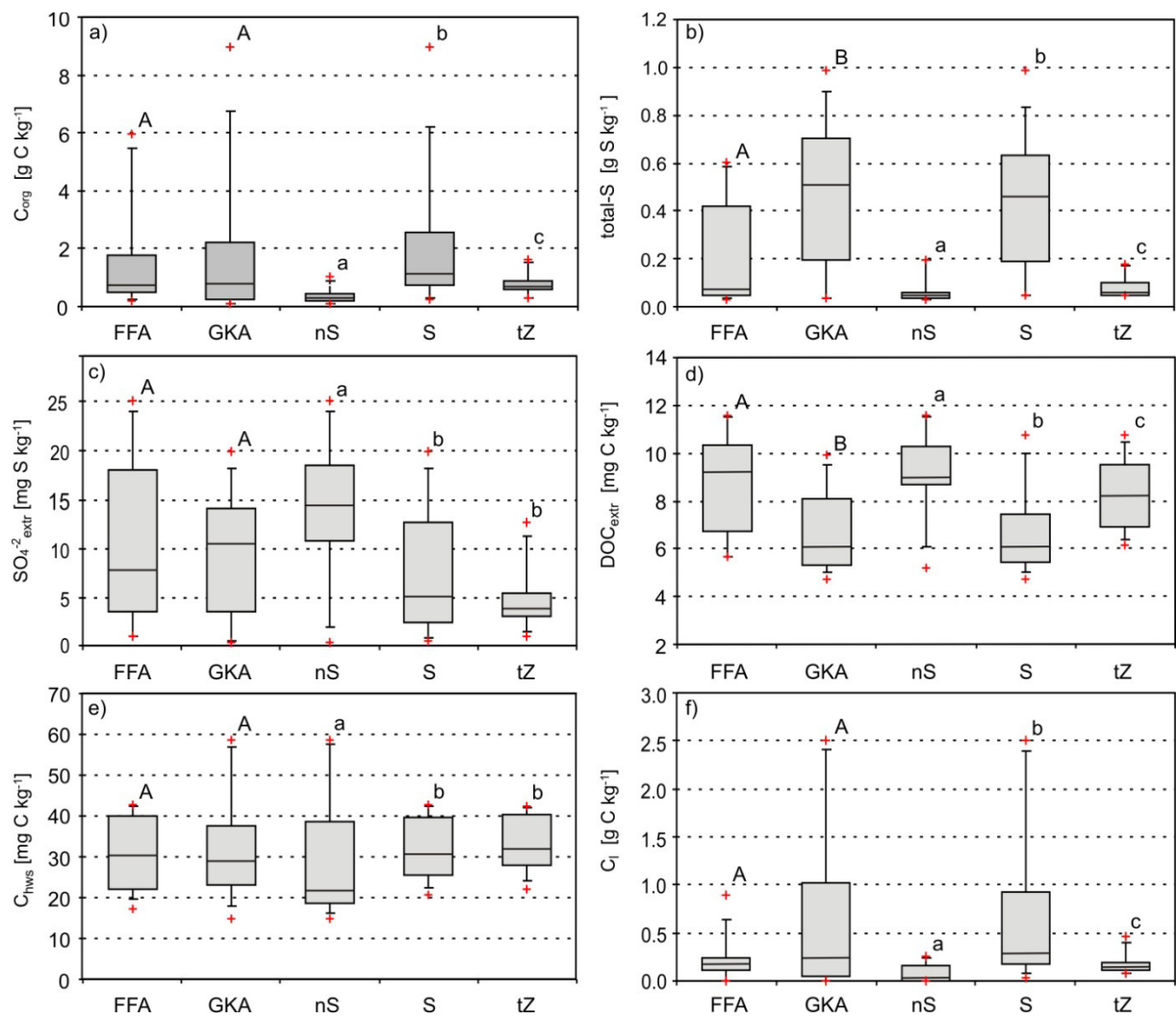


Fig. S2. Distribution of different sediment parameters in the aquifer material from the Fuhrberger Feld aquifer (FFA) and the Großenkneten aquifer (GKA) and in the various established groups of aquifer material: a) organic carbon, b) total sulphur, c) extractable sulphate, d) extractable dissolved organic carbon, e) hot water soluble organic carbon, f) potassium permanganate labile organic carbon. n S, S and tZ indicate non-sulphidic -, sulphidic and transition zone aquifer material, respectively. Different uppercase letters above the box-plots indicate significant differences between FFA and GKA material, different small letters show significant differences between n S, S and tZ (Kruskal-Wallis-Test ($P < 0.05$)).

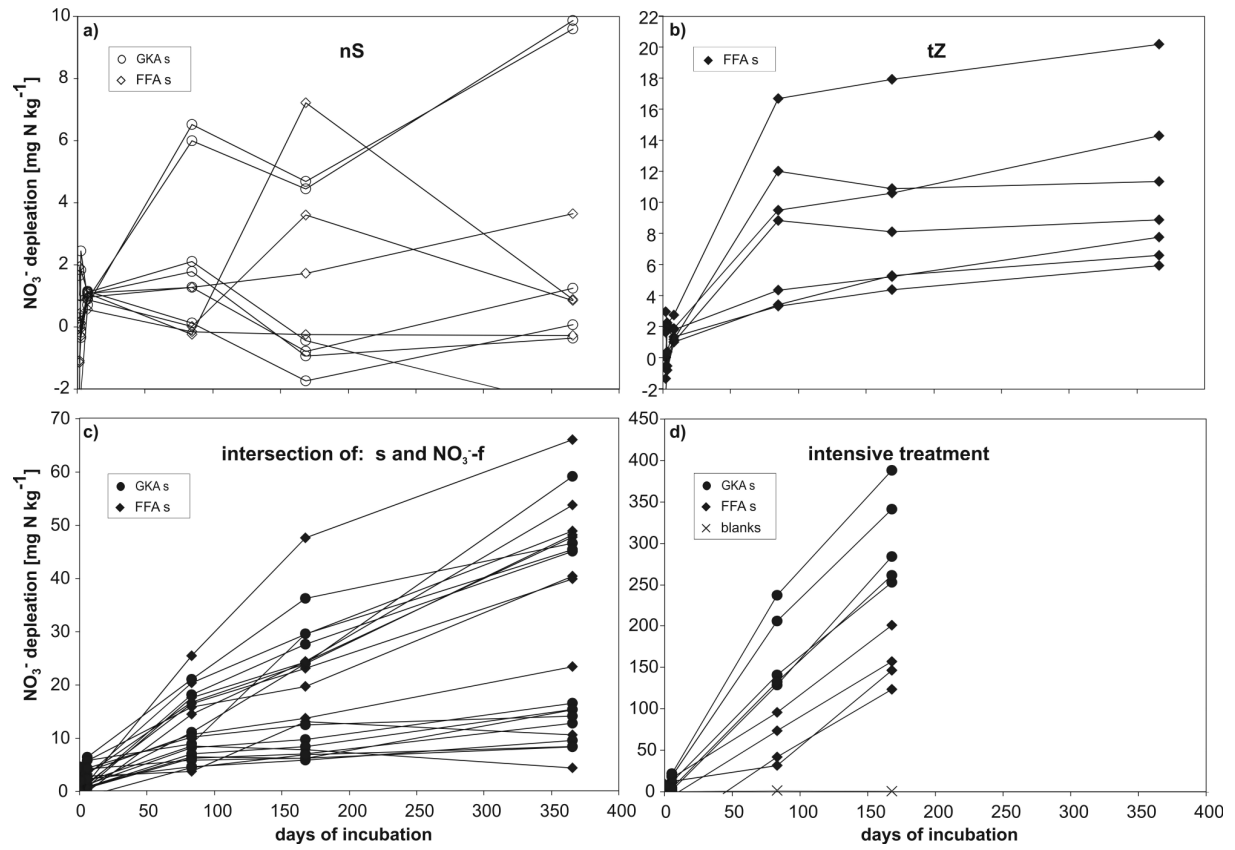


Fig. S3. Measured NO_3^- consumption during incubations. (The NO_3^- concentrations at the last sampling date of intensive incubations were not measured.)

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