

1 Supporting information: Quantification of
2 individual Rare Earth Elements from industrial
3 sources in sewage sludge.

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16 Content: 3 tables, 8 figures

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19 **Tables**20 **Table S1:** Properties and origin (location) of the soil samples.

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Soil identifier	Soil characteristics	Soil texture (0–20 cm)	Bedrock	Location	Landuse	Emission sources
NABO - 53 GEM 1-3A I/1	partially decarbonated calcareous brown earth, rich in clay	Loamy clay	Limestone/Oolithic lime	Gempen (ct. SO)	Artificial meadow, agriculture	Industrial zone, urban area, metal industry
NABO - 62 BET 1-3A IV/1	partially decarbonated calcareous brown earth, rich in clay	Clayey silty clay	Limestone	Bettlach (ct. SO)	Forest/ woodland dominant	Settlement area
NABO - 103 HAE 1-3A III/1	Brown earth-Pseudogley	Loam	Loose rock / fine-grained alluvial	Härkingen (ct. SO)	Agriculture	Motorway
NABO - 9 BR 1-1A I/1	Para-brown earth (eroded)	Sandy loam	Loose rock, silty, loess	Binningen (ct. BL)	Agriculture	Urban area
NABO - 27 JU 1-1A I/1	Pseudogley (acidic)	Loam	Loose rock, (calcareous)	Jussy (ct. GE)	Forest	Urban area
NABO - 32 LB 1-1A I/1	Brown earth (acidic)	Loamy clay	Limestone	La Brevine (ct. NE)	(Alpine) Pasture	Urban area
NABO - 74 MOE 1-1A I/1	Pseudogley (weakly acidic)	Clayey loam	Loose rock, (calcareous)	Mörschwil (ct. SG)	Meadow	Urban area
NABO - 76 SEN 1-1A II/1	Brown earth	Sandy loam	Loose rock	Sent (ct. GR)	Forest	Suburban area
NABO - 97 LUG 1-2A II/1	Brown earth (gleytic), weakly acidic	Sandy loam	Loose rock, loamy	Lugano (ct. TI)	City park	City
NABO – 99 VISP	Brown earth	Sandy loam	Limestone slate	Visp (ct. VS)	Forest	Suburban area

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24 **Table S2:** REE contents of the sludge samples. All concentration values are given in mg kg⁻¹ (dry mass) and correspond to triplicate or
 25 duplicate digestions. In addition to the REE contents, the daily digested sludge production (kg (dry mass) / day) and the number of
 26 connected people are provided. Nr refers to the numbers on the Swiss map (Figure S2). The WWTP are sorted according to the
 27 connected population equivalents.

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Name	La	Ce	Pr	Nd	Sm	Eu	Gd	Tb	Dy	Ho	Er	Tm	Yb	Lu	SludgeProd	Population	Nr
Vernier	5.24	7.57	0.66	2.78	0.48	0.13	0.47	0.08	0.49	0.07	0.22	0.03	0.22	0.03	24300	440750	63
Zuerich	7.01	10.57	1.06	4.01	0.97	0.16	0.67	0.11	0.50	0.10	0.31	0.04	0.32	0.04	25000	429371	5
Basel	6.37	11.41	0.79	2.94	0.54	0.12	0.51	0.07	0.38	0.07	0.23	0.03	0.21	0.03	35600	268317	25
Lausanne	5.48	11.46	1.33	4.79	1.05	0.30	0.81	0.13	0.57	0.11	0.30	0.04	0.29	0.04	28500	235359	52
Bern	4.60	6.18	0.56	2.46	0.49	0.10	0.44	0.06	0.31	0.06	0.19	0.03	0.25	0.03	17300	218965	6
Emmen	3.20	4.74	0.54	2.03	0.44	0.07	0.34	0.05	0.26	0.05	0.15	0.02	0.15	0.02	9270	178555	14
Cham	6.35	9.17	0.84	3.32	0.59	0.12	0.49	0.08	0.43	0.08	0.26	0.03	0.31	0.03	7400	143102	19
Winterthur	6.86	7.89	0.64	2.50	0.46	0.10	0.44	0.06	0.36	0.06	0.21	0.03	0.23	0.03	5200	130101	3
Bioggio	25.47	71.46	0.86	3.22	0.75	0.13	0.57	0.09	0.48	0.09	0.29	0.04	0.28	0.03	5800	124990	50
Uetendorf	5.23	6.29	0.63	2.46	0.65	0.11	0.44	0.06	0.40	0.07	0.19	0.03	0.24	0.03	8200	123223	13
Ramsen	8.57	13.44	1.35	4.98	0.82	0.16	5.47	0.11	0.61	0.12	0.66	0.04	0.32	0.05	5130	97591	29
Zuchwil	17.55	29.42	1.50	5.59	1.01	0.21	0.97	0.13	0.83	0.16	0.45	0.06	0.45	0.06	5700	90668	24
Bruegg	16.75	29.03	1.45	5.53	1.04	0.23	1.01	0.15	0.79	0.16	0.44	0.06	0.48	0.07	2740	85034	11
Birsfelden	18.12	43.65	1.26	4.76	0.88	0.16	0.79	0.12	0.64	0.12	0.36	0.05	0.38	0.05	5400	82473	26
Dietikon	14.12	14.50	0.93	3.08	0.55	0.15	0.55	0.08	0.45	0.09	0.29	0.03	0.26	0.03	4300	78844	4
Aarau	6.62	11.27	1.01	5.58	1.04	0.209	0.83	0.15	0.66	0.12	0.36	0.05	0.40	0.07	4300	74022	40
Ittigen	7.39	13.40	0.66	2.62	0.53	0.11	0.47	0.07	0.41	0.07	0.23	0.03	0.23	0.03	3800	70195	7
Turgi	9.72	10.94	1.11	4.32	0.94	0.17	0.76	0.11	0.63	0.12	0.36	0.05	0.41	0.05	3300	58524	51
Laupen	6.29	11.43	1.29	4.76	0.95	0.18	0.84	0.13	0.72	0.15	0.45	0.06	0.37	0.06	3300	57779	10
Hofen	26.71	170.06	0.89	3.39	0.63	0.12	1.87	0.09	0.43	0.08	0.23	0.03	0.27	0.03	3300	56379	32
Thal	25.21	95.92	1.53	5.41	0.91	0.19	1.10	0.12	0.67	0.14	0.39	0.05	0.46	0.05	5500	55968	33
Neuhausen	4.45	7.20	0.76	2.96	0.54	0.12	0.54	0.09	0.50	0.10	0.26	0.04	0.29	0.03	2600	55740	28
Chur	7.42	11.29	0.70	2.73	0.50	0.13	0.49	0.07	0.41	0.08	0.30	0.03	0.48	0.03	3700	54610	38
Giubiasco	4.63	5.96	0.62	2.53	0.51	0.10	0.50	0.06	0.36	0.07	0.20	0.02	0.23	0.02	2400	53201	48

Winznau	7.05	12.09	1.21	4.75	0.87	0.19	0.80	0.11	0.67	0.13	0.34	0.05	0.36	0.05	2200	52223	23		
Vevey	10.67	22.74	1.91	2.17	0.39	0.10	0.43	0.06	0.34	0.07	0.19	0.03	0.22	0.03	2300	51937	55		
Locarno	8.85	10.01	1.09	3.79	0.79	0.17	0.78	0.10	0.58	0.10	0.33	0.04	0.31	0.04	3800	51373	49		
Duebendorf	4.47	6.23	0.43	1.47	0.28	0.06	0.65	0.04	0.26	0.05	0.13	0.02	0.17	0.02	2350	46619	2		
Bilten	6.03	7.60	0.77	2.86	0.52	0.14	0.61	0.08	0.49	0.09	0.27	0.04	0.27	0.04	1700	44667	18		
Windisch	23.00	14.70	1.68	6.07	10.33	0.23	1.51	0.15	0.87	0.17	0.44	0.06	0.48	0.05	2150	44634	44		
Fribourg	4.71	8.20	0.85	3.40	0.60	0.13	0.59	0.07	0.39	0.08	0.22	0.03	0.24	0.03	4100	43681	22		
Reinach	7.58	7.02	0.78	2.95	0.62	0.12	0.54	0.08	0.48	0.09	0.28	0.03	0.29	0.04	1200	41000	45		
Neuchatel	3.38	5.57	0.50	2.05	0.37	0.08	0.42	0.06	0.29	0.06	0.17	0.02	0.24	0.02	1600	40567	62		
La Chaux-de-Fonds	5.19	8.77	0.89	3.41	0.67	0.15	0.62	0.09	0.55	0.11	0.34	0.05	0.33	0.04	2000	37455	61		
Opfikon	5.41	5.54	0.58	2.23	0.46	0.09	0.36	0.06	0.26	0.05	0.14	0.02	0.15	0.02	3500	37167	9		
Dagmarsellen	6.01	7.31	0.78	2.96	0.55	0.12	0.49	0.08	0.43	0.09	0.26	0.04	0.32	0.03	2210	35885	15		
Gland	5.53	7.88	0.80	3.09	0.55	0.14	0.74	0.09	0.45	0.09	0.23	0.04	0.24	0.04	1260	34409	53		
Yverdon	112.69	500.74	2.52	3.69	0.58	0.12	1.00	0.11	0.51	0.10	0.29	0.05	0.35	0.03	1900	32842	56		
Vuippens	5.10	8.90	0.99	3.61	0.70	0.19	1.15	0.10	0.57	0.11	0.34	0.05	0.31	0.05	2200	31765	21		
Altdorf	7.26	9.81	1.10	4.05	0.74	0.16	0.72	0.11	0.64	0.12	0.37	0.05	0.44	0.05	1300	30409	16		
Weinfelden	5.43	8.13	0.92	3.36	0.75	0.16	0.66	0.09	0.58	0.12	0.36	0.06	0.38	0.06	2500	30136	47		
Wohlen	7.26	10.26	1.13	4.24	0.83	0.15	0.77	0.11	0.63	0.12	0.35	0.05	0.36	0.05	1740	29973	43		
Rancate	5.35	7.43	0.83	3.21	0.65	0.14	0.60	0.09	0.50	0.10	0.26	0.04	0.27	0.04	1440	29009	51		
Schwyz	7.15	8.33	0.94	3.67	0.68	0.15	0.66	0.09	0.53	0.11	0.31	0.04	0.34	0.04	1500	28243	17		
Brig	3.45	6.16	0.59	2.33	0.56	0.12	0.47	0.07	0.39	0.08	0.20	0.03	0.22	0.02	1800	27702	57		
Unterseen	4.80	8.81	0.92	3.52	0.59	0.11	0.61	0.08	0.43	0.08	0.23	0.03	0.29	0.02	1500	25413	8		
Kaisten	8.22	12.72	1.44	5.66	1.19	0.20	0.98	0.15	0.86	0.17	0.46	0.07	0.51	0.06	1800	24024	46		
Buchs	34.52	143.92	1.60	5.89	0.97	0.20	1.02	0.26	0.85	0.17	0.48	0.06	0.55	0.06	1435	23913	34		
Nyon	8.84	6.26	0.66	2.52	0.43	0.09	0.57	0.07	0.38	0.07	0.20	0.03	0.22	0.02	1400	23331	54		
Zwingen	22.22	19.37	2.27	8.08	1.47	0.34	1.27	0.18	1.06	0.21	0.62	0.10	0.77	0.12	1100	23305	27		
Ecublens	7.06	11.88	1.26	4.45	0.86	0.17	0.77	0.11	0.72	0.14	0.40	0.07	0.36	0.06	860	19542	20		
Porrentruy	11.48	21.03	2.42	9.42	1.77	0.34	1.73	0.24	1.38	0.27	0.76	0.11	0.74	0.11	990	16029	64		
Richterswil	5.74	7.89	0.86	3.01	0.53	0.10	0.54	0.07	0.39	0.07	0.20	0.03	0.23	0.03	515	14127	1		
Leuk	7.57	14.55	1.62	6.24	1.27	0.25	1.13	0.17	1.00	0.20	0.56	0.09	0.53	0.09	685	13413	58		

Visp	18.68	18.38	3.19	13.15	2.53	0.63	2.87	0.40	2.65	0.54	1.56	0.19	1.25	0.17	13500	12985	60
Unterehrendingen	9.68	17.22	2.08	7.70	1.49	0.34	1.58	0.21	1.29	0.24	0.67	0.09	0.65	0.07	770	12785	42
Appenzell	4.47	7.15	0.79	3.01	0.58	0.12	0.57	0.07	0.43	0.08	0.25	0.03	0.29	0.04	660	10175	30
Davos	4.76	7.03	0.83	3.02	0.62	0.13	0.59	0.09	0.52	0.09	0.30	0.04	0.33	0.04	821	9901	37
Evionnaz	6.73	12.35	1.31	4.90	1.06	0.22	0.96	0.13	0.84	0.15	0.45	0.06	0.42	0.05	120	6263	59
Scuol	9.95	9.50	0.96	3.49	0.79	0.17	0.74	0.12	0.71	0.14	0.43	0.06	0.41	0.05	170	3533	36
Poschiavo	7.92	15.14	1.64	6.03	1.18	0.23	1.12	0.18	1.06	0.20	0.55	0.08	0.54	0.08	164	3271	35
Disentis	7.37	13.46	1.61	6.28	1.32	0.27	1.14	0.16	0.92	0.19	0.52	0.06	0.46	0.06	110	1877	39
Innerkirchen	5.73	8.60	0.88	3.23	0.56	0.11	0.71	0.07	0.40	0.09	0.25	0.04	0.24	0.04	50	873	12

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31 **Table S3:** REE contents of the soil samples. All concentration values are given in mg kg^{-1} (dry mass) and correspond to triplicate or
 32 duplicate digestions. The numbers in the names correspond to the number in Figure S1.
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Name	La	Ce	Pr	Nd	Sm	Eu	Gd	Tb	Dy	Ho	Er	Tm	Yb	Lu
103_HAE	32.28	68.02	7.76	30.26	5.66	1.08	4.58	0.68	3.91	0.78	2.18	0.31	2.00	0.30
62_BET	36.96	78.50	8.55	32.85	6.19	1.23	5.22	0.82	4.98	0.99	2.90	0.42	2.76	0.42
53_GEM	51.41	102.90	12.08	47.49	9.62	1.99	8.66	1.31	7.61	1.51	4.25	0.59	3.72	0.55
9_BR	38.51	90.63	9.06	34.94	7.14	1.21	6.17	0.86	5.11	1.00	2.80	0.42	2.65	0.37
27_JU	33.81	71.30	7.76	29.50	5.39	1.02	4.86	0.65	3.88	0.74	2.17	0.31	2.02	0.28
32_LB	49.29	105.24	11.49	44.43	8.53	1.67	7.66	1.07	6.19	1.19	3.31	0.47	3.13	0.43
74_MOE	33.28	72.33	7.65	29.84	5.95	1.14	5.44	0.80	4.72	0.95	2.72	0.38	2.58	0.34
76_SEN	37.50	84.52	9.04	34.99	6.93	1.29	6.05	0.77	4.51	0.83	2.41	0.34	2.27	0.28
97_LUG	38.77	83.84	9.29	35.38	6.78	1.34	5.92	0.80	4.58	0.88	2.36	0.32	2.18	0.30
99_VSP	31.21	68.44	7.83	29.27	5.99	1.17	5.16	0.76	4.42	0.83	2.26	0.31	1.93	0.26

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Figures

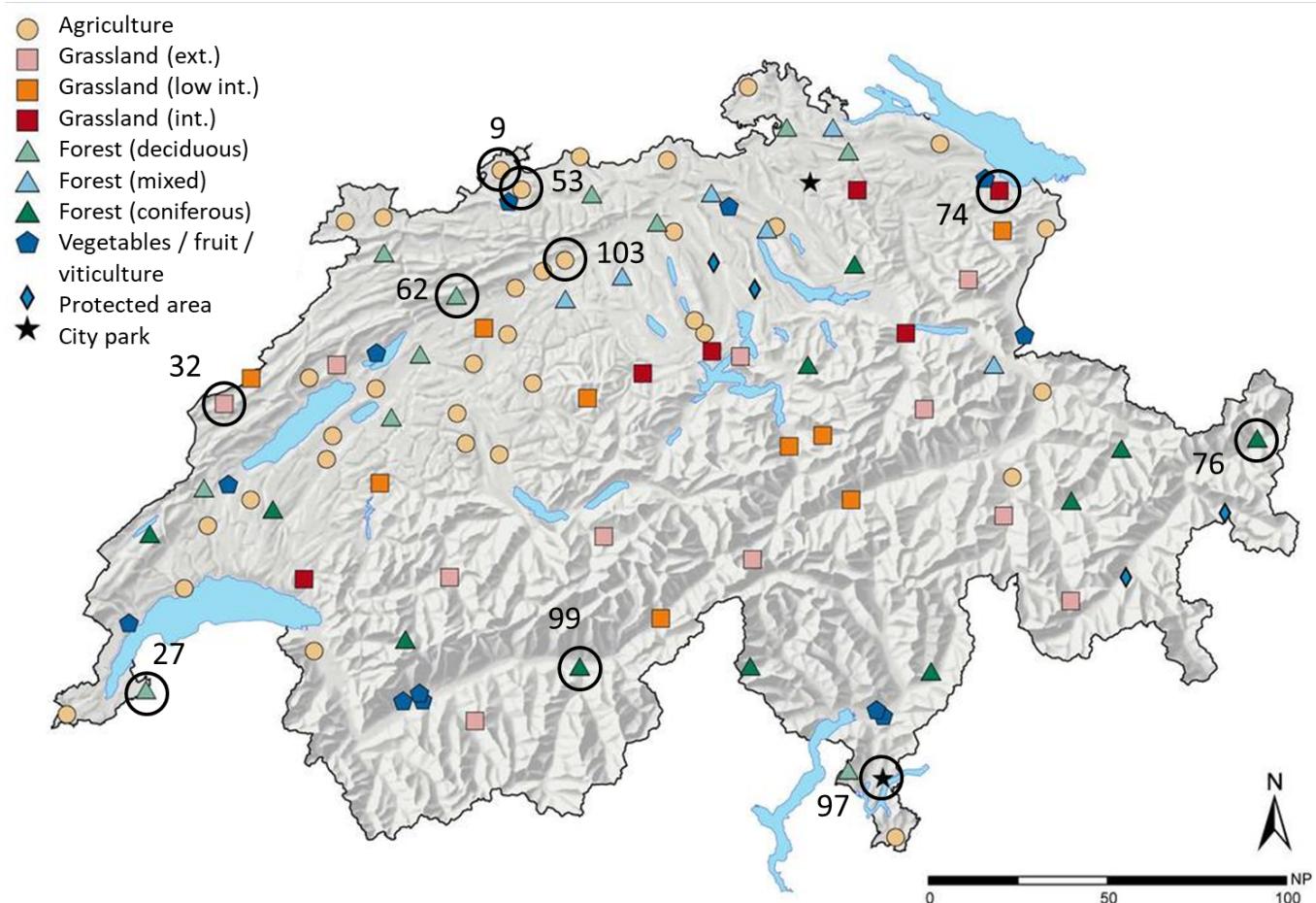


Figure S1: Location of the soil samples (black circles) and associated land use, indicated by the different symbols. The numbers refer to the numbers given in table S2. Other symbols refer to additional sites from the NABO.

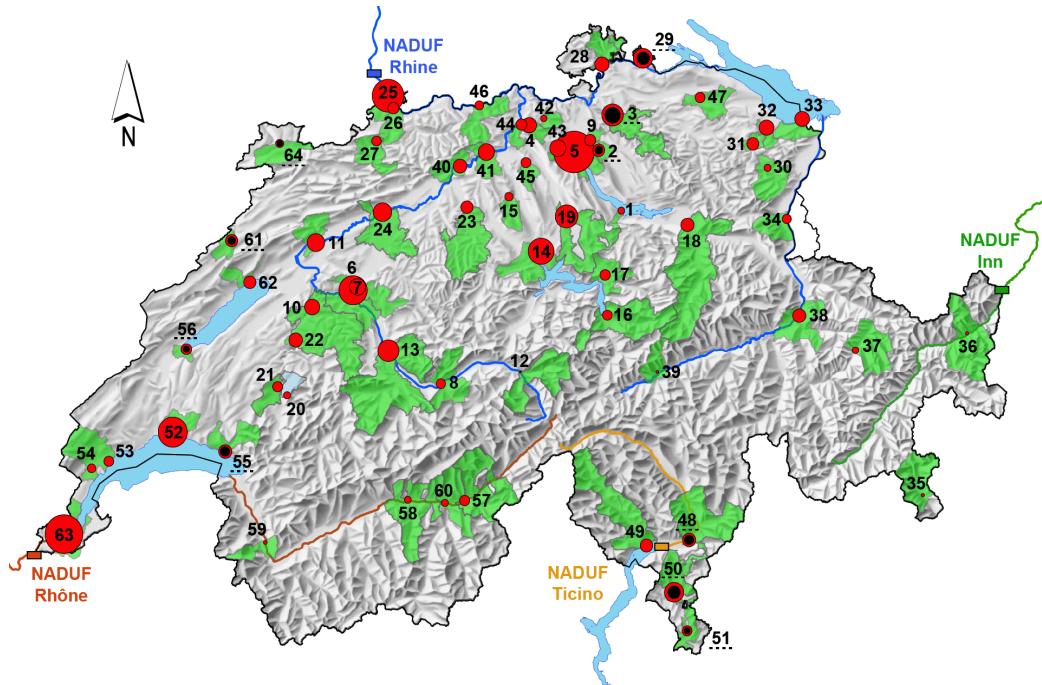


Figure S2: Map of Switzerland with locations of the selected WWTPs (red dots) and the corresponding catchment areas (green areas). The diameter of the red dots reflects the capacities of the corresponding WWTPs (number of connected population equivalents). Modified after (Vriens et al., 2017). WWTP number 31 (St. Gallen) was not included in the current study due to shortage of available materials

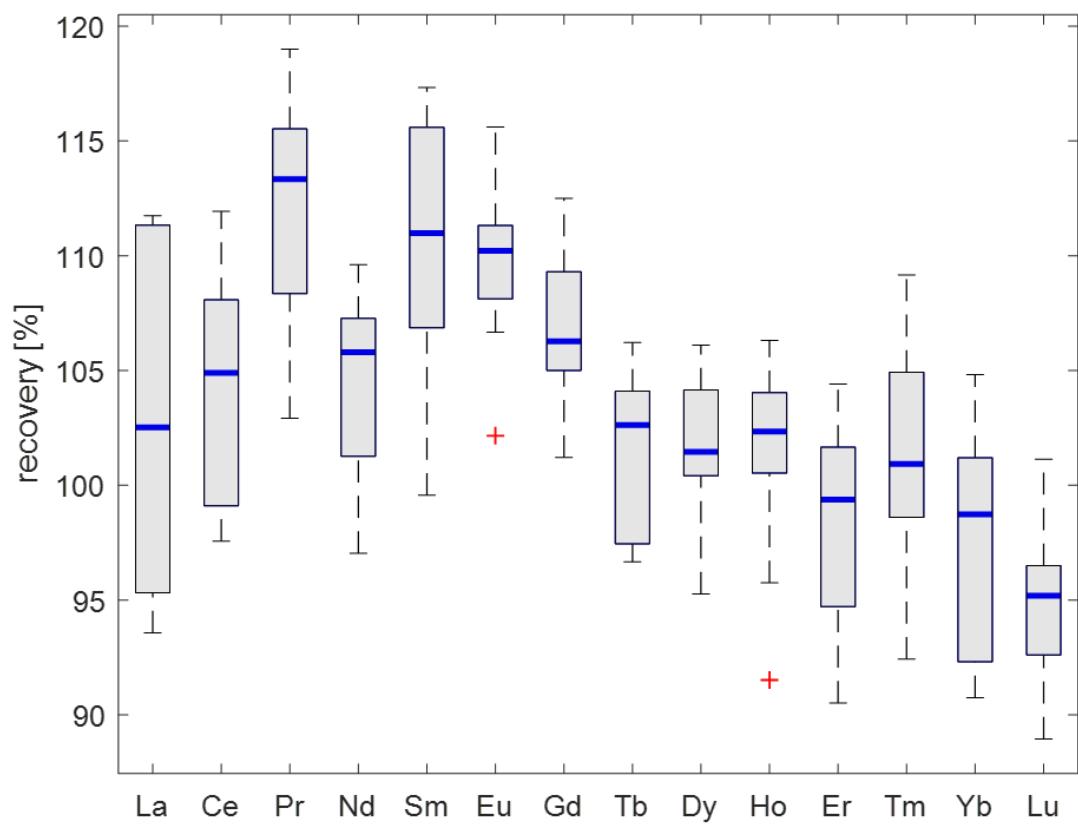


Figure S3: Recoveries of the REEs derived from multiple ($n = 10$) digestions and ICP MS measurements of the reference material BCR® – 667 (estuarine sediment). On each box, the central mark (blue line) indicates the median, and the bottom and top edges of the box indicate the 25th and 75th percentiles, respectively. The whiskers extend to the most extreme data points not considered outliers, and the outliers are plotted individually using the '+' symbol. Outliers are defined as values that deviate more than 1.5 times from the interquartile range (top or bottom of the box).

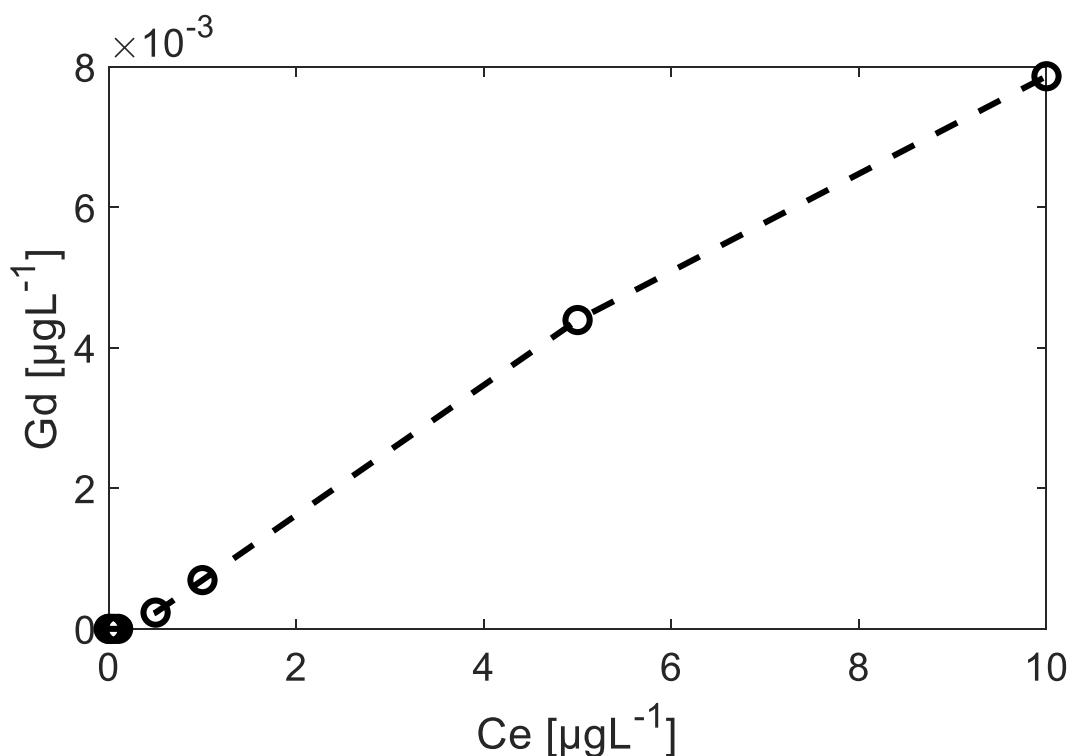


Figure S4: Interference of Ce (formation of CeOH) on ^{157}Gd . The counts of Gd were quantified based on a single element calibration. Correcting the Gd concentrations following this calibration curve only marginally affected the calculated, industrial fraction of Gd. Nevertheless, Gd values were corrected, accordingly.

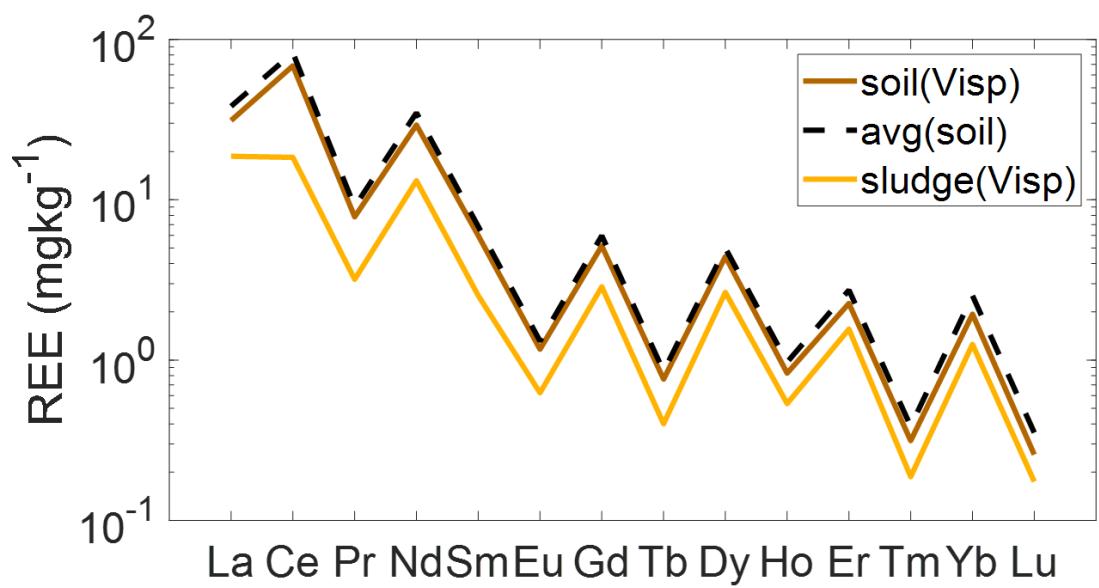


Figure S5: REE pattern of the sludge from the WWTP in Visp together with the average REE patterns of the soils and the soil collected within the catchment area of the WWTP of Visp. The REE pattern of the sludge from Visp is visibly depleted in Ce.

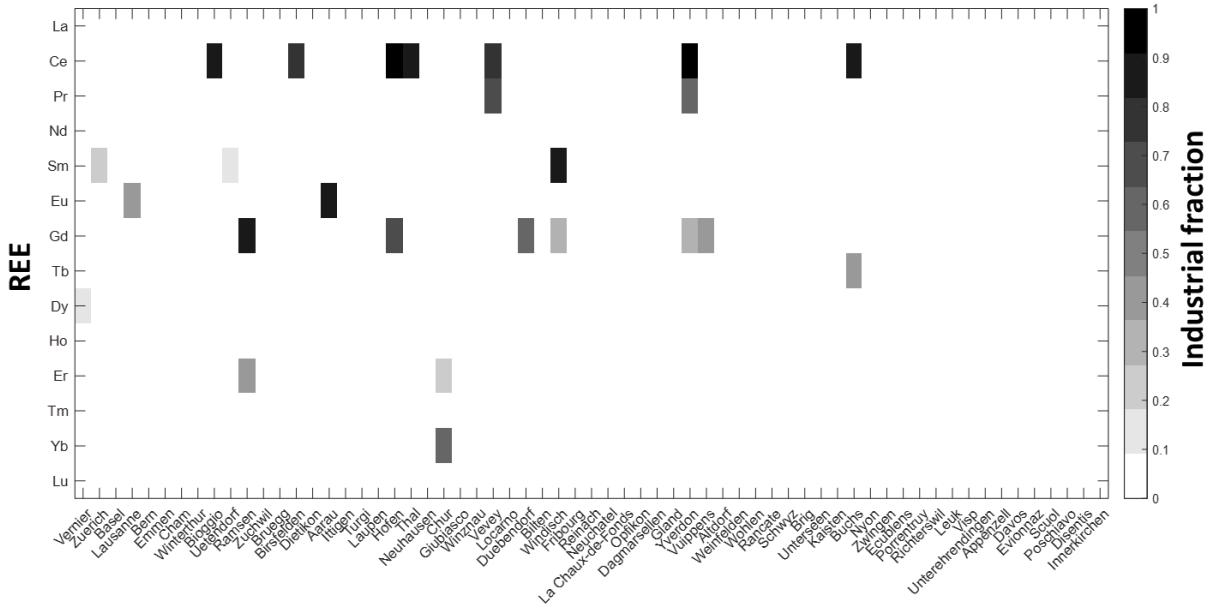


Figure S6: Heat map of the f_{REE}^{IND} for the individual REEs and the specific WWTPs based on the pattern fitting approach (A2). Increasing industrial fractions are represented by an increasing darkness.

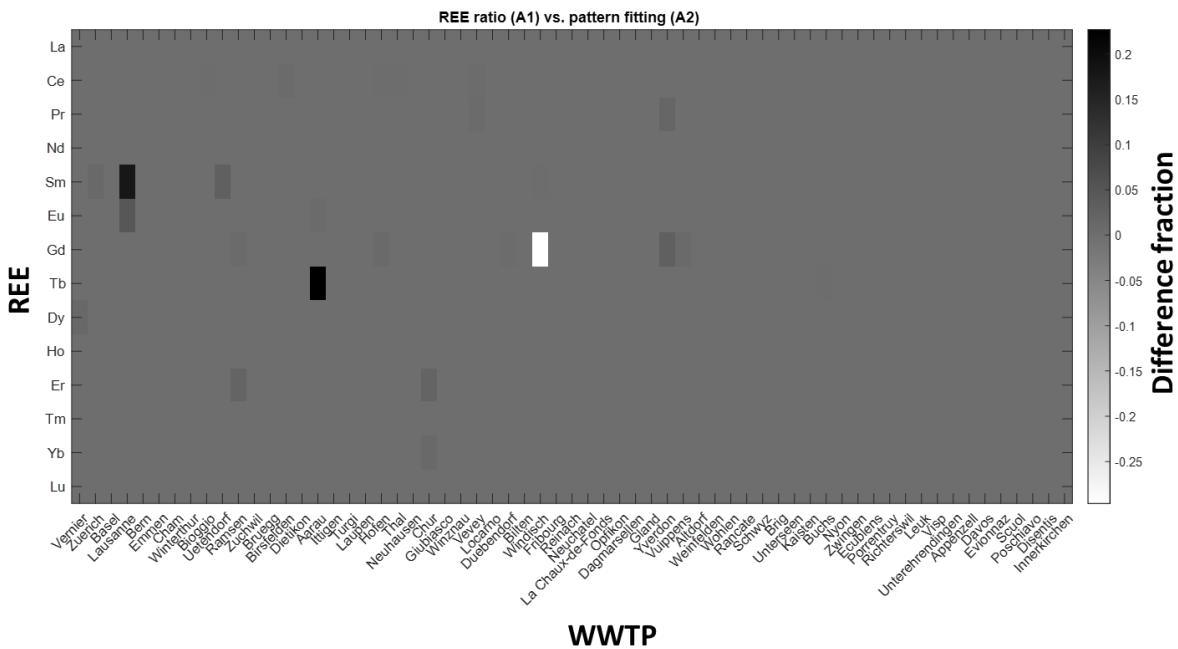


Figure S7: Difference heat map of the f_{REE}^{IND} for the individual REEs and the specific WWTPs based on the ratio (A1) and the pattern fitting (A2) approach. The WWTPs that receive substantial industrial fractions of a specific REE were almost identical for both approaches, which is reflected by the medium gray tone (0, identical result from both approaches) of the largest part of the difference heat map. Slight differences in the absolute industrial fractions are mostly within 5–10% and may be explained by variation of the REE patterns of the soils.

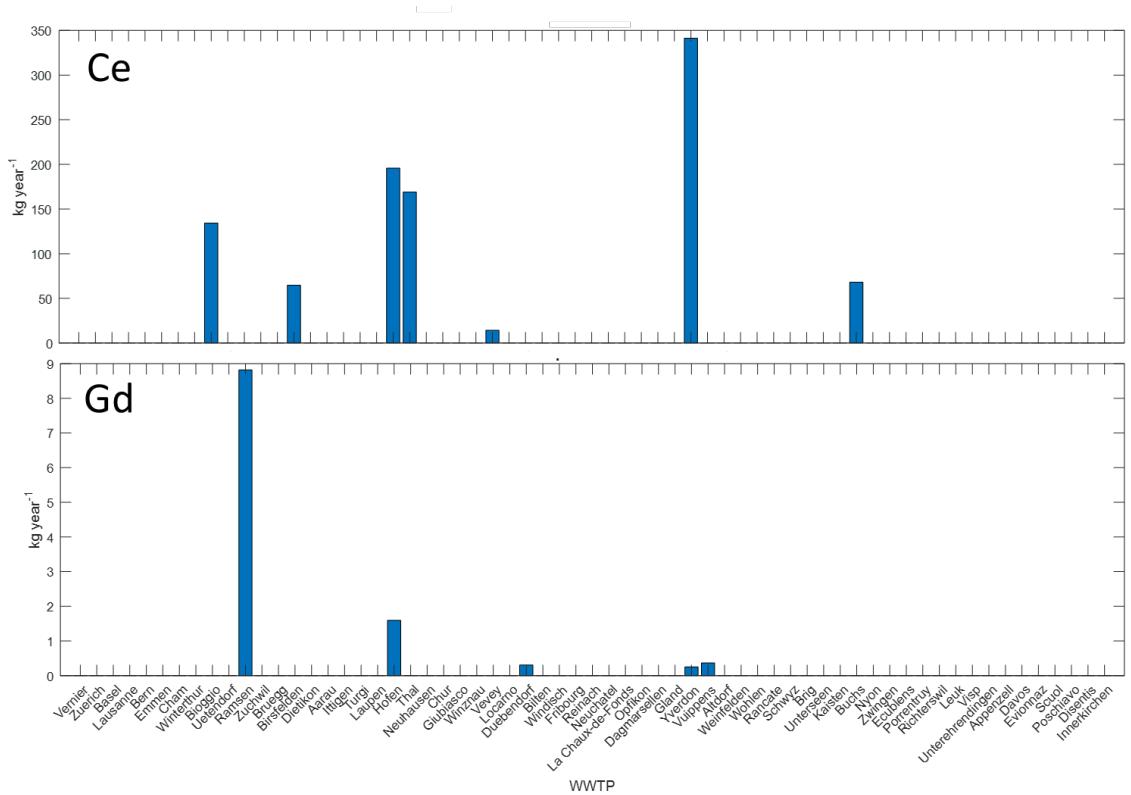


Figure S8: Annual fluxes of Ce (A) and Gd (B) from industrial sources calculated for the 63 WWTPs.

References:

- Vriens, B., Voegelin, A., Hug, S.J., Kaegi, R., Winkel, L.H.E., Buser, A.M., Berg, M., 2017. Quantification of Element Fluxes in Wastewaters: A Nationwide Survey in Switzerland. Environ. Sci. Technol. 51, 10943–10953.
<https://doi.org/10.1021/acs.est.7b01731>