

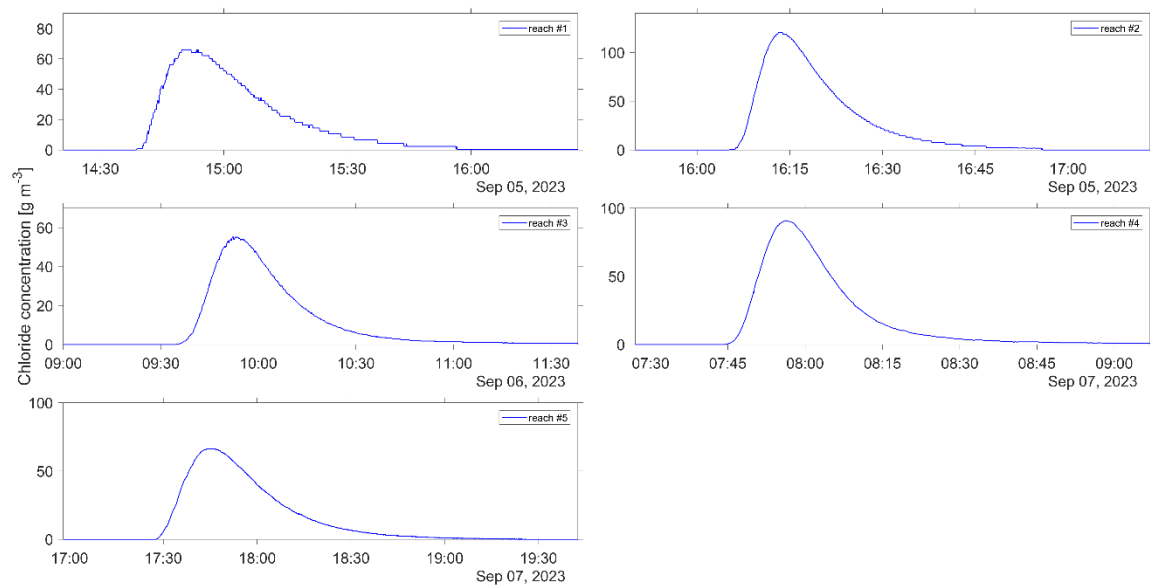


Figure S1: Breakthrough curves measured at the downstream end of each reach (reaches #1 - #5)

Table S1: Information content (top 10%) for reaches #2 - #5

Parameter	D	α	Q_{fix} A _{TS}	q _i	SUM	D	α	Q_{LHS} A _{TS}	q _i	SUM	D	α	Q_{out} A _{TS}	q _i	SUM
Reach #2															
$H(X)$	3.91	3.91	3.91	-		3.91	3.91	3.91	0.00		3.91	3.91	3.91	3.91	
$H(X chloride)$	1.48	3.81	3.77	-		1.54	3.81	3.75	0.00		1.76	3.81	3.74	2.49	
$H(X radon)$	3.87	3.77	1.92	-		3.90	3.90	3.84	0.00		3.89	3.90	3.80	0.95	
$H(X (radon \cap chloride))$	1.49	2.18	1.95	-		1.56	3.80	3.74	0.00		1.81	3.81	3.75	0.83	
$D_{KL}(X chloride, X)$	2.42	0.10	0.14	-	2.67	2.37	0.10	0.15	0.00	2.63	2.15	0.09	0.16	1.42	3.83
$D_{KL}(X radon, X)$	0.04	0.14	1.99	-	2.17	0.01	0.01	0.07	0.00	0.08	0.01	0.01	0.11	2.96	3.09
$D_{KL}\left(X (radon \cap chloride), X\right)$	2.42	1.73	1.96	-	6.10	2.35	0.11	0.16	0.00	2.62	2.09	0.10	0.16	3.08	5.43
Reach #3															
$H(X)$	3.91	3.91	3.91	-		3.91	3.91	3.91	0.00		3.91	3.91	3.91	3.91	
$H(X chloride)$	1.85	3.85	3.78	-		1.88	3.85	3.77	0.00		1.96	3.86	3.76	2.93	
$H(X radon)$	3.90	3.72	1.67	-		3.90	3.86	3.18	0.00		3.91	3.90	3.84	1.20	
$H(X (radon \cap chloride))$	1.76	2.14	1.65	-		1.81	3.39	3.56	0.00		1.82	3.85	3.80	0.80	
$D_{KL}(X chloride, X)$	2.05	0.06	0.13	-	2.24	2.03	0.06	0.14	0.00	2.22	1.95	0.04	0.15	0.98	3.12
$D_{KL}(X radon, X)$	0.01	0.18	2.24	-	2.43	0.01	0.04	0.73	0.00	0.78	0.00	0.00	0.07	2.71	2.78
$D_{KL}\left(X (radon \cap chloride), X\right)$	2.15	1.77	2.25	-	6.17	2.10	0.51	0.34	0.00	2.96	2.09	0.05	0.10	3.11	5.35
Reach #4															
$H(X)$	3.91	3.91	3.91	-		3.91	3.91	3.91	0.00		3.91	3.91	3.91	3.91	
$H(X chloride)$	1.99	3.86	3.75	-		1.67	3.85	3.81	0.00		1.71	3.86	3.80	3.90	
$H(X radon)$	3.89	3.70	1.61	-		3.89	3.84	2.98	0.00		3.90	3.89	3.76	2.46	
$H(X (radon \cap chloride))$	1.81	2.28	1.55	-		1.67	3.35	3.39	0.00		1.64	3.78	3.83	1.91	
$D_{KL}(X chloride, X)$	1.92	0.05	0.16	-	2.13	2.24	0.05	0.10	0.00	2.39	2.20	0.05	0.11	0.00	2.36
$D_{KL}(X radon, X)$	0.02	0.21	2.29	-	2.52	0.01	0.07	0.93	0.00	1.01	0.00	0.01	0.15	1.45	1.61
$D_{KL}\left(X (radon \cap chloride), X\right)$	2.10	1.62	2.36	-	6.08	2.24	0.56	0.52	0.00	3.31	2.27	0.13	0.08	2.00	4.48
Reach #5															
$H(X)^b$	3.91	3.91	3.91	-		3.91	3.91	3.91	0.00		3.91	3.91	3.91	3.91	
$H(X chloride)$	1.77	3.86	3.77	-		1.80	3.86	3.77	0.00		1.74	3.87	3.73	1.89	
$H(X radon)$	3.91	3.77	1.71	-		3.90	3.90	3.78	0.00		3.90	3.91	3.57	0.79	
$H(X (radon \cap chloride))$	1.64	2.54	1.93	-		1.81	3.89	3.67	0.00		1.69	3.89	3.46	0.78	
$D_{KL}(X chloride, X)$	2.14	0.05	0.14	-	2.32	2.11	0.05	0.14	0.00	2.29	2.17	0.04	0.17	2.01	4.40
$D_{KL}(X radon, X)$	0.00	0.14	2.20	-	2.34	0.00	0.00	0.12	0.00	0.13	0.00	0.00	0.34	3.11	3.45
$D_{KL}\left(X (radon \cap chloride), X\right)$	2.27	1.37	1.98	-	5.61	2.10	0.02	0.24	0.00	2.36	2.21	0.01	0.45	3.12	5.80

Table S2: Overview of the sensitivity based on the K-S test for all model parameters (D , α , A_{TS} , and the groundwater inflow (q_i)) for calibrating OTIS-R with chloride and radon. Results are presented for the top 10%

Reach	Parameter	Q_{fix}		Q_{LHS}		Q_{out}	
		chloride	radon	chloride	radon	chloride	radon
Reach #1	D	high	slight	high	slight	high	slight
	α	moderate	moderate	slight	slight	slight	slight
	A_{TS}	high	high	moderate	high	moderate	moderate
	q_i					moderate	high
Reach #2	D	high	slight	high	slight	high	slight
	α	high	moderate	high	slight	moderate	slight
	A_{TS}	high	high	high	moderate	high	moderate
	q_i					moderate	high
Reach #3	D	high	slight	high	slight	high	slight
	α	insensitive	moderate	moderate	slight	moderate	slight
	A_{TS}	moderate	high	moderate	high	high	slight
	q_i					slight	high
Reach #4	D	high	slight	high	slight	high	slight
	α	moderate	moderate	moderate	slight	moderate	slight
	A_{TS}	moderate	high	moderate	high	moderate	insensitive
	q_i					slight	high
Reach #5	D	high	slight	high	slight	high	slight
	α	moderate	moderate	moderate	slight	moderate	slight
	A_{TS}	moderate	high	moderate	moderate	high	high
	q_i					slight	high

Table S3: Spearman rank correlation coefficients for all parameters across all reaches. Only the top 1% of results are shown. Statistically significant correlations are marked with an asterisk (*)

Reach #1		chloride			radon (k _{low})	
Parameter	Q _{fix}	Q _{LHS}	Q _{out}	Q _{fix}	Q _{LHS}	Q _{out}
D - α	0.57*	0.55*	0.52*	0.01	0.04	-0.01
D - A _{TS}	-0.10*	-0.10*	-0.02	-0.01	-0.003	-0.05*
D - q _I		-0.10*	-0.11*		-0.02	0.07*
α - A _{TS}	-0.26*	-0.27*	-0.23*	-0.91*	-0.22*	-0.08*
α - q _I		-0.04	0.03		-0.07*	-0.04
A _{TS} - q _I		-0.05	-0.26*		-0.86*	-0.84*
Reach #2		chloride			radon (k _{low})	
D - α	0.12*	0.07*	0.04	-0.04	-0.04	-0.01
D - A _{TS}	-0.16*	-0.05	-0.03	0.02	0.03	0.11*
D - q _I		-0.25*	-0.21*		-0.05	-0.16*
α - A _{TS}	-0.56*	-0.51*	-0.45*	-0.92*	-0.14*	-0.09*
α - q _I		0.13*	0.04		0.08*	0.10*
A _{TS} - q _I		-0.22*	-0.11*		-0.70*	-0.61*
Reach #3		chloride			radon (k _{low})	
D - α	0.62*	0.58*	0.43*	-0.01	-0.03	-0.004
D - A _{TS}	-0.38*	-0.29*	-0.02	-0.03	0.01	0.10*
D - q _I		-0.03	-0.32*		-0.005	-0.18*
α - A _{TS}	-0.47*	-0.40*	-0.22*	-0.91*	-0.39*	-0.10*
α - q _I		0.04	0.05		-0.13*	0.05*
A _{TS} - q _I		-0.17*	-0.52*		-0.72*	-0.77*
Reach #4		chloride			radon (k _{low})	
D - α	0.52*	0.52*	0.39*	0.02	0.02	0.01
D - A _{TS}	-0.24*	-0.28*	-0.10*	-0.01	-0.01	-0.03
D - q _I		-0.03	-0.28*		0.01	0.04
α - A _{TS}	-0.33*	-0.41*	-0.27*	-0.91*	-0.50*	-0.11*
α - q _I		-0.01	-0.10*		-0.15*	-0.02
A _{TS} - q _I		-0.12*	-0.34*		-0.65*	-0.86*
Reach #5		chloride			radon (k _{low})	
D - α	0.53*	0.53*	0.40*	0.004	0.005	-0.02
D - A _{TS}	-0.16*	-0.10*	0.08*	0.21*	-0.005	0.28*
D - q _I		-0.14*	-0.29*		0.17*	0.30*
α - A _{TS}	-0.32*	-0.30*	-0.12*	-0.87*	-0.09*	-0.05
α - q _I		-0.01	-0.01		0.05	0.01
A _{TS} - q _I		-0.21*	-0.37*		-0.53*	-0.39*