

Supporting information

Selenium distribution and speciation in waters of pristine alpine lakes from central-western Pyrenees (France-Spain)

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Supporting information:

Number of Figures SI: 5

Number of Tables SI: 5

Figure SI 1. Example chromatogram of Nère subsurface water sample of June 2017 without (blue line) and with selenite and selenate standard additions (orange line: 5 ng L⁻¹ selenite and selenate additions). Concentrations of compounds were: < LoQ, 4.4 ± 0.3 ng Se L⁻¹ for Se(IV) and Se(VI) respectively.

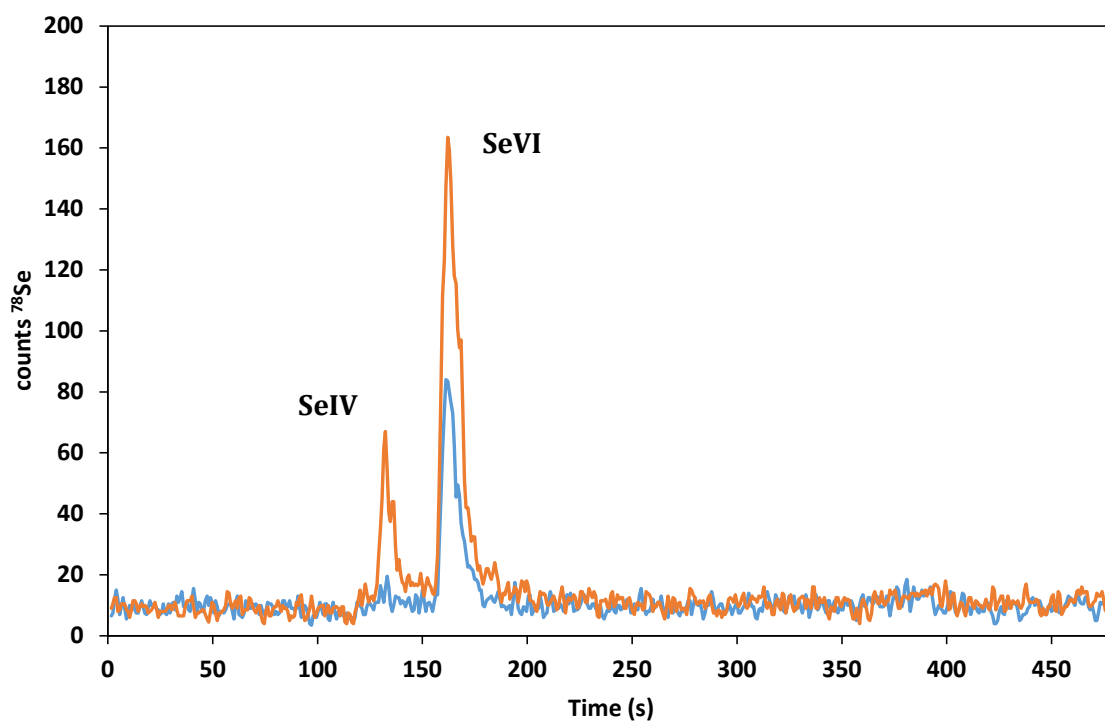


Figure SI 2. Bulk versus total dissolved selenium concentrations. Lake groups color code: pDe-GR in blue, De-SR in green, PT-SR in yellow and Cr-SR in red. Dotted line has slope 1.

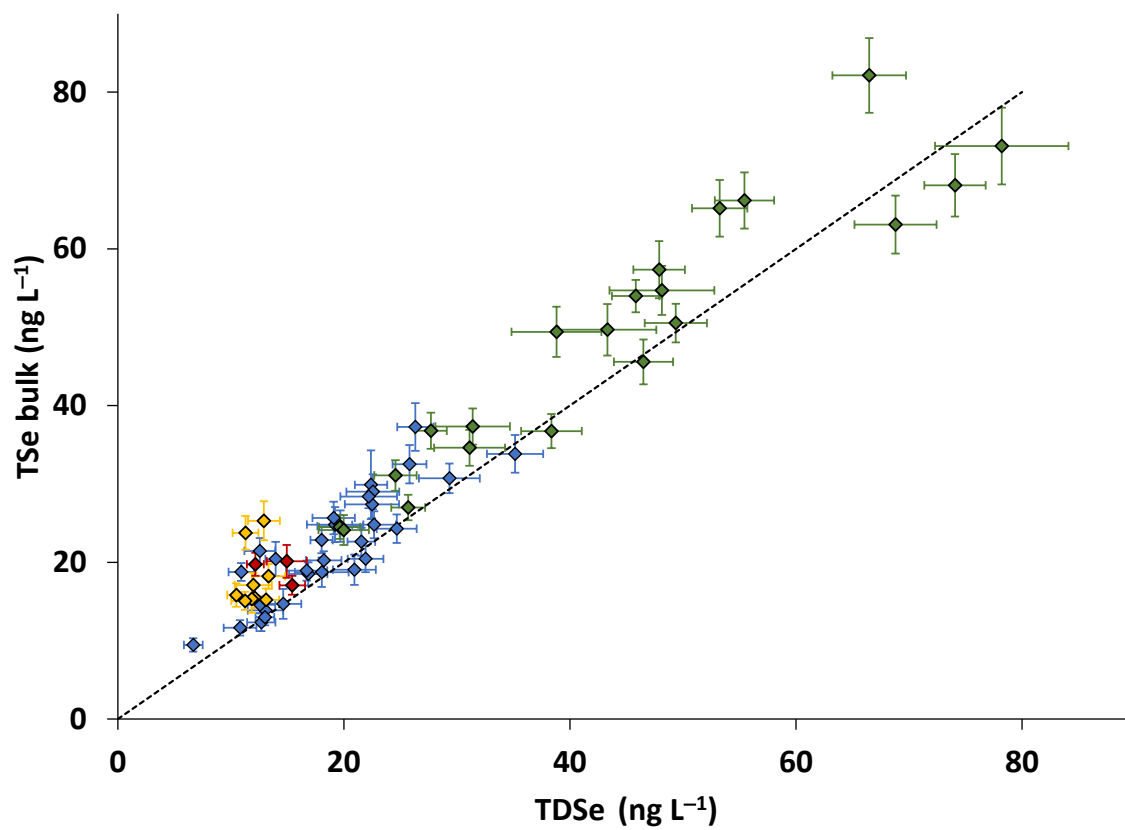


Figure SI 3. Total dissolved selenium concentration as a function of A) dissolved inorganic carbon concentration and B) sulphate concentration. Sabocos lake presenting outlier DIC values was excluded from correlations. Lake groups color code: pDe-GR in blue, De-SR in green, PT-SR in yellow and Cr-SR in red.

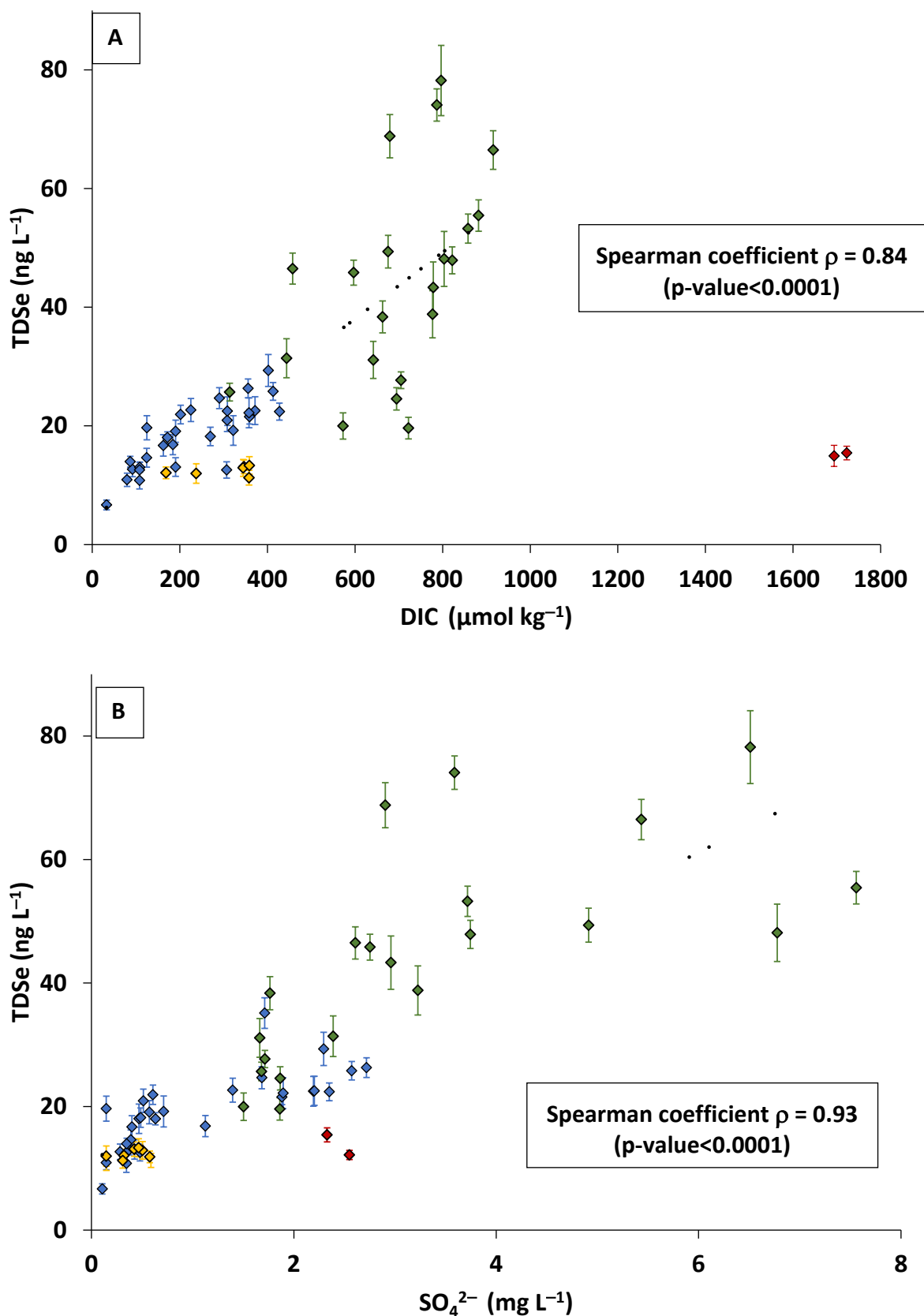


Figure SI 4. Selenate versus total dissolved selenium concentrations. Lake groups color code: pDe-GR in blue, De-SR in green, PT-SR in yellow and Cr-SR in red. Dotted line has slope 1.

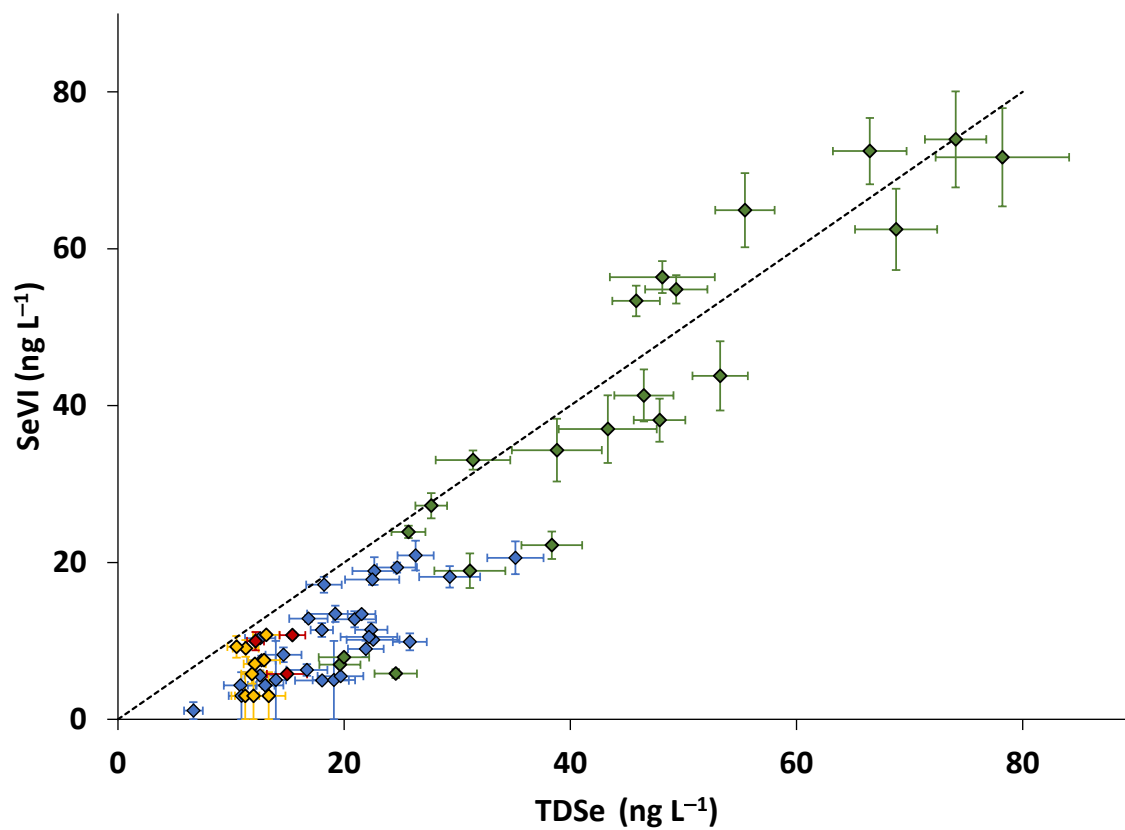


Figure SI 5. Depth profiles obtained from lakes Arratille and Azul Superior (De-SR lakes): A) temperature ($^{\circ}\text{C}$) and SO_4^{2-} (mg L^{-1}) (top axis), Conductivity (Cond.; $\mu\text{S cm}^{-1}$), oxygen saturation (Ox. Sat. in %) and NO_3^- (10^{-2}mg L^{-1}) (bottom axis); B) non-volatile Se speciation and total Se in ng Se L^{-1} ; C) dissolved total volatile Se and species in pg L^{-1} .

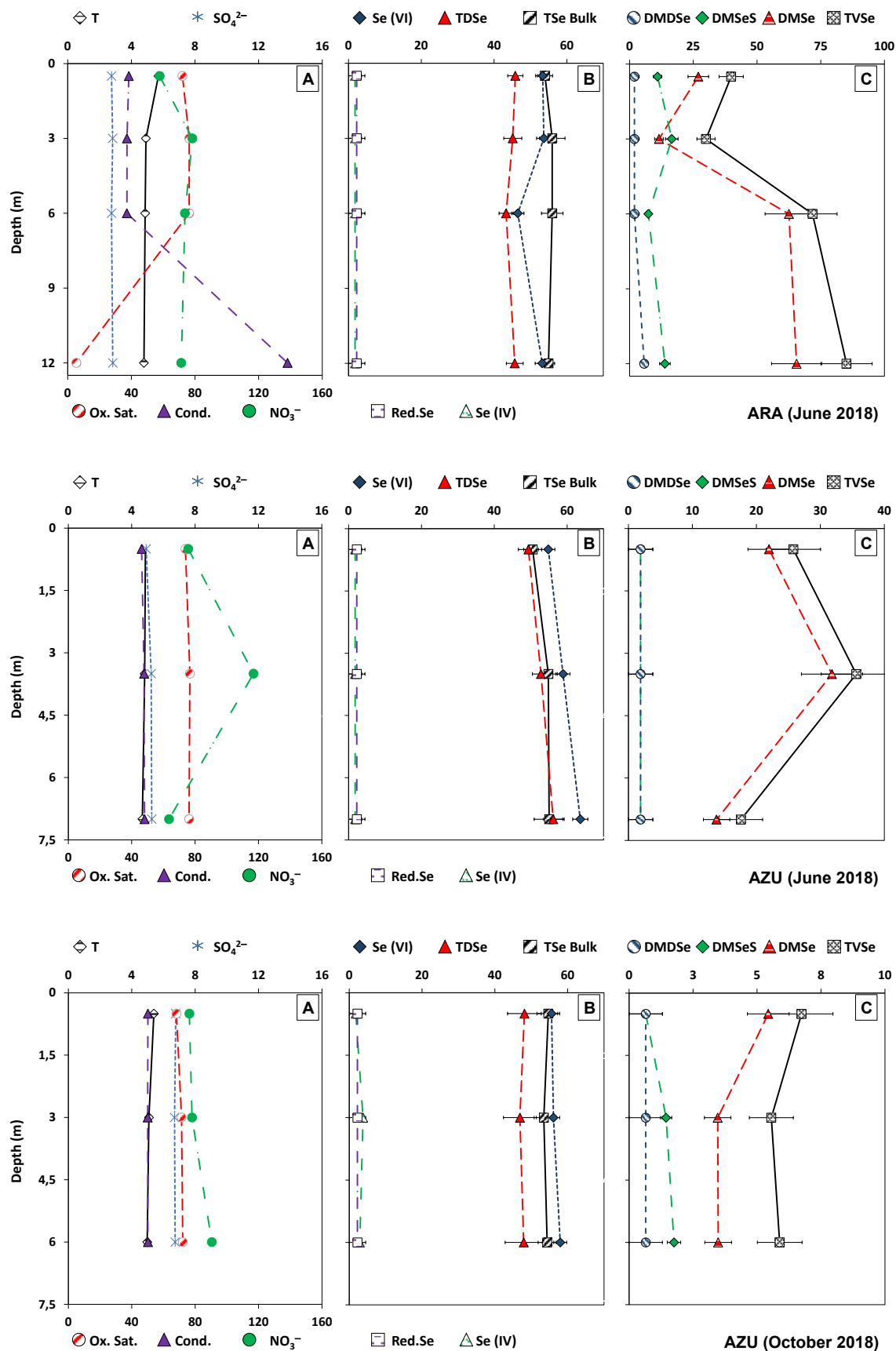


Table SI 1. Raw data for subsurface samples of main anions (Cl^- , NO_3^- , SO_4^{2-}), main physicochemical parameters (temperature (T), conductivity (Cond.), pH, redox potential (E_H), non-purgable organic carbon (NPOC) and dissolved inorganic carbon (DIC). LoD for measured anions were in the ranges: $(0.01\text{--}0.05)\cdot\text{mg L}^{-1}$ for Cl^- , $(0.01\text{--}0.14)\cdot\text{mg L}^{-1}$ for NO_3^- and $(0.03\text{--}0.30)\cdot\text{mg L}^{-1}$ SO_4^{2-} . *associated error corresponds to standard deviation of three replicated ($n=3$) points sampled/lake.

Lake	Cl^-	NO_3^- mg L^{-1}	SO_4^{2-}	T (° C)	Cond. ($\mu\text{S cm}^{-1}$)	pH	E (mV)	NPOC (mg C L^{-1})	DIC ($\mu\text{mol kg}^{-1}$)
pDe-GR lakes – June 2017									
Pey	$0.16 \pm 0.01^*$	$0.41 \pm 0.06^*$	$0.28 \pm 0.03^*$	5.45	17	$6.01 \pm 0.01^*$	208	$0.63 \pm 0.01^*$	$92 \pm 3^*$
Cam	$0.17 \pm 0.01^*$	$0.40 \pm 0.06^*$	$0.39 \pm 0.01^*$	8.71	8	$6.42 \pm 0.02^*$	160	$0.68 \pm 0.05^*$	$124 \pm 2^*$
Opa	0.16	$0.65 \pm 0.08^*$	$0.51 \pm 0.01^*$	-	-	$6.92 \pm 0.01^*$	-	$1.06 \pm 0.07^*$	$308 \pm 4^*$
Pou	$0.23 \pm 0.03^*$	$0.40 \pm 0.03^*$	$0.43 \pm 0.01^*$	9.46	12	$6.75 \pm 0.05^*$	202	$1.0 \pm 0.2^*$	$190 \pm 1^*$
Ner	$0.16 \pm 0.01^*$	$0.30 \pm 0.08^*$	$0.38 \pm 0.01^*$	11.13	7	$6.27 \pm 0.04^*$	119	$0.78 \pm 0.01^*$	$109 \pm 2^*$
Arn	$0.17 \pm 0.01^*$	$0.66 \pm 0.01^*$	$1.40 \pm 0.02^*$	8.43	18	$6.83 \pm 0.02^*$	122	$1.0 \pm 0.1^*$	$225 \pm 1^*$
Coa	$0.15 \pm 0.01^*$	$0.21 \pm 0.02^*$	$0.47 \pm 0.02^*$	7.11	23	6.42	97	$1.02 \pm 0.05^*$	134
Xua	$0.20 \pm 0.04^*$	$0.90 \pm 0.06^*$	$0.61 \pm 0.03^*$	4.63	12	$6.68 \pm 0.02^*$	110	$0.85 \pm 0.05^*$	$202 \pm 5^*$
Pan	$0.33 \pm 0.09^*$	$0.7 \pm 0.1^*$	$2.3 \pm 0.1^*$	10.29	27	7.02	195	$1.5 \pm 0.2^*$	402
Bac	$0.23 \pm 0.01^*$	$0.7 \pm 0.1^*$	$1.71 \pm 0.04^*$	-	-	$6.94 \pm 0.02^*$	-	$1.23 \pm 0.09^*$	$310 \pm 3^*$
pDe-GR lakes – June 2018									
Pey	0.07	0.10	<LoD	6.41	16	4.87	36	0.84	33
Cam	0.13	0.34	0.35	2.43	5	5.80	172	0.85	108
Opa	0.17	0.56	0.49	3.29	15	6.48	91	0.71	270
Arn	0.19	0.62	1.13	7.45	13	6.33	200	0.77	184
Coa	0.24	0.41	0.35	10.09	7	5.85	169	1.03	108
Pan	0.27	0.71	1.88	11.18	28	6.67	176	1.01	359
Bac	0.16	0.57	1.68	12.45	25	6.55	129	0.94	290

Table SI 1. (Continued)

Lake	Cl ⁻	NO ₃ ⁻	SO ₄ ²⁻	T	Cond.	pH	E	NPOC	DIC
pDe-GR lakes – June 2019									
Arn	0.10	0.79	1.27	-	-	-	-	5.33	-
Coa	0.12	0.56	0.39	-	-	-	-	1.31	-
Pan	0.36	0.55	2.70	-	-	-	-	1.41	-
Bac	0.13	1.08	1.33	-	-	-	-	1.53	-
pDe-GR lakes – October 2017									
Pey	0.17 ± 0.01*	<LoD*	0.35 ± 0.03*	9.57	7	6.00	108	1.40	87
Cam	0.19 ± 0.01*	0.21 ± 0.01*	0.63 ± 0.02*	9.71	14	6.65 ± 0.03*	121	1.27 ± 0.22*	172 ± 2*
Opa	0.16 ± 0.01*	0.58 ± 0.04*	0.71 ± 0.02*	9.89	25	6.93 ± 0.04*	141	1.32 ± 0.06*	322 ± 3*
Arn	0.19 ± 0.01*	0.66 ± 0.02*	2.72 ± 0.07*	10.19	31	7.07 ± 0.03*	117	1.6 ± 0.3*	356 ± 3*
Coa	0.20 ± 0.01*	<LoD*	0.57 ± 0.03*	11.08	15	6.67 ± 0.05*	81	1.9 ± 0.1*	190 ± 5*
Bac	0.18 ± 0.01*	0.37 ± 0.02*	2.35 ± 0.05*	10.71	36	7.10 ± 0.05*	114	1.8 ± 0.3*	413 ± 12*
Pan	0.33 ± 0.11*	0.43 ± 0.01*	2.6 ± 0.2*	11.12	51	7.02 ± 0.07*	146	2.1 ± 0.3*	430 ± 5*
pDe-GR lakes – October 2018									
Pey	0.14	<LoD	<LoD	6.81	5	5.94	151	1.64	79
Cam	0.18	0.21	0.40	8.47	10	6.36	222	1.08	162
Opa	0.17	0.51	0.48	5.90	17	6.74	127	1.13	307
Arn	0.21	0.81	2.19	7.41	20	6.79	221	-	309
Coa	0.25	0.09	<LoD	6.18	7	6.02	163	1.87	125
Pan	0.68	0.63	2.20	9.87	25	6.64	193	2.03	372
Bac	0.22	0.82	1.89	8.97	32	6.75	207	1.05	358

Table SI 1. (Continued)

Lake	Cl ⁻	NO ₃ ⁻	SO ₄ ²⁻	T	Cond.	pH	E	NPOC	DIC
De-SR lakes – June 2017									
Bad	0.20 ± 0.04*	0.77 ± 0.01*	3.59 ± 0.06*	-	-	7.46	-	2.0 ± 0.2*	787 ± 3*
Ara	0.20 ± 0.03*	0.64 ± 0.03*	2.90 ± 0.03*	9.53	46	7.35 ± 0.02*	271	1.6 ± 0.2*	680 ± 3*
Azu	0.20 ± 0.02*	0.9 ± 0.2*	6.51 ± 0.04*	8.22	58	7.47	131	2.1 ± 0.2*	797
Pec	0.18 ± 0.01*	0.79 ± 0.06*	2.61 ± 0.04*	8.84	36	7.13 ± 0.02*	198	1.51 ± 0.02*	457 ± 2*
Par	0.49 ± 0.03*	0.5 ± 0.1*	1.76 ± 0.05*	10.61	41	7.19 ± 0.03*	118	2.8 ± 0.4*	663 ± 2*
Ord	0.18 ± 0.01*	0.12 ± 0.03*	1.86 ± 0.09*	18.29	61	7.34 ± 0.03*	157	2.7 ± 0.4*	695 ± 32*
De-SR lakes – June 2018									
Bad	0.19	1.13	5.43	5.89	34	7.21	207	0.89	915
Ara	0.17	0.58	2.75	5.70	38	6.99	84	0.85	597
Azu	0.18	0.76	4.91	4.87	46	7.17	149	0.70	675
Pec	0.17	1.03	1.68	4.33	21	6.64	149	0.65	314
Par	0.50	0.46	1.71	-	-	-	-	2.18	705
Ord	0.20	0.20	1.86	17.50	57	7.02	97	2.26	722
De-SR lakes – June 2019									
Azu	0.11	1.16	5.02	-	-	-	-	0.94	-
Pec	0.10	0.69	0.77	-	-	-	-	1.30	-
Ord	0.19	0.38	1.59	-	-	-	-	1.66	-
De-SR lakes – October 2017									
Bad	0.20 ± 0.05*	0.41 ± 0.08*	3.7 ± 0.1*	9.72	67	7.59 ± 0.04*	46	3.0 ± 0.6*	858 ± 14*
Ara	0.15 ± 0.01*	0.21 ± 0.02*	3.74 ± 0.04*	9.85	65	7.91 ± 0.03*	80	2.8 ± 0.3*	822 ± 3*
Azu	0.17 ± 0.01*	0.68 ± 0.03*	7.56 ± 0.05*	8.77	72	7.60 ± 0.05*	102	2.5 ± 0.1*	882 ± 6*

Table SI 1. (Continued)

Lake	Cl ⁻	NO ₃ ⁻	SO ₄ ²⁻	T	Cond.	pH	E	NPOC	DIC
De-SR lakes – October 2018									
Bad	0.15	0.29	2.96	5.33	45	7.37	136	1.26	778
Ara	0.15	0.17	3.23	8.32	50	7.23	170	1.79	777
Azu	0.16	0.76	6.78	5.39	50	7.27	110	0.94	803
Pec	0.19	0.69	2.39	8.13	35	6.79	216	1.08	444
Par	0.68	0.57	1.66	9.53	34	6.99	240	2.30	642
Ord	0.39	0.14	1.50	9.32	48	6.90	225	4.63	573
PT-SR lakes – June 2018									
Gen	0.36	0.18	0.42	6.87	20	6.72	202	0.86	346
Ber	0.23	0.37	0.32	-	-	-	-	0.77	168
Rou	0.30	0.14	0.50	-	-	-	-	0.87	345
PT-SR lakes – June 2019									
Gen	0.32	0.44	0.59	7.57	30	-	285	1.15	-
Ber	0.24	0.22	<LoD	2.14	9	-	206	1.12	-
Rou	0.26	0.30	0.58	12.69	34	-	234	1.45	-
PT-SR lakes – October 2018									
Gen	0.26	<LoD	0.31	12.80	24	6.75	96	1.38	358
Ber	0.31	<LoD	<LoD	12.09	15	7.43	38	1.58	237
Rou	0.29	<LoD	0.47	13.38	25	6.85	36	1.86	359
Cr-SR lake (Sabocos) – June 2018, 2019 and October 2018									
Sab J18	0.21	0.49	2.33	17.25	115	7.53	207	1.71	1723
Sab J19	0.15	0.76	2.55	11.87	130	-	138	1.80	-
Sab O18	0.20	0.37	2.54	10.70	101	7.64	245	1.96	1694

Table SI 2. Subsurface concentrations of total bulk (TSe Bulk), filtered (TDSe) and volatile (TVSe) selenium, and of its species: selenite (Se(IV)), selenate (Se(VI)), and calculated Red.Se (TDSe – Se(IV) – Se(VI)); dimethylselenide (DMSe), dimethyl selenide sulphide (DMSeS) and dimethyl diselenide (DMDSe). Values are grouped by lake (*italic*) and prevailing bedrock is indicated in parenthesis. The average value for each lake is indicated in bold. n.s. means no sample and values under the limit of quantification are indicated as <LoQ. Values are presented either in ng Se L⁻¹ (for non-volatile Se species) or pg Se L⁻¹ (for volatile Se data) ± the associated error. The associated error corresponds to the analytical uncertainty except for June 2017 where the error combines uncertainties of the analysis and sampling (three replicated (n=3) points sampled/lake).

Year	Month	TSe Bulk	TDSe	Se(IV) ng Se L⁻¹	Se(VI)	Red.Se	TVSe	DMSe pg Se L⁻¹	DMSeS	DMDSe
Peyregnets de Cambalès (pDe-GR lake)										
2017	June	12 ± 1	13 ± 1	<LoQ	7.5 ± 0.6	<LoQ	81 ± 12	77 ± 12	<LoQ	<LoQ
2017	October	20 ± 2	14 ± 1	<LoQ	<LoQ	<LoQ	34 ± 5	31 ± 5	<LoQ	<LoQ
2018	June	9 ± 1	7 ± 1	<LoQ	<LoQ	<LoQ	19 ± 3	15 ± 2	<LoQ	<LoQ
2018	October	19 ± 1	11 ± 1	<LoQ	<LoQ	<LoQ	12 ± 2	11 ± 2	<LoQ	<LoQ
Grand Lac de Cambalès (pDe-GR lake)										
2017	June	15 ± 2	15 ± 2	<LoQ	8 ± 1	5 ± 3	45 ± 7	41 ± 6	<LoQ	<LoQ
2017	October	23 ± 2	18 ± 1	<LoQ	11 ± 1	<LoQ	52 ± 8	49 ± 7	<LoQ	<LoQ
2018	June	15 ± 1	13 ± 1	<LoQ	10.3 ± 0.6	<LoQ	41 ± 6	37 ± 6	<LoQ	<LoQ
2018	October	19 ± 1	17 ± 2	<LoQ	6.3 ± 0.7	<LoQ	4 ± 1	2.6 ± 0.4	<LoQ	<LoQ
Petite Opale (pDe-GR lake)										
2017	June	19 ± 2	21 ± 2	<LoQ	13 ± 1	6 ± 3	90 ± 13	86 ± 13	<LoQ	<LoQ
2017	October	25 ± 2	19 ± 3	<LoQ	13 ± 1	<LoQ	77 ± 10	63 ± 9	4.3 ± 0.6	10 ± 1
2018	June	20 ± 1	18 ± 2	<LoQ	17 ± 1	<LoQ	22 ± 4	18 ± 3	<LoQ	<LoQ
2018	October	21 ± 2	13 ± 1	<LoQ	5.6 ± 0.7	<LoQ	12 ± 2	11 ± 2	1.3 ± 0.2	<LoQ
Pourtet (pDe-GR lake)										
2017	June	14 ± 1	13 ± 2	<LoQ	4.3 ± 0.4	7 ± 2	22 ± 4	18 ± 3	<LoQ	<LoQ

Table SI 2. (Continued)

Year	Month	TSe Bulk	TDS _{Se}	Se(IV) ng Se L ⁻¹	Se(VI)	Red.Se	TVSe	DMSe pg Se L ⁻¹	DMSeS	DMDSe
Nère (pDe-GR lake)										
2017	June	13 ± 1	13 ± 1	<LoQ	4.3 ± 0.4	7 ± 3	65 ± 10	61 ± 9	<LoQ	<LoQ
Arnales (pDe-GR lake)										
2017	June	25 ± 2	23 ± 2	<LoQ	19 ± 2	<LoQ	77 ± 11	73 ± 11	<LoQ	<LoQ
2017	October	37 ± 3	26 ± 2	<LoQ	21 ± 2	<LoQ	46 ± 7	43 ± 7	<LoQ	<LoQ
2018	June	19 ± 1	17 ± 2	<LoQ	13 ± 1	<LoQ	32 ± 5	29 ± 4	<LoQ	<LoQ
2018	October	27 ± 2	22 ± 2	<LoQ	18 ± 1	<LoQ	56 ± 8	54 ± 8	<LoQ	<LoQ
2019	June	25 ± 3	n.s.	<LoQ	22 ± 2	–	n.s.	n.s.	n.s.	n.s.
Coanga (pDe-GR lake)										
2017	June	19 ± 2	18 ± 2	<LoQ	4.9 ± 0.4	11 ± 3	113 ± 17	110 ± 16	<LoQ	<LoQ
2017	October	26 ± 2	19 ± 2	<LoQ	<LoQ	<LoQ	55 ± 7	47 ± 7	4.9 ± 0.7	3.1 ± 0.5
2018	June	12 ± 1	11 ± 1	<LoQ	4.3 ± 0.4	5 ± 2	71 ± 10	67 ± 10	<LoQ	<LoQ
2018	October	25 ± 2	20 ± 2	<LoQ	5.6 ± 0.5	12 ± 3	29 ± 4	27 ± 4	<LoQ	<LoQ
2019	June	15 ± 1	n.s.	<LoQ	6 ± 1	–	n.s.	n.s.	n.s.	n.s.
Xuans (pDe-GR lake)										
2017	June	20 ± 2	22 ± 2	3.2 ± 0.5	9 ± 1	10 ± 2	94 ± 14	89 ± 13	<LoQ	<LoQ
Baños de Panticosa (pDe-GR lake)										
2017	June	31 ± 2	29 ± 3	<LoQ	18 ± 1	9 ± 4	118 ± 13	49 ± 7	67 ± 10	<LoQ
2017	October	30 ± 4	22 ± 1	<LoQ	11 ± 1	<LoQ	67 ± 7	29 ± 4	35 ± 5	4.1 ± 0.6
2018	June	23 ± 2	22 ± 1	<LoQ	13.4 ± 0.4	6 ± 2	n.s.	n.s.	n.s.	n.s.
2018	October	29 ± 2	23 ± 2	<LoQ	10.2 ± 0.4	11 ± 3	n.s.	n.s.	n.s.	n.s.
2019	June	20 ± 1	n.s.	<LoQ	23 ± 2	–	n.s.	n.s.	n.s.	n.s.

Table SI 2. (Continued)

Year	Month	TSe Bulk	TDSe	Se(IV) ng Se L ⁻¹	Se(VI)	Red.Se	TVSe	DMSe pg Se L ⁻¹	DMSeS	DMDSe
Bachimaña Bajo (pDe-GR lake)										
2017	June	34 ± 2	35 ± 2	<LoQ	21 ± 2	13 ± 4	n.s.	n.s.	n.s.	n.s.
2017	October	33 ± 2	26 ± 2	<LoQ	10 ± 1	10 ± 6	69 ± 7	41 ± 6	25 ± 4	3.0 ± 0.4
2018	June	24 ± 2	25 ± 2	<LoQ	19 ± 1	<LoQ	95 ± 14	92 ± 14	<LoQ	<LoQ
2018	October	28 ± 2	22 ± 3	<LoQ	10.5 ± 0.5	10 ± 3	22 ± 2	12 ± 2	10 ± 2	1.1 ± 0.2
2019	June	27 ± 3	n.s.	<LoQ	23 ± 2	–	n.s.	n.s.	n.s.	n.s.
Badète (De-SR lake)										
2017	June	68 ± 4	74 ± 3	<LoQ	74 ± 6	<LoQ	12 ± 3	7 ± 1	<LoQ	<LoQ
2017	October	65 ± 4	53 ± 2	<LoQ	44 ± 4	<LoQ	74 ± 9	59 ± 9	12 ± 2	2.5 ± 0.4
2018	June	82 ± 5	66 ± 3	<LoQ	72 ± 4	<LoQ	18 ± 3	14 ± 2	<LoQ	<LoQ
2018	October	50 ± 3	43 ± 4	<LoQ	37 ± 4	<LoQ	9 ± 1	6.0 ± 0.9	2.3 ± 0.4	<LoQ
Arratille (De-SR lake)										
2017	June	63 ± 4	69 ± 4	<LoQ	62 ± 5	<LoQ	85 ± 11	14 ± 2	68 ± 10	<LoQ
2017	October	57 ± 4	48 ± 2	<LoQ	38 ± 3	<LoQ	119 ± 14	17 ± 3	91 ± 14	10 ± 1
2018	June	56 ± 3	46 ± 3	<LoQ	51 ± 2	<LoQ	27 ± 3	12 ± 2	13 ± 2	<LoQ
2018	October	49 ± 3	39 ± 4	<LoQ	34 ± 4	<LoQ	4 ± 1	3.0 ± 0.5	<LoQ	<LoQ
Azul Superior (De-SR lake)										
2017	June	73 ± 5	78 ± 6	<LoQ	72 ± 6	<LoQ	70 ± 8	44 ± 7	24 ± 4	<LoQ
2017	October	66 ± 4	55 ± 3	<LoQ	65 ± 5	<LoQ	39 ± 5	31 ± 5	6 ± 1	1.9 ± 0.3
2018	June	51 ± 2	49 ± 3	<LoQ	55 ± 2	<LoQ	26 ± 4	22 ± 3	<LoQ	<LoQ
2018	October	55 ± 3	48 ± 5	<LoQ	56 ± 2	<LoQ	7 ± 1	5 ± 1	<LoQ	<LoQ
2019	June	69 ± 5	n.s.	3.5 ± 1	71 ± 5	–	n.s.	n.s.	n.s.	n.s.

Table SI 2. (Continued)

Year	Month	TSe Bulk	TDSe	Se(IV) ng Se L ⁻¹	Se(VI)	Red.Se	TVSe	DMSe	DMSeS	DMDSe
							pg Se L ⁻¹			
Pecico (De-SR lake)										
2017	June	46 ± 3	47 ± 3	<LoQ	41 ± 3	<LoQ	36 ± 6	32 ± 5	<LoQ	<LoQ
2018	June	27 ± 2	26 ± 1	<LoQ	24 ± 1	<LoQ	41 ± 6	37 ± 6	<LoQ	<LoQ
2018	October	37 ± 2	31 ± 3	<LoQ	33 ± 1	<LoQ	n.s.	n.s.	n.s.	n.s.
2019	June	26 ± 2	n.s.	<LoQ	23 ± 2	–	n.s.	n.s.	n.s.	n.s.
Paradis (De-GR lake)										
2017	June	37 ± 2	38 ± 3	<LoQ	22 ± 2	14 ± 4	n.s.	n.s.	n.s.	n.s.
2018	June	37 ± 2	28 ± 1	<LoQ	27 ± 2	<LoQ	n.s.	n.s.	n.s.	n.s.
2018	October	35 ± 2	31 ± 3	<LoQ	19 ± 2	<LoQ	n.s.	n.s.	n.s.	n.s.
Ordicuso (De-GR lake)										
2017	June	31 ± 2	25 ± 2	<LoQ	5.8 ± 0.6	17 ± 3	484 ± 53	99 ± 15	339 ± 51	46 ± 7
2018	June	24 ± 2	20 ± 2	<LoQ	7.0 ± 0.3	11 ± 3	n.s.	n.s.	n.s.	n.s.
2018	October	24 ± 2	20 ± 2	<LoQ	7.9 ± 0.3	10 ± 3	11 ± 1	9 ± 1	2.0 ± 0.3	<LoQ
2019	June	25 ± 2	n.s.	<LoQ	17 ± 3	–	n.s.	n.s.	n.s.	n.s.
Gentau (PT-SR lake)										
2018	June	15 ± 1	13 ± 1	<LoQ	11 ± 1	<LoQ	17 ± 3	9 ± 1	6 ± 1	<LoQ
2018	October	15 ± 1	11 ± 1	<LoQ	<LoQ	<LoQ	5.3 ± 0.8	3.0 ± 0.5	1.7 ± 0.3	<LoQ
2019	June	24 ± 2	11 ± 1	<LoQ	9 ± 1	<LoQ	3 ± 1	1.5 ± 0.2	<LoQ	<LoQ
Bersau (PT-SR lake)										
2018	June	15 ± 1	12 ± 1	<LoQ	7.1 ± 0.3	<LoQ	12 ± 1	9 ± 1	1.2 ± 0.2	<LoQ
2018	October	17 ± 2	12 ± 2	<LoQ	<LoQ	<LoQ	8 ± 1	3.7 ± 0.6	2.8 ± 0.4	<LoQ
2019	June	16 ± 1	10 ± 1	<LoQ	9 ± 1	<LoQ	3 ± 1	2.8 ± 0.4	<LoQ	<LoQ

Table SI 2. (Continued)

Year	Month	TSe Bulk	TDSe	Se(IV) ng Se L ⁻¹	Se(VI)	Red.Se	TVSe	DMSe	DMSes	DMDSe
							pg Se L ⁻¹			
Roumassot (PT-SR lake)										
2018	June	25 ± 2	13 ± 1	<LoQ	7.5 ± 0.3	<LoQ	36 ± 5	29 ± 4	4.7 ± 0.7	<LoQ
2018	October	18 ± 2	13 ± 1	<LoQ	<LoQ	<LoQ	6 ± 1	3.6 ± 0.5	1.8 ± 0.3	<LoQ
2019	June	15 ± 2	12 ± 1	<LoQ	6 ± 1	<LoQ	4 ± 1	3.0 ± 0.5	<LoQ	<LoQ
Sabocos (Cr-SR lake)										
2018	June	17 ± 1	15 ± 1	<LoQ	10.7 ± 0.4	<LoQ	28 ± 4	17 ± 3	12 ± 2	<LoQ
2018	October	20 ± 2	15 ± 2	<LoQ	5.8 ± 0.5	8 ± 2	9 ± 1	1.5 ± 0.2	5 ± 1	<LoQ
2019	June	20 ± 1	12 ± 1	<LoQ	10 ± 1	<LoQ	<LoQ	<LoQ	<LoQ	<LoQ

Table SI 3. Spearman correlation matrix of major components in lake waters and total dissolved selenium. Moderate ($|\rho|>0.4$), strong ($|\rho|>0.6$) and very strong ($|\rho|>0.8$) correlations are indicated with gradual orange tones. Sabocos lake presenting outlier DIC values was excluded.

	TDSe	NO ₃ ⁻	SO ₄ ²⁻	Cl ⁻	DIC	NPOC
TDSe	1					
NO ₃ ⁻	0.60	1				
SO ₄ ²⁻	0.93	0.58	1			
Cl ⁻	0.03	0.003	0.02	1		
DIC	0.84	0.35	0.88	0.22	1	
NPOC	0.37	-0.23	0.35	0.36	0.52	1

Table SI 4. Depth profiles raw data for Arratille, Azul Superior, Gentau and Sabocos lakes. Data include non-gaseous dissolved total Se bulk (TSe Bulk), TSe filtered (TDSe) and Se speciation: selenate (Se(VI)) and the calculated Red.Se fraction; and total volatile Se and gaseous Se speciation: dimethylselenide (DMSe), dimethyl selenide sulphide (DMSeS) and dimethyl diselenide (DMDSe).

n.s. means no sample and values under the limit of quantification are indicated as <LoQ. Values are presented either in ng Se L⁻¹ (for non-volatile Se species) or pg Se L⁻¹ (for volatile Se data) ± the associated error. The associated error corresponds to the analytical uncertainty. Parameters shown are temperature (Temp.), conductivity (Cond.), oxygen saturation (Ox. Sat.), dissolved oxygen (DO), nitrate (NO₃⁻) and sulfate (SO₄²⁻). Se(IV) was in all cases <LoQ.

Depth m	TSe Bulk	TDSe	Se(VI)	Red.Se	TVSe	DMSe	DMSeS	DMDSe	T ° C	Cond. µS cm ⁻¹	Ox. Sat. %	DO mg L ⁻¹	NO ₃ ⁻ mg L ⁻¹	SO ₄ ²⁻ mg L ⁻¹
Arratille (De-SR lake) – June 2018														
0.5	54 ± 2	46 ± 2	53 ± 2	<LoQ	40 ± 5	27 ± 4	11 ± 2	<LoQ	5.70	38	72	9.0	0.58	2.75
3	56 ± 4	45 ± 3	54 ± 2	<LoQ	30 ± 4	12 ± 2	17 ± 2	<LoQ	4.91	37	76	9.8	0.78	2.82
6	56 ± 3	43 ± 2	47 ± 2	<LoQ	72 ± 10	63 ± 9	7 ± 1	<LoQ	4.87	37	76	9.8	0.74	2.75
12	55 ± 2	46 ± 2	53 ± 2	<LoQ	85 ± 10	66 ± 10	14 ± 2	6 ± 1	4.79	138	5	0.7	0.71	2.83
Azul Superior (De-SR lake) – June 2018														
0.5	51 ± 2	49 ± 3	55 ± 2	<LoQ	26 ± 4	22 ± 3	<LoQ	<LoQ	4.87	46	74	9.5	0.76	4.91
3.5	55 ± 2	53 ± 2	59 ± 2	<LoQ	36 ± 5	32 ± 5	<LoQ	<LoQ	4.82	48	77	9.8	1.17	5.23
7	55 ± 4	56 ± 3	64 ± 2	<LoQ	18 ± 3	14 ± 2	<LoQ	<LoQ	4.67	48	76	9.8	0.64	5.26
Azul Superior (De-SR lake) – October 2018														
0.5	55 ± 3	48 ± 5	56 ± 2	<LoQ	7 ± 1	5 ± 1	<LoQ	<LoQ	5.39	50	68	8.6	0.76	6.78
3	54 ± 3	47 ± 5	56 ± 2	<LoQ	6 ± 1	3 ± 1	1.4 ± 0.2	<LoQ	5.08	50	71	9.1	0.78	6.70
6	54 ± 3	48 ± 5	58 ± 2	<LoQ	6 ± 1	3 ± 1	1.8 ± 0.3	<LoQ	4.98	50	72	9.2	0.90	6.73

Table SI. 4 (Continued)

Depth	TSe Bulk	TDSe	Se(VI)	Red.Se	TVSe	DMSe	DMSes	DMDSe	T	Cond.	Ox. Sat.	DO	NO ₃ ⁻	SO ₄ ²⁻
Gentau (PT-SR lake) – June 2018														
0.5	15 ± 1	13 ± 1	10.8 ± 0.4	<LoQ	17 ± 3	9 ± 1	6 ± 1	<LoQ	6.87	20	82	10.0	0.18	0.42
5	14 ± 1	13 ± 1	9.5 ± 0.4	<LoQ	33 ± 3	19 ± 3	9 ± 1	4.5 ± 0.7	4.97	20	78	10.0	0.18	0.43
8	13 ± 1	14 ± 1	8.5 ± 0.3	<LoQ	11 ± 3	7 ± 1	<LoQ	<LoQ	4.37	38	27	3.5	0.04	0.49
12	13 ± 2	13 ± 1	4.2 ± 0.1	7 ± 2	31 ± 3	6 ± 1	12 ± 2	13 ± 2	4.23	47	3	0.3	0.03	0.68
18	10 ± 1	22 ± 2	3.5 ± 0.1	17 ± 2	96 ± 11	9 ± 1	14 ± 2	73 ± 11	4.40	67	1	0.2	0.08	0.66
Gentau (PT-SR lake) – October 2018														
0.5	15 ± 1	11 ± 1	<LoQ	<LoQ	5 ± 1	2.9 ± 0.4	1.7 ± 0.3	<LoQ	12.80	24	79	8.4	<LoD	0.31
5	16 ± 1	11 ± 1	<LoQ	<LoQ	10 ± 1	7 ± 1	2.2 ± 0.3	<LoQ	12.68	24	78	8.2	<LoD	0.30
8	15 ± 2	11 ± 1	<LoQ	<LoQ	6 ± 1	3.5 ± 0.5	1.7 ± 0.3	<LoQ	12.62	24	77	8.1	<LoD	0.33
12	16 ± 1	13 ± 1	<LoQ	<LoQ	2 ± 1	<LoQ	<LoQ	<LoQ	8.52	44	33	3.8	<LoD	0.47
17	21 ± 2	14 ± 2	<LoQ	<LoQ	9 ± 1	3.3 ± 0.5	5 ± 1	<LoQ	6.05	56	2	0.2	0.14	<LoD
Gentau (PT-SR lake) – June 2019														
0.5	24 ± 2	11 ± 1	9 ± 1	<LoQ	<LoQ	<LoQ	<LoQ	<LoQ	7.57	30	79	9.5	0.44	0.59
5	16 ± 2	11 ± 1	9 ± 1	<LoQ	6 ± 1	<LoQ	3.0 ± 0.4	1.9 ± 0.3	6.76	31	84	10.3	0.27	0.54
8	15 ± 1	11 ± 1	7 ± 1	<LoQ	5 ± 1	<LoQ	3.6 ± 0.5	<LoQ	5.82	35	91	11.4	0.18	0.58
12	15 ± 1	11 ± 1	<LoQ	8 ± 2	n.s.	n.s.	n.s.	n.s.	4.51	42	20	2.6	0.41	0.71
17.5	16 ± 1	10 ± 1	<LoQ	7 ± 2	8 ± 1	3.7 ± 0.6	<LoQ	3.3 ± 0.5	4.30	47	4	0.5	0.13	0.70

Table SI. 4 (Continued)

Depth	TSe Bulk	TDSe	Se(VI)	Red.Se	TVSe	DMSe	DMSes	DMDSe	T	Cond.	Ox. Sat.	DO	NO ₃ ⁻	SO ₄ ²⁻
Sabocos (Cr-SR lake) – June 2018														
0.5	17 ± 1	15 ± 1	10.7 ± 0.4	<LoQ	28 ± 4	17 ± 3	9 ± 1	<LoQ	17.25	115	102	9.8	0.49	2.33
5	19 ± 2	16 ± 2	12.4 ± 0.4	<LoQ	26 ± 3	12 ± 2	12 ± 2	<LoQ	10.70	115	82	9.1	0.48	2.51
9	19 ± 2	19 ± 1	11.9 ± 0.4	5 ± 2	8 ± 3	<LoQ	4 ± 1	<LoQ	7.57	119	61	7.3	0.45	3.21
15	19 ± 2	19 ± 1	11.7 ± 0.4	6 ± 2	10 ± 3	7 ± 1	<LoQ	<LoQ	6.01	120	49	6.1	0.54	3.07
20	18 ± 1	19 ± 1	13.2 ± 0.4	<LoQ	19 ± 3	15 ± 2	<LoQ	<LoQ	5.13	124	8	1.1	0.50	3.02
25	17 ± 1	17 ± 2	10.1 ± 0.3	5 ± 3	80 ± 8	36 ± 5	10 ± 1	34 ± 5	4.80	127	1	0.2	0.46	2.74
Sabocos (Cr-SR lake) – October 2018														
0.5	19 ± 2	15 ± 2	6 ± 1	6 ± 2	9 ± 1	1.5 ± 0.2	6 ± 0.9	1.6 ± 0.2	10.70	101	78	8.7	0.37	2.54
4	19 ± 1	15 ± 2	6 ± 1	8 ± 3	10 ± 1	1.7 ± 0.3	7 ± 1	1.7 ± 0.3	10.56	102	78	8.7	0.15	2.54
8	22 ± 2	15 ± 2	6 ± 1	7 ± 2	8 ± 1	1.5 ± 0.2	5 ± 1	1.7 ± 0.3	10.53	102	77	8.6	0.17	2.55
12	18 ± 1	14 ± 2	9.5 ± 0.4	<LoQ	11 ± 1	7 ± 1	2.5 ± 0.4	2.1 ± 0.3	7.87	119	62	7.4	0.34	3.09
18	19 ± 2	14 ± 2	15 ± 1	<LoQ	4 ± 1	2.8 ± 0.4	<LoQ	<LoQ	5.97	117	35	4.4	0.44	3.10
23	19 ± 2	16 ± 2	11.5 ± 0.4	<LoQ	9 ± 1	8 ± 1	<LoQ	<LoQ	5.70	121	5	0.6	0.20	3.03
Sabocos (Cr-SR lake) – June 2019														
0.5	20 ± 1	12 ± 1	10 ± 1	<LoQ	<LoQ	<LoQ	<LoQ	<LoQ	11.87	130	79	8.6	0.76	2.55
5	20 ± 2	12 ± 1	9 ± 1	<LoQ	6 ± 1	<LoQ	4 ± 0.6	<LoQ	10.18	133	82	9.3	0.61	2.40
9	22 ± 2	15 ± 1	13 ± 2	<LoQ	6 ± 1	<LoQ	4.6 ± 0.7	<LoQ	7.07	130	76	9.2	0.71	2.79
13	21 ± 2	16 ± 1	15 ± 1	<LoQ	<LoQ	<LoQ	<LoQ	<LoQ	6.09	129	70	8.7	0.78	2.80
18	22 ± 2	17 ± 1	17 ± 2	<LoQ	<LoQ	<LoQ	<LoQ	<LoQ	5.26	134	48	6.1	0.79	3.02
25	24 ± 2	17 ± 2	17 ± 1	<LoQ	6 ± 1	3.9 ± 0.6	<LoQ	<LoQ	4.87	140	7	0.9	0.57	2.73

Table SI 5. Estimated fluxes for each investigated lake of Se wet deposition (with data from Roulier *et al.*⁵¹) and Se gaseous emission (using the model of Cole and Caraco⁵³ adapted by Lancelleur *et al.*⁵) for windspeeds of 3 m · s⁻¹). Selenium input and emission is presented as g Se · year⁻¹ and as % of selenium stock in the corresponding lake. Calculation considering an ice-covering period of 6 months.

Lake	Surface m ²	Volume m ³	Stock Se g	Input Se		Se emitted (wind 3 m s ⁻¹)	
				g yr ⁻¹	%	g yr ⁻¹	%
PEY	11690	52987	0.59	0.59	100	0.034	5,8
CAM	34640	208745	3.23	1.74	54	0.090	2,8
OPA	6380	25788	0.46	0.32	70	0.020	4.3
POU	59470	387705	5.06	2.99	59	-	-
NER	29100	178522	2.32	1.46	63	-	-
PAR	4300	16570	0.54	0.22	40	-	-
BAD	69660	243990	14.46	3.50	24	0.304	2.1
ARA	58650	264307	13.30	2.95	22	0.437	3,3
BAC	30770	193335	5.21	1.55	30	0.174	3.3
PAN	55000	470072	11.26	2.76	25	0.455	4.0
AZU	38870	273527	15.80	1.95	12	0.141	0.9
PEC	9090	38954	1.35	0.46	34	-	-
ARN	25980	84579	1.87	1.31	70	0.120	6.4
ORD	3720	14171	0.30	0.19	62	-	-
COA	5810	23208	0.39	0.29	74	0.021	5.3
XUA	29740	183875	4.03	1.49	37	-	-
SAB	95610	1183798	16.79	4.80	29	0.132	0,8
GEN	86240	993736	11.82	4.33	37	0.069	0,6
BER	128230	2266475	26.12	6.44	25	0.062	0,2
ROU	51450	424200	5.39	2.58	48	0.023	0,4
min	3720	14171	0.30	0.19	12%	0.020	0,2%
max	128230	2266475	26.12	6.44	100%	0.455	6,4%
median	32705	201040	4.55	1.64	39%	0.105	3.0%

