Trace element behaviour in sediments of Ugandan part of Lake Victoria – Results from sequential extraction and chemometrical evaluation

Environmental Earth Science

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Supplementary information

Table S1 Indication of the element concentrations of the quality standards used to verify the calibration of each element, their certified concentrations (NIST 1640a and TMDA-64-3, both natural water standards; all data in [µg/l]; -- : not certified) and the recovery rate (RR) of the corresponding measurement. Both standards had been measured in a tenfold dilution. All elements except Li for the NIST 1640a standard fulfil the required recovery rate of 90–110%. Since the limit of determination for Li is 0.5 µg/l, this is to be expected measuring an effective concentration of 0.041 µg/l

Table S2 Indication of the element contents of the quality sediment standards used to validate the applied method (LGC6187 for pseudototal digestion; BC701 for BCR-extraction), their certified contents (all data in [mg/kg], except Fe ([g/kg])) and the recovery rate (RR) of the corresponding method. For the pseudototal digestion, all elements except V and Sn fulfil the required recovery rate of 90–110%. Both element contents though still lie inside the 95% confidence interval published by the company providing the sediment standard (LGC, England). Thus, the used pseudototal digestion method is validated. The BCR701 standard shows six of in total 24 element contents with a RR outside the boarders of 90–110%, affecting four different elements. Deviations might have occurred because of less experience with the BCR extraction in comparison with the laboratories participating in the validation of this method. Additionally, four of the six element contents originate from the pseudototal digestion of the residue after the third extraction step, which element contents only are indicative and not certified. Some uncertainty might have come from that as well

Table S3 Element contents [mg/kg] (except for [a]: [g/kg]) of the elements not listed in table 3 (paper), sediment pH and redox potential ΔE [mV] (T corrected) with indication of the Geo-Index I_{geo} in brackets (Class 0: 1–1.5x RV; Class 1: 1.5–3x RV; Class 2: 3–6x RV; Class 3: 6–12x RV; Class 4: 12–24x RV; *italics:* RV, **bold**: maximum value, NM: not measured)

Table S4 Complete indication of the results of the factor analysis using the normalised data containing pseudototal element contents, TP, LOI and the closest distance between shore and the SP (left: factor loadings (positive correlation (>0.7): light grey; negative correlation (<0.7): dark grey); right: factor scores)

Table S5 Indication of the fraction percentages (FP [%]; Eq. 1) of each of the studied elements resulting from the BCR-extraction (number indicates the fraction) *: Irregular data caused by Cu contamination of the 2nd fraction of the sequential extraction (as seen in the blanks of the according measurement series as well). After checking, no major influence of these measurement errors on the statistical analyses was detectable though, probably due to a sufficiently big dataset (Figs. 4 a) and S1 a); Tables 5 and S2)

Table S6 Indication of the summed content (SC [mg/kg] (except for [a]: [g/kg]); eq. 2) of each of the studied elements resulting from the BCR-extraction (nd = not determinable due to demonstrable Cu contamination in 2nd fraction) *: Irregular data caused by Cu contamination of the 2nd fraction of the sequential extraction (as seen in the blanks of the according measurement series as well). After checking, no major influence of these measurement errors on the cluster analysis was detectable though, probably due to a sufficiently big dataset (Fig. S1 b))

Table S7 a) Factor loadings resulting from the factor analysis using the normalised data containing the fraction percentages in each fraction of all of the studied elements (light grey = factor loading > 0.7, positive correlation; dark grey = factor loading < 0.7, negative correlation); b) to e) Indication of the associated factor scores for the different samples in each fraction of the sequential extraction

Fig S1 Cluster analyses of the element composition using Ward´s method with the squared Euclidean distance and Z-transformed fraction percentages (a) and summed contents (b)

Table S1 Indication of the element concentrations of the quality standards used to verify the calibration of each element, their certified concentrations (NIST 1640a and TMDA-64-3, both natural water standards; all data in [µg/l]; -- : not certified) and the recovery rate (RR) of the corresponding measurement. Both standards had been measured in a tenfold dilution. All elements except Li for the NIST 1640a standard fulfil the required recovery rate of 90–110%. Since the limit of determination for Li is 0.5 µg/l, this is to be expected measuring an effective concentration of 0.041 µg/l

	pseudototal digestion						BCR extraction					
	NIST 1640a			TMDA-64-3			NIST 1640a			TMDA-64-3		
	found	certified	RR [%]	found	certified	RR [%]	found	certified	RR [%]	found	certified	RR [%]
Li	n.d.	0.41		157	143	109.8	n.d.	0.41		147	143	102.8
Be	3.0	3.03	99.0	143	157	91.1	3.2	3.03	105.6	154	157	98.1
Mg	1071	1058.6	101.2	--	--	--	1069	1058.6	101.0	--	--	--
Al	57	53.0	107.5	304	291	104.5	50	53.0	94.3	281	291	96.6
Ca	5756	5615.0	102.5	--	--	--	5865	5615.0	104.5	--	--	--
Ti	--	--	--	124	123	100.8	--	--	--	123	123	100.0
V	15	15.05	99.7	288	279	103.2	15	15.05	99.7	286	279	102.5
Cr	40	40.54	98.7	295	283	104.2	41	40.54	101.1	287	283	101.4
Mn	39	40.39	96.6	303	292	103.8	40	40.39	99.0	297	292	101.7
Fe	37	36.8	100.5	320	298	107.4	37	36.80	100.5	307	298	103.0
Co	21	20.24	103.8	260	250	104.0	21	20.24	103.8	253	250	101.2
Ni	26	25.32	102.7	265	252	105.2	25	25.32	98.7	261	252	103.6
Cu	91	85.75	106.1	271	261	103.8	90	85.75	105.0	271	261	103.8
Zn	59	55.64	106.0	331	320	103.4	56	55.64	100.6	325	320	101.6
As	7.6	8.08	94.1	168	164	102.4	8.6	8.08	106.5	168	164	102.4
Se	19	20.13	94.4	158	154	102.6	22	20.13	109.3	157	154	101.9
Rb	1.2	1.2	100.2	31	30.9	100.3	1.2	1.2	100.2	31	30.9	100.3
Sr	125	126.03	99.2	658	628	104.8	126	126.03	100.0	661	628	105.3
Mo	46	45.60	100.9	304	286	106.3	46	45.6	100.9	298	286	104.2
Cd	4.1	3.99	102.7	270	258	104.7	4.1	3.99	102.7	268	258	103.9
Sn	--	--	--	282	281	100.4	--	--	--	284	281	101.1
Ba	155	151.8	102.1	308	287	107.3	154	151.8	101.4	303	287	105.6
Pb	12	12.1	99.2	289	280	103.2	13	12.1	107.4	292	280	104.3

Table S2 Element contents of the quality sediment standards used to validate the applied method (LGC6187 for pseudototal digestion; BC701 for BCR-extraction), their certified contents (all data in [mg/kg], except Fe ([g/kg])) and the recovery rate (RR) of the corresponding method. For the pseudototal digestion, all elements except V and Sn fulfil the required recovery rate of 90–110%. Both element contents though still lie inside the 95% confidence interval published by the company providing the sediment standard (LGC, England). Thus, the used pseudototal digestion method is validated. The BCR701 standard shows six of in total 24 element contents with a RR outside the boarders of 90–110%, affecting four different elements. Deviations might have occurred because of less experience with the BCR extraction in comparison with the laboratories participating in the validation of this method. Additionally, four of the six element contents originate from the pseudototal digestion of the residue after the third extraction step, which element contents only are indicative and not certified. Some uncertainty might have come from that as well

		V	Cr	Mn	Fe	Ni	Cu	Zn	As	Se	Cd	Sn	Pb
	found	44	89	1242	25	36	86	456	26	1.1	2.8	7.7	78
LGC6187	certified	38.3 ± 6.5	84 ± 9.4	1240 ± 60	23.6 ± 1.5	34.7 ± 1.7	83.6 ± 4.1	439 ± 26	24 ± 3.2	1.2 ± 0.2	2.7 ± 0.3	6.8 ± 1.8	77.2 ± 4.5
	RR [%]	114.9	106.0	100.2	106.3	103.7	102.9	103.9	108.3	91.7	103.7	113.2	101.0

		Cr	Ni	Cu	Zn	Cd	Pb
BCR701 – 1. fraction	found	2.7	15.1	48.0	216.0	7.5	3.1
	certified	2.26	15.4	49.3	205	7.34	3.18
	RR [%]	119.5	98.1	97.4	105.4	102.2	97.5
BCR701 – 2. fraction	found	47	28	135	116	3.7	124
	certified	45.7	26.6	124	114	3.77	126
	RR [%]	102.8	105.3	108.9	101.8	98.1	98.4
BCR701 – 3. fraction	found	137	16	67	46	0.26	10
	certified	143	15.3	55.2	45.7	0.27	9.3
	RR [%]	95.8	104.6	121.4	100.7	96.3	107.5
BCR701 – 4. fraction	found	75	37	34	96	0.12	13
	indicative	62.5	41.4	38.5	95	0.13	11
	RR [%]	120.0	89.4	88.3	101.1	92.3	118.2

Table S3 Indication of the element contents [mg/kg] (except for [a]: [g/kg]) of the elements not listed in table 3 (paper), sediment pH and redox potential ΔE [mV] (T corrected) with indication of the Geo-Index I_{geo} in brackets (Class 0: 1–1.5x RV; Class 1: 1.5–3x RV; Class 2: 3–6x RV; Class 3: 6–12x RV; Class 4: 12–24x RV; *italics:* RV, **bold**: maximum value, NM: not measured)

	Li	Be	Mg[a]	Al[a]	Ca[a]	Ti	V	Mn	Fe[a]	Co	Se	Rb	Sr	Mo	Sn	Ba	pH	ΔE
1_00–05	33 (2)	2.2 (1)	2.4 (1)	49 (1)	6.3 (1)	602 (1)	52 (0)	395 (2)	32 (0)	14 (1)	0.5 (1)	37 (1)	55 (0)	1.2 (0)	2.3 (1)	165 (0)	NM	NM
1_10-15	37 (2)	2.3 (1)	2.9 (1)	57 (1)	3.9 (0)	956 (1)	57 (1)	301 (1)	33 (1)	16 (1)	*<0.2* (0)	42 (1)	52 (0)	1.4 (0)	2.2 (1)	168 (0)	NM	NM
1_30-36	**66 (3)**	**4.0** (2)	**3.5** (1)	**81** (2)	3.0 (0)	1007 (1)	83 (1)	183 (1)	39 (1)	**16** (1)	0.4 (1)	**53** (1)	*46* (0)	3.8 (2)	2.8 (1)	163 (0)	NM	NM
2_00-05	13 (0)	1.3 (0)	2.7 (1)	34 (0)	4.6 (1)	906 (1)	42 (0)	550 (2)	40 (1)	12 (0)	1.0 (2)	34 (1)	73 (1)	2.0 (1)	2.2 (1)	**271 (1)**	7.2	61
2_10-15	20 (1)	2.5 (1)	3.2 (1)	46 (1)	4.2 (0)	979 (1)	65 (1)	804 (3)	**48** (1)	14 (1)	1.6 (3)	42 (1)	77 (1)	2.7 (1)	2.1 (0)	245 (1)	6	57
3_00-05	12 (0)	1.8 (1)	2.9 (1)	27 (0)	3.6 (0)	611 (1)	*35* (0)	**1331 (4)**	34 (1)	*8.3* (0)	1.5 (3)	37 (1)	65 (0)	*1.0* (0)	**3.1** (1)	251 (1)	7	41
3_10-15	12 (0)	2.0 (1)	3.1 (1)	32 (0)	3.8 (0)	1064 (1)	49 (0)	1224 (3)	34 (1)	9.5 (0)	0.3 (0)	40 (1)	70 (1)	1.0 (0)	*1.4* (0)	230 (1)	6.8	49
3_24-30	12 (0)	2.3 (1)	2.9 (1)	*25* (0)	3.4 (0)	*391* (0)	39 (0)	1107 (3)	29 (0)	9.1 (0)	0.4 (1)	34 (1)	63 (0)	1.1 (0)	1.6 (0)	185 (0)	6.7	30
4_00-05	13 (0)	1.4 (0)	1.9 (0)	39 (1)	**9.4** (2)	629 (1)	69 (1)	248 (1)	34 (1)	11 (0)	**1.8** (3)	26 (0)	**109** (1)	3.8 (2)	2.1 (0)	207 (1)	7	41
4_10-15	15 (0)	1.7 (1)	1.9 (0)	34 (0)	3.8 (0)	693 (1)	76 (1)	170 (1)	30 (0)	12 (0)	1.7 (3)	24 (0)	64 (0)	3.5 (2)	1.8 (0)	148 (0)	6.8	49
4_32-38	14 (0)	1.7 (1)	1.6 (0)	27 (0)	3.1 (0)	425 (0)	78 (1)	132 (0)	29 (0)	13 (1)	1.0 (2)	*21* (0)	50 (0)	3.4 (2)	2.8 (1)	*125* (0)	6.7	30
5_00-05	14 (0)	1.3 (0)	2.0 (0)	37 (0)	4.2 (0)	816 (1)	76 (1)	166 (1)	23 (0)	9.9 (0)	0.9 (2)	22 (0)	76 (1)	3.4 (2)	1.5 (0)	167 (0)	NM	NM
5_10-15	15 (0)	1.2 (0)	1.6 (0)	40 (1)	*2.9* (0)	807 (1)	73 (1)	*109* (0)	*21* (0)	11 (0)	0.6 (1)	23 (0)	57 (0)	4.5 (2)	1.7 (0)	130 (0)	NM	NM
6_00-05	15 (0)	1.2 (0)	1.8 (0)	42 (1)	4.8 (1)	938 (1)	74 (1)	202 (1)	28 (0)	11 (0)	0.9 (2)	26 (0)	74 (1)	3.1 (2)	1.8 (0)	164 (0)	NM	NM
6_10-15	14 (0)	1.3 (0)	*1.5* (0)	39 (1)	3.0 (0)	905 (1)	69 (1)	134 (0)	23 (0)	9.5 (0)	0.4 (1)	24 (0)	51 (0)	3.1 (2)	1.6 (0)	128 (0)	NM	NM
7_00-05	13 (0)	1.0 (0)	1.8 (0)	42 (1)	4.5 (1)	1109 (1)	75 (1)	247 (1)	38 (1)	13 (1)	0.5 (1)	21 (0)	80 (1)	**4.8** (2)	1.7 (0)	169 (0)	NM	NM
7_05-10	*11* (0)	*0.9* (0)	1.7 (0)	37 (0)	4.5 (1)	1159 (1)	65 (1)	230 (1)	38 (1)	12 (0)	0.8 (2)	22 (0)	72 (1)	3.8 (2)	1.8 (0)	175 (0)	NM	NM
7_10-15	17 (1)	1.6 (1)	1.8 (0)	46 (1)	4.6 (1)	**1329** (2)	**89** (1)	266 (1)	39 (1)	15 (1)	0.6 (1)	22 (0)	72 (1)	4.1 (2)	1.7 (0)	157 (0)	NM	NM
Range	11 - 66	0.9 - 4.0	1.5 - 3.5	25 - 81	2.9 - 9.4	391 - 1329	35 - 89	109 - 1331	21 - 48	8.3 - 16	0.2 - 1.8	21 - 53	46 - 109	1.0 - 4.8	1.4 - 3.1	125 - 271		
Mean	19	1,8	2284	40728	4313	851	65	433	32808	12	0,9	31	67	2,9	2,0	181		
Median	14	1,7	1979	39034	4056	905	69	248	33301	12	0,8	26	68	3,2	1,8	167		

legend Class 0 | Class 1 | Class 2 | Class 3 | Class 4 (I geo)

Table S4 Complete indication of the results of the factor analysis using the normalised data containing pseudototal element contents, TP, LOI and the closest distance between shore and the SP (left: factor loadings (positive correlation (> 0.7): light grey; negative correlation (< -0.7): dark grey); right: factor scores)

	F1	**F2**	**F3**
Li	-0.19	0.92	-0.24
Be	0.27	0.84	-0.16
Mg	0.66	0.71	0.13
Al	-0.43	0.86	0.00
Ca	-0.08	-0.11	0.51
Ti	-0.48	0.29	0.42
V	-0.89	0.03	0.05
Cr	-0.96	0.15	0.08
Mn	0.92	0.05	0.15
Fe	0.10	0.54	0.69
Co	-0.50	0.79	-0.02
Ni	-0.38	0.83	0.01
Cu	-0.60	0.03	0.33
Zn	0.29	0.65	0.44
As	-0.29	-0.25	0.70
Se	0.13	-0.30	0.53
Rb	0.50	0.84	-0.01
Sr	0.01	-0.37	0.81
Mo	-0.85	-0.16	0.25
Cd	0.48	-0.29	0.36
Sn	0.19	0.49	-0.05
Ba	0.69	0.14	0.64
Pb	-0.56	-0.35	0.65
LOI	-0.32	-0.84	0.32
TP	0.49	-0.52	0.38
distance	0.91	-0.08	0.01

	F1	**F2**	**F3**
1_00–05	0.22	0.68	-1.25
1_10-15	-0.06	1.39	-1.22
1_30-36	-0.68	3.15	-0.18
2_00-05	0.80	0.02	1.20
2_10-15	0.70	0.77	1.74
3_00-05	1.98	-0.11	0.15
3_10-15	1.48	0.02	-0.05
3_24-30	2.00	-0.48	-0.71
4_00-05	-0.17	-0.66	1.75
4_10-15	-0.40	-0.62	-0.43
4_32-38	-0.40	-0.71	-1.31
5_00-05	-0.38	-0.94	-0.17
5_10-15	-0.74	-0.89	-0.94
6_00-05	-0.57	-0.54	0.16
6_10-15	-0.54	-0.68	-1.08
7_00-05	-1.01	-0.13	0.73
7_05-10	-0.87	-0.28	0.74
7_10-15	-1.35	0.03	0.88

Table S5 Indication of the fraction percentages (FP [%]; Eq. 1) of each of the studied elements resulting from the BCR-extraction (number indicates the fraction) *: Irregular data caused by Cu contamination of the 2nd fraction of the sequential extraction (as seen in the blanks of the according measurement series as well). After checking, no major influence of these measurement errors on the statistical analyses was detectable though, probably due to a sufficiently big dataset (Figs. 4 a) and S1 a); Tables 5 and S2)

	As.1	**As.2**	**As.3**	**As.4**	**Cd.1**	**Cd.2**	**Cd.3**	**Cd.4**	**Co.1**	**Co.2**	**Co.3**	**Co.4**	**Cr.1**	**Cr.2**	**Cr.3**	**Cr.4**	**Cu.1**	**Cu.2**	**Cu.3**	**Cu.4**	**Ni.1**	**Ni.2**	**Ni.3**	**Ni.4**
1_00–05	1	3	22	73	38	41	10	11	10	10	37	43	0	6	20	73	0	36	18	46	5	6	19	70
1_10–15	2	4	26	69	58	42	0	0	11	10	40	39	0	6	26	68	0	36	28	36	7	8	23	61
1_30–36	1	11	20	68	48	31	17	4	14	9	37	40	0	2	29	69	0	32	41	27	12	10	21	57
2_00–05	0	6	46	47	42	46	5	7	11	7	38	45	1	5	26	68	0*	41*	24*	35*	4	6	30	60
2_10–15	0	3	55	42	44	43	4	9	10	5	48	37	0	5	32	63	0	21	30	49	7	6	33	54
3_00–05	1	4	64	32	41	44	11	4	7	9	35	49	0	2	18	80	0	24	33	44	4	7	27	63
3_10–15	0	3	55	41	39	42	13	5	8	9	35	48	0	2	24	74	0	27	39	34	5	6	33	56
3_24–30	0	3	53	44	43	39	10	8	7	8	33	52	0	2	17	81	0	24	23	54	6	6	28	60
4_10–15	0	8	50	41	46	50	0	3	6	3	61	29	0	2	45	52	0*	36*	40*	24*	6	6	36	52
4_32–38	1	4	56	39	53	31	9	7	7	4	63	26	1	3	49	47	0	20	45	35	9	7	37	46
5_00–05	1	3	45	52	53	31	10	6	7	4	60	30	0	2	42	56	0	20	51	29	7	6	31	56
5_10–15	2	3	50	45	61	35	4	1	10	4	62	25	0	2	49	49	0	30	50	20	12	6	33	49
6_00–05	1	1	50	48	41	53	3	3	4	3	62	30	0	2	40	58	0	18	46	37	4	5	27	63
6_10–15	2	2	50	47	53	36	9	3	10	5	58	27	0	2	47	51	0	22	50	27	8	6	28	57
7_00–05	2	2	38	59	44	46	3	7	4	3	56	38	0	3	38	59	0	17	54	29	4	4	27	66
7_05–10	0	6	32	63	41	42	8	9	3	3	60	35	0	2	39	59	0*	33*	46*	20*	4	4	25	68
7_10–15	1	1	41	58	45	31	11	13	6	4	59	32	0	3	45	52	0	17	62	21	4	5	30	61

Table S5 continuation

	Pb.1	Pb.2	Pb.3	Pb.4	Zn.1	Zn.2	Zn.3	Zn.4	Li.1	Li.2	Li.3	Li.4	Be.1	Be.2	Be.3	Be.4	Mg.1	Mg.2	Mg.3	Mg.4	Al.1	Al.2	Al.3	Al.4
1_00–05	1	56	14	29	11	17	8	64	1	1	17	82	18	31	24	28	19	7	12	62	0	3	5	91
1_10–15	2	55	18	26	10	17	11	63	1	1	14	85	19	24	33	24	17	9	15	59	0	3	7	90
1_30–36	2	60	17	21	11	16	22	52	1	0	10	89	18	24	34	24	14	8	18	61	0	1	6	93
2_00–05	1	48	11	40	14	24	9	54	1	1	16	82	25	36	20	19	22	6	12	60	0	4	5	90
2_10–15	1	35	19	44	14	18	11	57	1	0	14	84	17	23	31	29	19	6	15	60	0	3	8	89
3_00–05	1	53	21	24	10	26	14	50	1	1	18	80	19	28	22	32	18	8	17	57	0	3	12	85
3_10–15	2	51	22	25	7	20	15	57	1	1	14	84	18	27	25	29	16	8	19	58	0	3	13	84
3_24–30	1	47	19	33	7	18	10	64	1	0	14	85	12	20	34	33	15	8	13	65	0	2	9	89
4_10–15	1	37	16	46	20	21	13	47	0	0	15	84	21	43	18	17	27	7	17	49	0	3	8	89
4_32–38	1	32	21	47	19	18	11	52	1	0	14	85	17	22	31	30	19	6	21	54	0	3	9	88
5_00–05	2	49	21	28	27	22	13	38	1	1	21	77	16	30	24	30	34	7	19	39	1	6	10	83
5_10–15	2	41	21	36	27	19	13	40	1	1	24	74	20	27	26	27	29	6	21	44	1	5	13	81
6_00–05	1	49	18	32	27	22	10	41	1	1	23	76	15	29	24	33	30	7	16	46	0	4	8	88
6_10–15	2	49	18	31	16	14	25	44	1	0	19	80	23	27	22	28	25	6	19	50	0	5	10	85
7_00–05	2	50	22	26	27	21	13	39	1	1	23	75	16	22	21	40	35	7	17	41	0	3	9	87
7_05–10	1	55	16	28	31	22	11	36	2	1	21	76	9	34	20	37	35	6	15	45	0	4	8	88
7_10–15	1	51	22	26	21	21	14	44	1	1	21	77	13	32	22	33	28	6	21	44	0	3	10	86

Table S5 continuation

	Ca.1	Ca.2	Ca.3	Ca.4	Ti.1	Ti.2	Ti.3	Ti.4	V.1	V.2	V.3	V.4	Mn.1	Mn.2	Mn.3	Mn.4	Fe.1	Fe.2	Fe.3	Fe.4	Rb.1	Rb.2	Rb.3	Rb.4
1_00–05	73	20	1	6	0	0	5	95	1	42	13	44	44	32	7	16	1	30	9	60	2	4	4	90
1_10–15	62	28	4	6	0	0	4	96	1	41	13	45	33	35	9	22	1	26	13	59	2	4	7	87
1_30–36	62	31	2	5	0	0	0	99	2	38	15	44	36	22	11	31	2	14	26	58	2	3	5	90
2_00–05	68	24	4	4	0	0	7	93	0	38	16	46	52	25	7	16	0	29	14	57	2	5	5	88
2_10–15	65	24	5	7	0	0	6	94	1	36	20	44	51	28	9	11	1	32	19	48	2	4	7	87
3_00–05	55	30	5	10	0	0	1	99	0	25	19	56	48	38	6	7	0	25	15	60	3	6	14	77
3_10–15	54	31	6	9	0	0	1	99	0	23	21	56	51	34	7	8	0	19	17	63	3	6	17	74
3_24–30	50	30	7	12	0	0	7	93	0	22	18	60	52	34	5	9	0	18	13	69	2	4	9	84
4_10–15	66	25	5	4	0	0	6	94	2	47	20	31	33	17	14	36	1	21	20	57	3	3	7	87
4_32–38	63	26	6	5	0	0	8	91	1	49	24	26	35	17	15	33	2	28	22	48	2	3	10	85
5_00–05	70	22	4	5	0	0	4	96	2	41	25	31	47	16	9	29	1	15	16	68	4	4	11	81
5_10–15	69	21	4	6	0	0	11	88	5	43	25	27	40	12	11	37	2	13	27	59	3	4	11	83
6_00–05	71	23	2	4	0	0	31	69	2	37	23	38	47	18	6	29	1	19	10	70	2	4	6	88
6_10–15	70	23	3	3	0	0	6	94	3	43	22	32	42	14	7	37	2	19	14	65	2	3	7	88
7_00–05	69	20	3	7	0	0	1	98	1	27	23	49	24	15	11	50	1	17	13	68	3	4	10	82
7_05–10	72	19	4	5	0	0	5	94	1	34	19	46	20	15	11	54	1	16	8	75	3	4	6	86
7_10–15	66	21	4	9	0	0	3	97	1	34	25	40	21	18	11	50	1	17	16	66	3	4	11	82

Table S5 continuation

	Sr.1	Sr.2	Sr.3	Sr.4	Se.1	Se.2	Se.3	Se.4	Mo.1	Mo.2	Mo.3	Mo.4	Sn.1	Sn.2	Sn.3	Sn.4	Ba.1	Ba.2	Ba.3	Ba.4
1_00–05	68	20	2	10	1	1	91	7	0	1	53	46	0	0	0	100	34	41	5	21
1_10–15	57	26	3	14	1	4	87	8	0	1	41	58	0	0	0	100	25	45	7	23
1_30–36	51	32	4	13	5	6	83	6	0	1	19	80	0	0	0	100	8	51	9	32
2_00–05	56	21	4	19	6	4	85	5	0	1	76	23	0	0	0	99	32	39	9	20
2_10–15	52	21	6	21	0	4	86	9	0	1	64	35	0	0	0	100	23	44	11	22
3_00–05	50	27	7	16	1	5	80	14	1	2	27	70	0	0	1	99	32	45	11	12
3_10–15	48	27	7	18	0	3	86	11	1	1	25	73	0	0	1	99	37	37	12	15
3_24–30	45	28	6	21	0	5	90	5	0	1	46	52	0	0	0	100	34	38	9	19
4_10–15	50	20	5	26	2	3	95	1	0	1	52	47	0	0	1	99	14	42	12	33
4_32–38	44	20	6	30	1	2	90	7	1	1	52	46	0	0	0	100	22	33	12	33
5_00–05	62	18	4	16	0	3	71	26	0	0	38	61	0	0	0	99	33	34	10	23
5_10–15	60	17	5	18	1	0	88	11	0	1	39	60	0	0	1	99	21	41	13	26
6_00–05	62	19	3	17	0	0	86	14	0	0	79	21	0	0	0	100	36	32	7	24
6_10–15	59	19	4	19	0	1	84	15	0	0	74	25	0	0	0	100	36	31	9	24
7_00–05	59	17	4	20	1	2	76	20	0	0	23	77	0	0	0	99	30	35	10	25
7_05–10	58	16	3	22	19	7	72	2	0	0	77	23	0	0	0	100	24	40	9	26
7_10–15	54	17	4	25	0	2	85	12	0	0	27	73	0	0	0	99	22	37	12	29

Table S6 Indication of the summed content (SC [mg/kg] (except for ^{a}: [g/kg]); eq. 2) of each of the studied elements resulting from the BCR-extraction (nd = not determinable due to demonstrable Cu contamination in 2nd fraction) *: Irregular data caused by Cu contamination of the 2nd fraction of the sequential extraction (as seen in the blanks of the according measurement series as well). After checking, no major influence of these measurement errors on the cluster analysis was detectable though, probably due to a sufficiently big dataset (Fig. S1 b))

	As	**Cd**	**Co**	**Cr**	**Cu**	**Ni**	**Pb**	**Zn**	**Li**	**Be**	**Mg^{a}**	**Al^{a}**	**Ca^{a}**	**Ti**	**V**	**Mn**	**Fe^{a}**	**Rb**	**Sr**	**Se**	**Mo**	**Sn**	**Ba**
1_00–05	2.6	0.06	12	48	26	29	11	58	28	1.6	2.1	36	6.5	554	49	368	28	34	58	0.5	1.2	2.6	148
1_10–15	2.4	0.06	14	54	30	32	12	67	32	2.0	2.6	40	3.9	617	53	262	28	38	49	0.7	1.5	2.5	144
1_30–36	3.4	0.12	15	64	38	41	15	89	47	2.9	2.9	55	3.0	592	68	164	31	44	41	1.2	3.2	3.3	127
2_00–05	5.7	0.22	10	42	96*	22	17	75	12	1.4	2.6	29	4.7	578	53	516	37	31	72	2.0	2.0	2.1	225
2_10–15	5.8	0.23	11	45	32	23	18	83	18	2.2	3.0	34	4.5	716	57	765	42	38	76	2.0	2.5	2.6	222
3_00–05	3.5	0.24	8.0	36	34	23	14	83	11	1.7	3.0	24	3.9	613	39	1369	32	33	68	1.4	1.1	4.4	250
3_10–15	4.3	0.22	8.2	29	36	19	14	76	12	2.2	3.1	26	3.8	699	42	1235	30	36	67	1.4	1.1	2.5	219
3_24–30	3.1	0.24	8.4	28	25	18	14	85	13	2.3	3.0	27	3.7	746	40	1099	29	38	64	1.4	1.0	2.8	183
4_10–15	4.9	0.10	11	65	96*	19	16	59	12	1.2	1.8	28	4.0	533	73	154	28	22	60	1.7	3.0	2.0	134
4_32–38	4.6	0.14	12	65	29	19	17	60	13	1.6	1.6	31	3.4	624	79	135	28	24	52	1.5	3.2	3.3	129
5_00–05	4.7	0.18	8.3	63	39	20	20	62	11	1.3	2.0	28	4.4	625	69	158	19	20	75	1.4	3.1	2.5	157
5_10–15	5.3	0.16	9.2	64	46	19	15	53	12	1.1	1.5	25	3.2	678	65	96	18	18	55	1.2	5.2	2.3	114
6_00–05	5.2	0.15	9.6	65	39	21	21	64	13	1.1	1.8	33	4.9	784	62	197	22	24	75	1.3	3.4	2.7	157
6_10–15	4.5	0.12	8.7	63	39	19	18	51	12	0.9	1.5	28	3.1	703	63	127	19	21	51	1.2	3.7	2.6	123
7_00–05	4.5	0.14	12	73	78	24	25	73	14	0.9	1.9	33	5.0	1027	73	217	33	20	77	1.3	3.7	2.6	156
7_05–10	4.6	0.14	11	72	243*	22	22	70	11	0.6	1.5	26	4.3	592	74	176	32	17	61	1.0	3.6	2.0	130
7_10–15	5.0	0.17	12	78	101	24	23	57	15	1.1	1.7	33	5.0	1075	80	209	32	19	69	1.3	4.4	2.6	136

Table S7 a) Factor loadings resulting from the factor analysis using the normalised data containing the fraction percentages in each fraction of all of the studied elements (light grey = factor loading > 0.7, positive correlation; dark grey = factor loading < 0.7, negative correlation)

b) to e) Indication of the associated factor scores for the different samples in each fraction of the sequential extraction

a)	As	Cd	Co	Cr	Cu	Ni	Pb	Zn	Li	Be	Mg	Al	Ca	Ti	V	Mn	Fe	Rb	Sr	Se	Mo	Sn	Ba
F1	0.61	-0.53	0.33	0.80	0.17	0.87	0.05	0.89	0.95	0.24	0.94	0.97	-0.30	0.97	0.49	0.24	0.86	0.97	-0.04	-0.29	0.46	0.98	-0.07
F2	0.70	-0.78	0.90	0.55	0.65	0.43	-0.16	-0.10	0.24	0.11	0.07	0.11	-0.67	0.10	0.08	-0.65	0.09	0.09	-0.74	0.93	0.77	0.03	-0.84
F3	0.15	-0.17	0.03	0.18	0.63	0.15	0.94	0.19	0.16	0.72	-0.18	0.20	-0.64	0.16	0.81	-0.34	0.45	0.19	-0.63	-0.04	0.05	0.17	0.34

fraction 1	F1	F2	F3
1_00–05	-0.29	-0.96	-1.51
1_10–15	-0.45	-0.77	-1.33
1_30–36	-0.45	-0.41	-1.53
2_00–05	-0.30	-0.95	-1.27
2_10–15	-0.30	-0.80	-1.51
3_00–05	-0.39	-0.85	-1.27
3_10–15	-0.40	-0.90	-1.19
3_24–30	-0.41	-0.84	-1.36
4_10–15	-0.33	-0.61	-1.43
4_32–38	-0.39	-0.70	-1.33
5_00–05	-0.09	-1.08	-1.59
5_10–15	-0.23	-0.63	-1.72
6_00–05	-0.11	-0.91	-1.73
6_10–15	-0.28	-1.02	-1.24
7_00–05	-0.22	-0.73	-1.57
7_05–10	-0.23	-0.81	-1.56
7_10–15	-0.34	-0.84	-1.34

b)

fraction 2	F1	F2	F3
1_00–05	-0.75	-0.88	1.59
1_10–15	-0.68	-0.96	1.30
1_30–36	-0.77	-0.90	1.34
2_00–05	-0.76	-0.82	1.62
2_10–15	-0.66	-0.88	0.90
3_00–05	-0.60	-1.08	1.04
3_10–15	-0.68	-0.87	0.89
3_24–30	-0.67	-0.83	0.61
4_10–15	-0.84	-0.85	1.77
4_32–38	-0.70	-0.60	0.87
5_00–05	-0.79	-0.69	1.29
5_10–15	-0.90	-0.75	1.66
6_00–05	-0.79	-0.91	1.25
6_10–15	-0.85	-0.66	1.26
7_00–05	-0.81	-0.81	1.02
7_05–10	-0.91	-0.63	1.55
7_10–15	-0.81	-0.66	1.22

c)

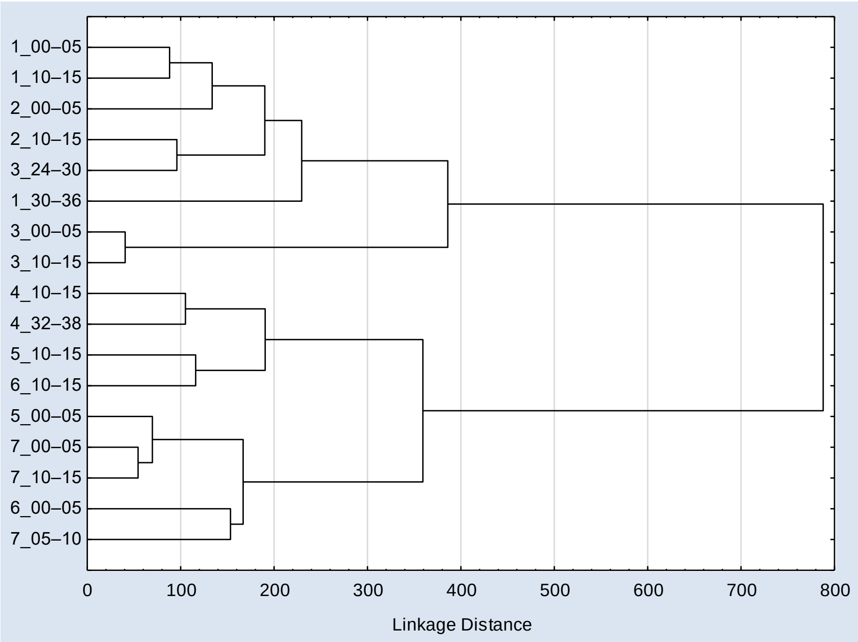
fraction 3	F1	F2	F3
1_00–05	-0.82	1.48	-0.38
1_10–15	-0.75	1.43	-0.02
1_30–36	-0.72	1.12	0.29
2_00–05	-0.63	1.66	-0.54
2_10–15	-0.58	1.61	-0.02
3_00–05	-0.61	1.32	-0.11
3_10–15	-0.59	1.28	0.06
3_24–30	-0.71	1.38	0.04
4_10–15	-0.50	1.79	-0.40
4_32–38	-0.48	1.68	0.07
5_00–05	-0.56	1.56	0.09
5_10–15	-0.63	1.74	-0.31
6_00–05	-0.56	1.90	-0.12
6_10–15	-0.49	1.80	-0.14
7_00–05	-0.64	1.48	0.09
7_05–10	-0.63	1.77	-0.33
7_10–15	-0.59	1.61	0.10

d)

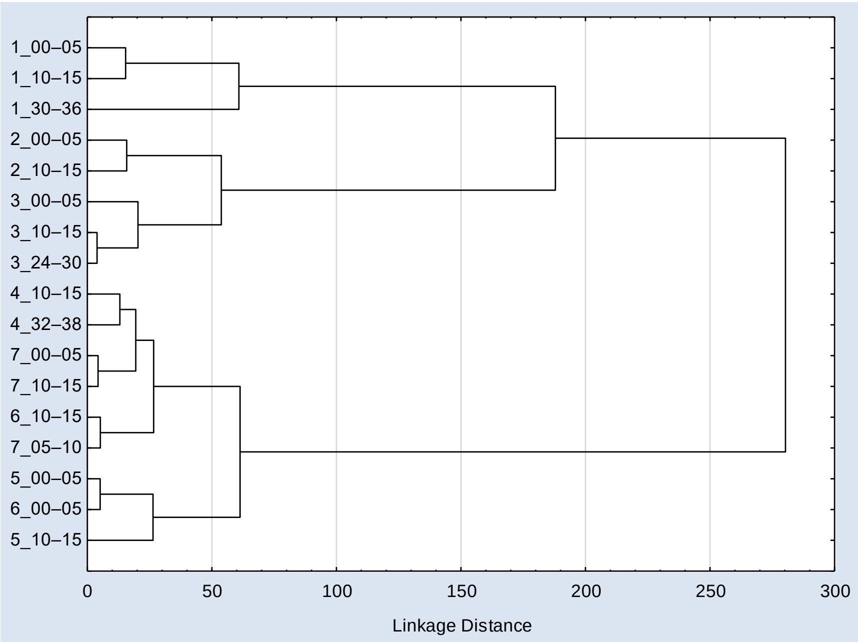
fraction 4	F1	F2	F3
1_00–05	1.85	0.37	0.30
1_10–15	1.88	0.30	0.04
1_30–36	1.94	0.19	-0.10
2_00–05	1.69	0.11	0.19
2_10–15	1.55	0.07	0.63
3_00–05	1.60	0.60	0.34
3_10–15	1.68	0.48	0.23
3_24–30	1.79	0.28	0.71
4_10–15	1.66	-0.33	0.06
4_32–38	1.57	-0.38	0.40
5_00–05	1.44	0.22	0.21
5_10–15	1.76	-0.36	0.37
6_00–05	1.47	-0.09	0.60
6_10–15	1.62	-0.12	0.12
7_00–05	1.67	0.06	0.46
7_05–10	1.76	-0.33	0.34
7_10–15	1.74	-0.11	0.02

e)

Fig S1 Cluster analyses of the element composition using Ward´s method with the squared Euclidean distance and Z-transformed (a) fraction percentages and (b) summed contents



a)



b)