Supplementary Information (SI) for Environmental Science: Processes & Impacts.

Supplementary

Continental fluxes of rare earth elements to coastal ecosystems across a wide geographic region varying in geology and climate

Table S1: Data quality validation for rare earth elements the St. Lawrence River for the 4 years of sampling (2017-2020): limits of detection (LoD), field blanks and recoveries of certified data.

REE	LoD (nM)	Number of samples under LoD (%)	Mean [REE] in field blank (nM)	Mean Recovery of SLRS-6[1] (%)
		n = 213	n = 19	n = 23
La	0.005	0	0.02	113
Ce	0.004	0	0.04	112
Pr	0.01	0.9	0.01	116
Nd	0.01	5	0.02	115
Sm	0.01	18	< LoD	111
Gd	0.01	12	< LoD	112
Tb	0.006	56	< LoD	110
Dy	0.006	16	< LoD	108
Ho	0.004	46	< LoD	111
Er	0.006	14	< LoD	110
Tm	0.006	86	< LoD	109
Yb	0.006	6	< LoD	109
Lu	0.006	75	< LoD	101

[1]Data cross-validated using reference values from CEAEQ, which routinely analyzes SLRS-6 under standardized conditions.

Table S2: Data quality validation for rare earth elements in the George and Koroc rivers for the 5 years of sampling (2019-2023): limits of detection (LoD), field blanks and recoveries of certified data.

REE	LoD ($ug \cdot L^{-1}$)	Mean [REE] in field blank (nM)[1]	Mean Recovery of SLRS-6 (%)
		n = 24	n = 8
La	0.0004	0.0022	110
Ce	0.0005	0.0033	108
Pr	0.0006	< LoD	111
Nd	0.0008	0.0015	109
Sm	0.001	< LoD	105
Gd	0.0008	< LoD	106
Tb	0.006	< LoD	100

Dy	0.001	< LoD	101
Ho	0.0006	0.0006	105
Er	0.001	< LoD	102
Tm	0.0008	< LoD	102
Yb	0.001	< LoD	97
Lu	0.0004	< LoD	98

[1]Only 11% of the 135 measured REEs concentrations were above the LoD

Table S3: Data quality validation for rare earth elements Hudson Bay rivers for the 3 years of sampling: limits of detection, field blanks and recoveries of certified data.

REE	LoD ($\mu g \cdot L^{-1}$)	Mean [REE] in field blank (nM)[1]	Mean Recovery of SLRS-6 (%)[1]
		n = 22	n = 22
La	0.0013	0.0021	110
Ce	0.0013	0.0036	106
Pr	0.0010	< LoD	107
Nd	0.0025	< LoD	107
Sm	0.0028	< LoD	103
Gd	0.0032	< LoD	107
Tb	0.0016	< LoD	103
Dy	0.0028	< LoD	101
Ho	0.0009	< LoD	108
Er	0.0019	< LoD	102
Tm	0.00013	< LoD	105
Yb	0.0032	< LoD	76
Lu	0.0013	< LoD	97

[1]SLRS-6 % recovery values are based on a comparison between our measurements and values reported in Yeghicheyan et al. (2017).

Table S4: Multiple linear regression model outputs to best predict the relative proportion (%) and the concentrations of filtered REEs measured in water of 40 rivers from the Province of Quebec

Variable	N	Coefficient	R^2	*p-value*	AIC
% filtered REEs	40				
Intercept		1.999	-	9.863e-09	-
pH		-0.172	0.50	4.485e-04	-17.126
Temperature (°C)		-0.021	**0.57**	1.273e-02	-21.925
Filtered concentrations of REEs	40				
Intercept		2.620	-	2.975e-09	-
DOC		0.112	0.66	8.999e-12	67.240
pH		-0.355	**0.74**	1.656e-03	58.426

Bold R^2 values represent the final selected model R^2.

Table S5. Fluxes[1], standard error (SE) and yields[1] of REEs in sampled rivers from the Province of Quebec

River	n[1]	Filtered Flux	SE	Total Flux	SE	Gd Flux	%Gd_{ant}	Filtered Yield	Total Yield
		Metric tons · $year^{-1}$		Metric tons · $year^{-1}$		kg · $year^{-1}$	%	kg · m^2 · $year^{-1}$	
Atlantic watershed - North									
Assomption	2	0.21	0.12	1.59	0.46	13.14	0	0.07	0.37
Batiscan	2	3.43	0.92	5.98	1.49	115.69	0	0.73	1.28
Bayonne[2]	1	0.03	0.08	0.31	0.02	1.24	12.7	0.09	0.87
Du Loup	2	0.81	0.12	4.08	0.99	24.40	0	0.50	2.54
La Romaine	2	9.19	1.42	-	-	171.77	0	0.64	-
Maskinongé	2	0.47	0.27	1.52	0.98	17.05	0	0.43	1.37
Ottawa	2	60.48	5.06	137.32	21.90	1957.28	0	0.41	0.94
Sainte-Anne	1	1.58	1.07	2.91	0.14	61.11	0	0.59	1.07
Saint-Maurice	4	27.55	4.06	42.44	5.17	817.67	0	0.64	0.98
St. Lawrence Downstream	4	29.38	5.16	739.67	101.89	707.70	0	0.02	0.55
Yamachiche[2]	2	0.16	0.02	0.69	0.28	4.98	0	0.61	2.51
Atlantic watershed – South									
Boyer	1	0.04	0.03	3.83	0.20	1.37	0	0.18	17.66
Bécancour	2	0.46	0.40	4.01	3.75	21.23	0	0.17	1.53
Châteauguay	1	0.05	0.01	0.46	0.04	7.07	49.7	0.02	0.19
Chaudière	1	0.39	0.01	1.11	0.08	34.62	54.8	0.06	0.17
Nicolet	2	0.51	0.44	13.50	12.50	20.08	0	0.15	3.99
Richelieu	2	1.28	0.28	24.12	4.40	66.75	18.7	0.05	1.02
Saint-François	2	1.09	0.74	4.46	2.01	66.98	24.4	0.11	0.44
Saint-Régis	1	0.20	-	1.92	-	138.89	97.8	2.11	20.69
Yamaska	2	0.39	0.22	12.02	2.79	24.25	49.7	0.08	2.51
Hudson Bay watershed									
Aquatuc	2	0.21	0.03	0.29	0.03	4.19	0	0.12	1.52

Beaver	2	0.64	0.07	0.97	0.18	15.50	0	0.22	0.33
Broadback	2	14.49	2.60	31.36	4.95	388.35	0	0.74	1.51
Caillet	2	0.45	0.07	0.72	0.06	9.72	0	0.95	1.53
ChinuSAW	2	0.02	0.001	0.05	0.02	0.47	0	0.50	1.19
Conn	2	0.74	0.08	1.14	0.18	15.67	0	1.07	1.65
Eastmain	2	3.41	0.51	6.00	1.27	85.79	0	0.86	1.51
Guillaume	2	1.34	0.22	1.91	0.22	26.36	0	1.13	1.61
Harricana	2	15.34	1.10	42.11	15.05	427.05	0	0.52	1.43
Jolicoeur	2	0.69	0.07	1.21	0.37	18.34	0	0.39	0.68
La Grande	2	54.68	5.40	84.03	15.89	1346.21	0	0.26	0.40
Maquata	2	0.46	0.03	0.67	0.17	11.92	0	0.44	0.65
Nottaway	2	61.95	6.64	111.02	25.72	1618.97	0	1.00	1.79
Old Factory	2	1.55	0.07	2.16	0.23	38.72	0	0.57	0.79
Pontax	2	5.83	0.87	15.26	5.14	141.63	0	0.74	1.93
Rupert	2	4.97	0.69	12.65	4.01	130.81	0	0.43	1.09
Salmon	2	0.004	0.001	0.008	0.002	0.11	0	0.05	0.12
Seal	2	0.55	0.16	0.95	0.14	12.53	0	0.35	0.60
Ungava Bay watershed									
George River	5	16.37	2.78	20.28	4.47	331.27	0	0.39	0.49
Koroc River	3	2.38	0.07	5.57	0.38	11.67	0	0.24	0.56

[1]Data represent mean discharge and mean concentrations based on triplicate samples collected across multiple years (n = number of years sampled)

[2]These rivers have been classified in the South of St. Lawrence subgroup since they exclusively drain the St. Lawrence lowlands by contrast to the others that also drain the Canadian Shield geology

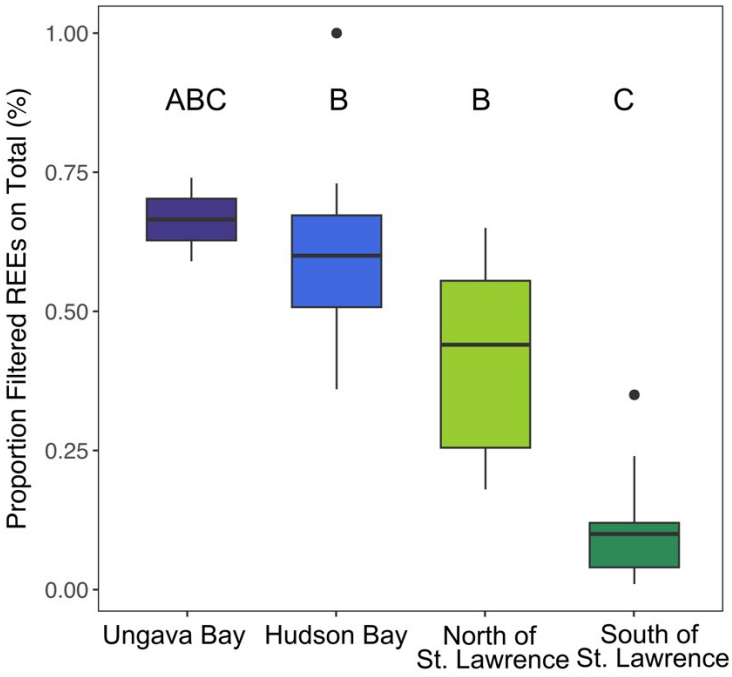

Figure S1. Proportion of filtered rare earth elements (REEs) relative to the total concentrations in rivers from the different watersheds of the Province of Quebec.

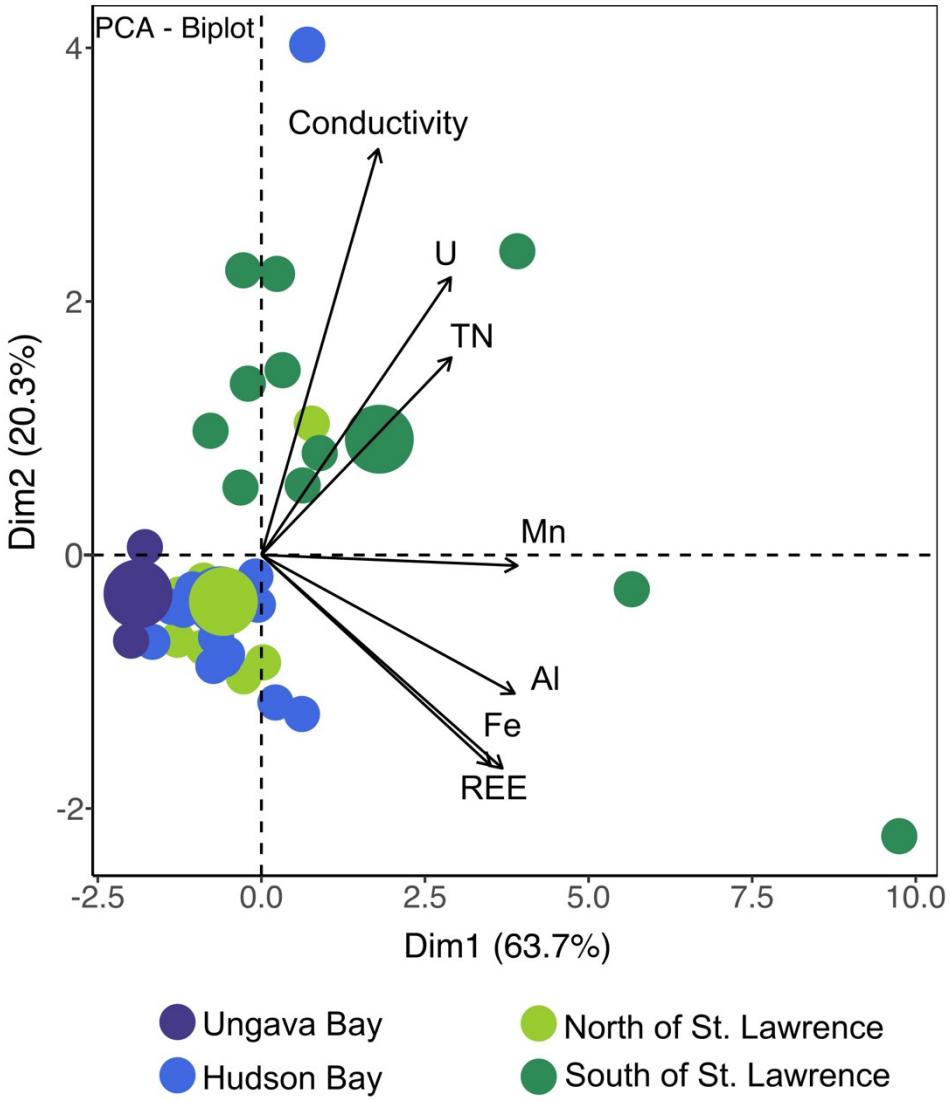

Figure S2. Principal component analysis in type 2 framing with the total concentrations of elements in rivers draining the different watersheds of the Province of Quebec. Larger circles represent the center of each group distribution.

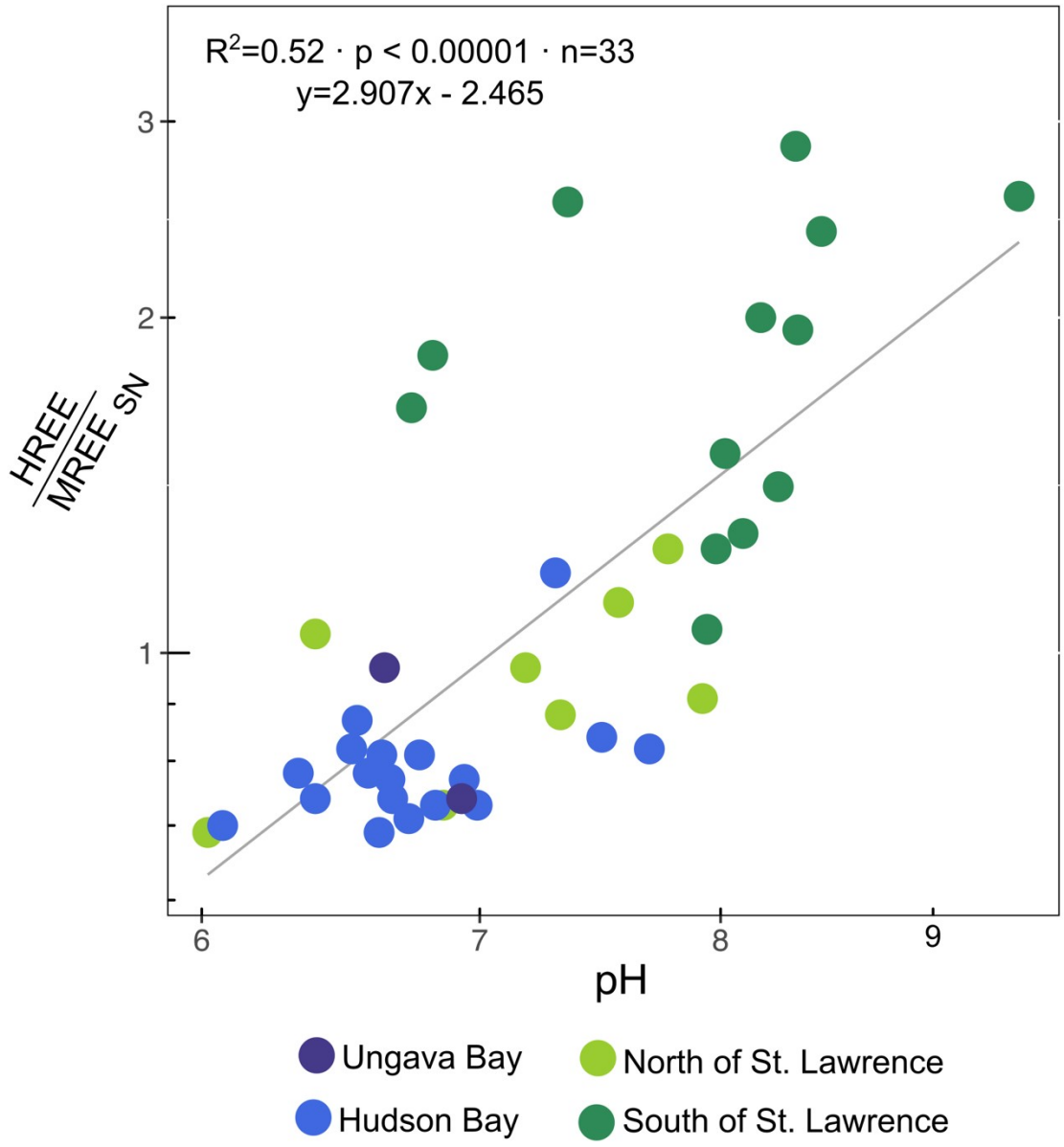

Figure S3. Standardized ratio of heavy versus middle rare earth elements $(HREEs/MREEs)_{SN}$ according to pH in rivers draining the different watersheds of the Province of Quebec.

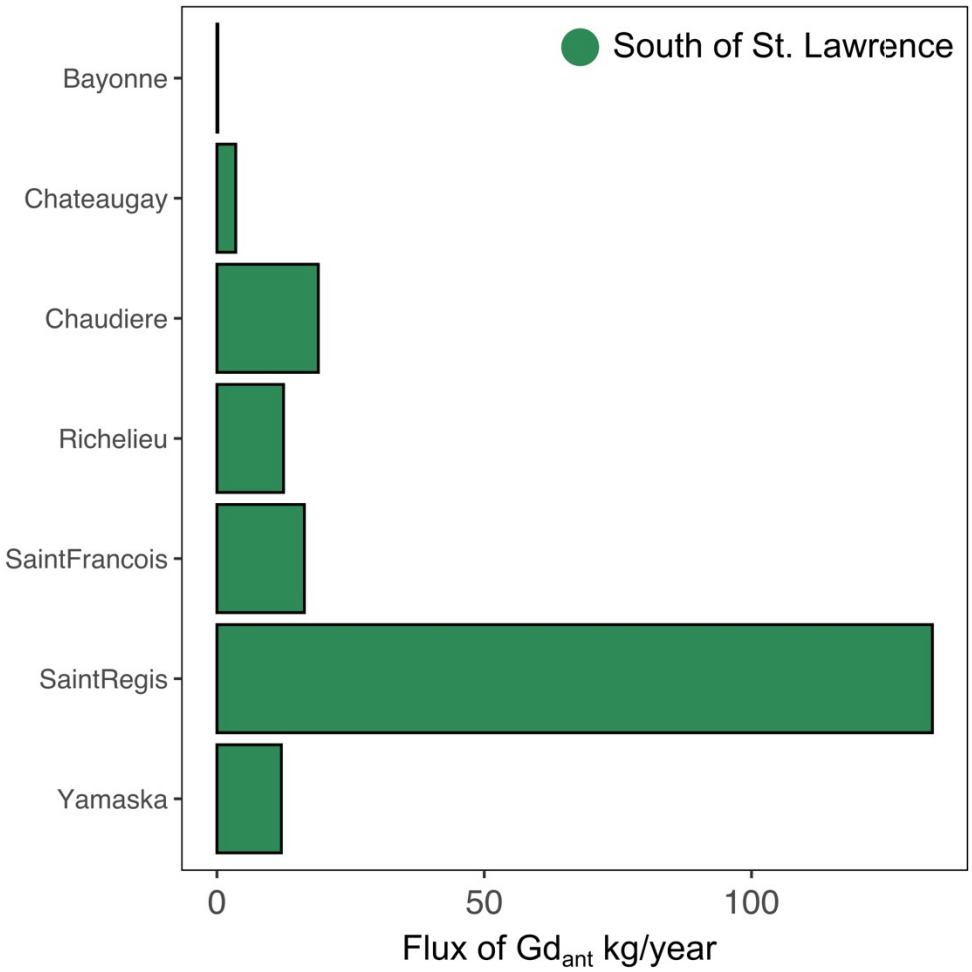

Figure S4. Fluxes of filtered gadolinium from anthropogenic origin (Gd_{ant}) in sampled rivers from the Province of Quebec. Anthropogenic Gd fluxes were calculated when the gadolinium anomaly (Gd/Gd*) was above 1.4.

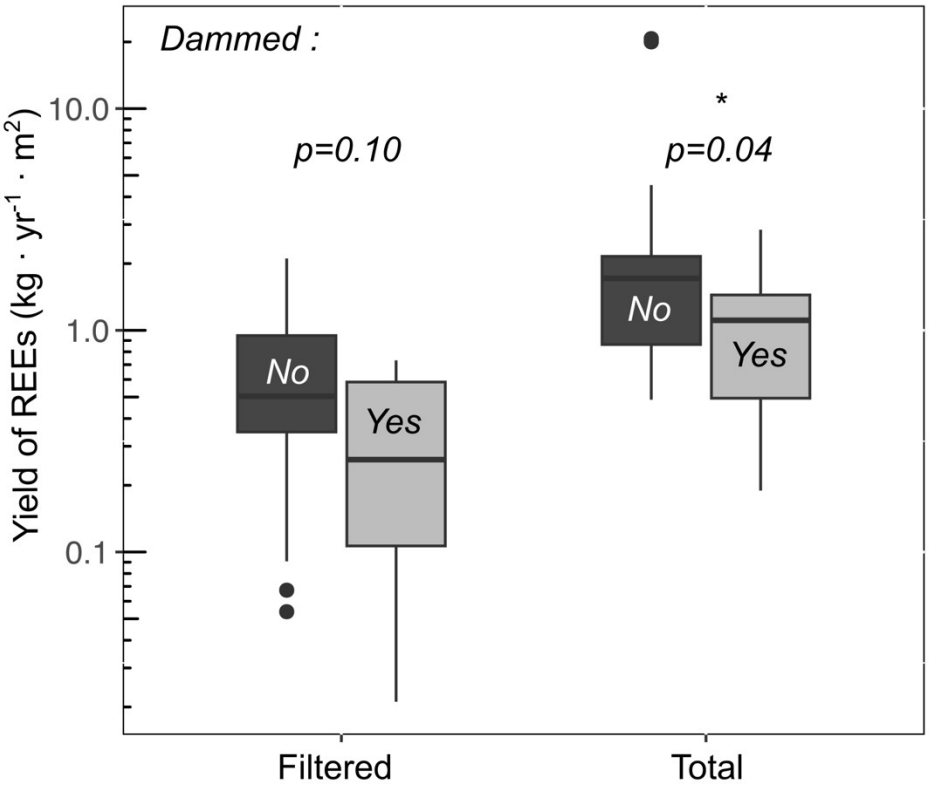

Figure S5. Yields of filtered and total rare earth elements (REEs) in dammed (yes) and undammed (no) rivers from the Province of Quebec. Dammed rivers (n=13) included mostly rivers draining the South of the province: Batiscan, Becancour, Chateauguay, Chaudiere, du Loup, La Romaine, Nicolet, Ottawa, Sainte-Anne, Saint-François, Saint-Maurice and Yamaska, in addition to La Grande River draining the Hudson Bay watershed. Undammed rivers included 26 rivers. Dammed rivers were characterized by the presence of one or several dams along their flow with a high impounding capacity according to the Quebec law on dam security.

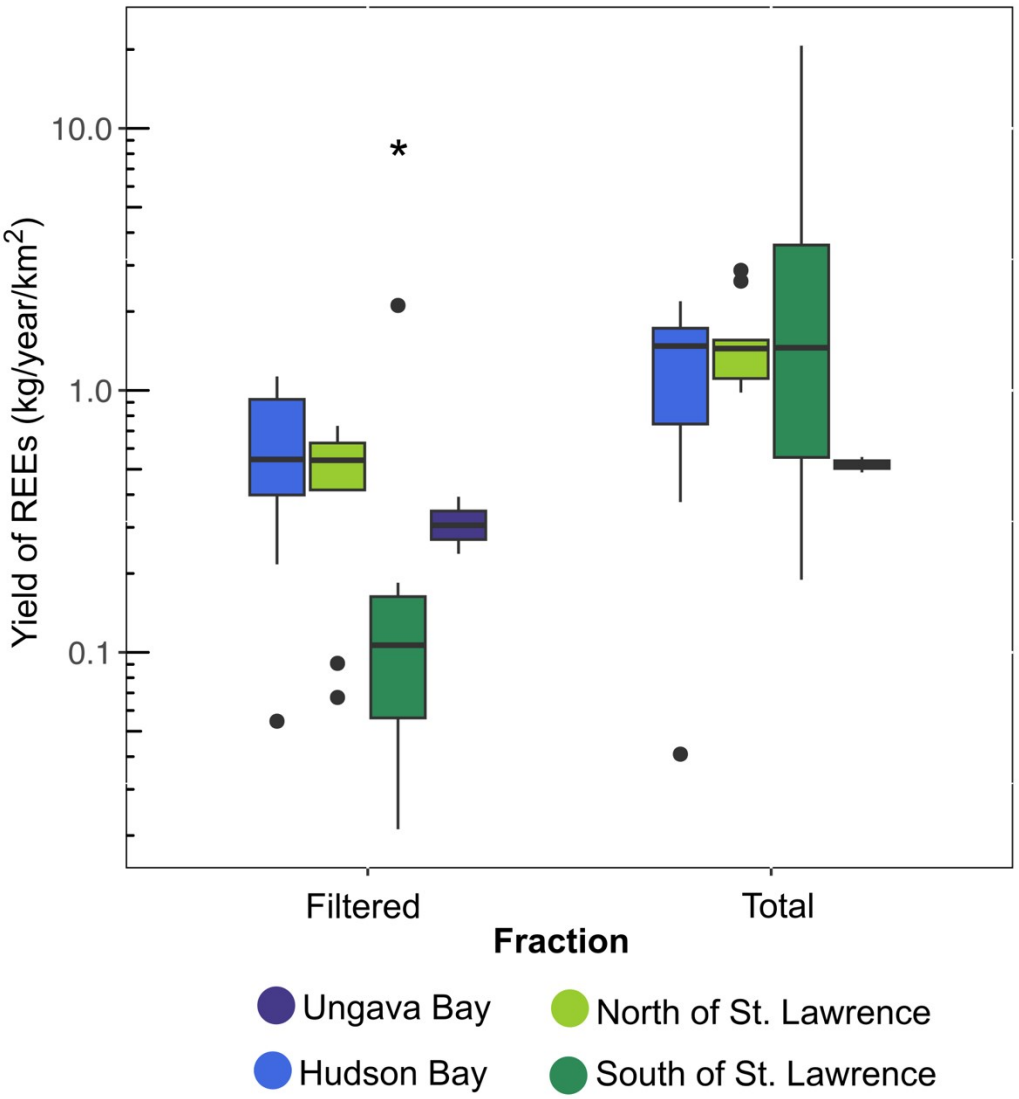

Figure S6. Yields of filtered and total concentrations of rare earth elements (REEs) in rivers from different watersheds in the Province of Québec. Significant difference is marked by an asterix (*) ($p = 0.01$).

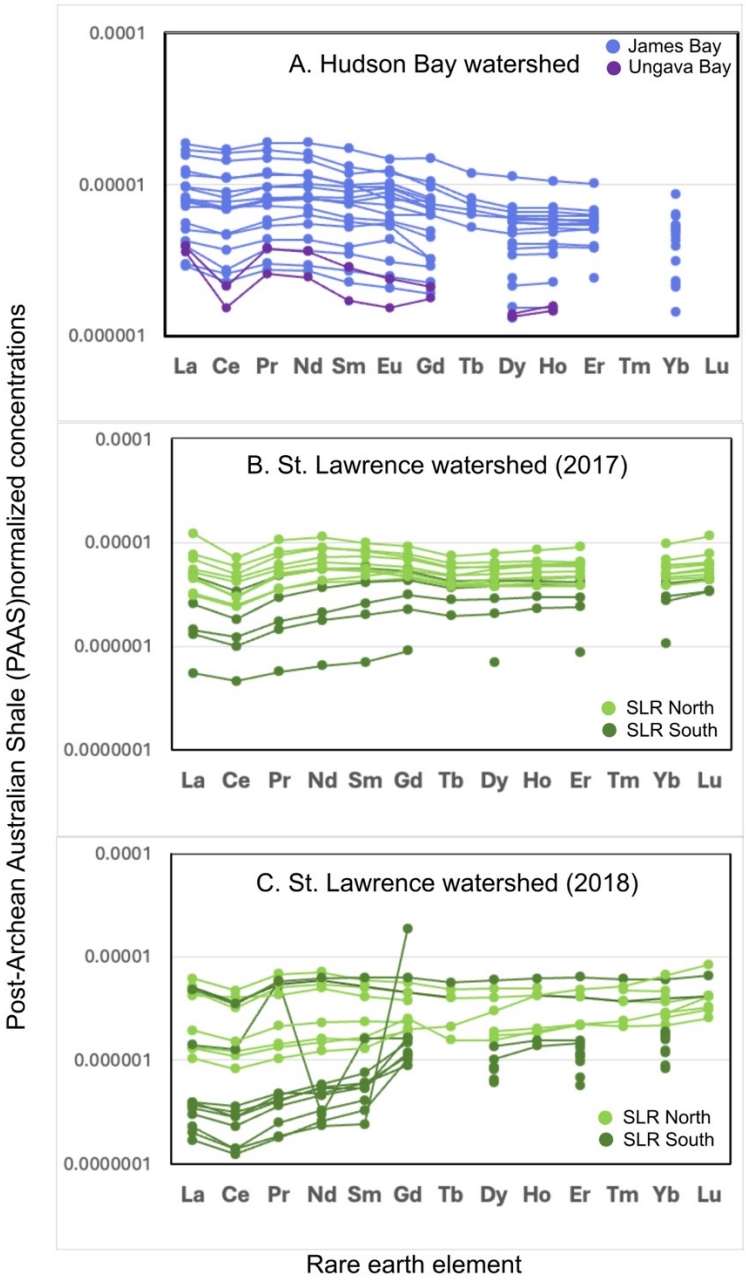

Figure S7. Normalized concentrations of filtered (0.45 µm) rare earth elements (REEs), based on Post-Archean Australian Shale (PAAS) values, in tributary waters from major watersheds across the province of Québec. Normalized data for the St. Lawrence River watershed are presented for 2017 (wet) and 2018 (dry), two years with contrasting precipitation regimes (Lafrenière et al., 2023).