Supplementary data for the article:

Milićević, T.; Relić, D.; Urošević, M. A.; Vuković, G.; Škrivanj, S.; Samson, R.; Popović, A. Integrated Approach to Environmental Pollution Investigation – Spatial and Temporal Patterns of Potentially Toxic Elements and Magnetic Particles in Vineyard through the Entire Grapevine Season. *Ecotoxicology and Environmental Safety* **2018**, *163*, 245–254. https://doi.org/10.1016/j.ecoenv.2018.07.078

Tables

Table S1: The descriptive statistics of physicochemical parameters: pH−acidity (-), SOM−soil organic matter (%), CEC−cation exchange capacity (cmol kg^{-1}), N (%), C (%) and H (%)

0–30 cm; 30–60 cm		April	May	June	July	August
pH (H_2O)	M	7.51	7.86	7.51	7.57	7.29
	SD	0.38	0.32	0.33	0.74	0.76
	Range	7.06–7.88	7.70–8.43	7.28–7.74	6.54–8.57	7.06–7.88
	C			6.57		
	SD			0.16		
pH (1 M KCl)	M	6.46	6.85	6.29	6.08	6.42
	SD	0.24	0.38	0.33	0.81	0.87
	Range	6.33–6.92	6.56–7.47	6.06–6.52	5.15–7.31	6.33–6.92
	C			5.09		
	SD			0.15		
pH (0.1 M $CaCl_2$)	M	6.73	7.27	6.74	6.68	6.91
	SD	0.21	0.33	0.43	0.77	0.83
	Range	6.53–7.06	6.98–7.77	6.43–7.04	5.60–7.71	6.53–7.06
	C			5.57		
	SD			0.12		
SOM (%)	M	0.81	0.96	0.89	0.96	0.79
	SD	0.08	0.13	0.27	0.12	0.17
	Range	0.76–0.81	0.72–1.03	0.70–1.09	0.81–2.06	0.48–0.96
	C			0.92		
	SD			0.09		
CEC (cmol kg^{-1})	M	28.45	27.89	28.02	27.64	26.58
	SD	3.10	2.02	3.76	2.29	2.88
	Range	22.03–30.34	24.03–28.35	22.13–32.20	23.92–29.34	22.72–31.34
	C			22.06		
	SD			2.63		
N (%)	M	0.13	0.16	0.16	0.14	0.14
	SD	0.02	0.03	0.02	0.01	0.03
	Range	0.11–0.16	0.10–0.16	0.14–0.21	0.13–0.15	0.08–0.16
	C			0.16		
	SD			0.04		
C (%)	M	1.68	1.57	1.71	1.37	1.61
	SD	0.22	0.50	0.27	0.09	1.04
	Range	1.36–1.87	1.04–2.25	0.52–1.98	1.29–1.49	1.06–4.09
	C			1.65		
	SD			0.41		
H (%)	M	0.80	0.77	0.80	0.81	0.77
	SD	0.04	0.07	0.05	0.03	0.11
	Range	0.79–0.89	0.74–0.89	0.62–0.98	0.78–0.85	0.53–0.84
	C			0.74		
	SD			0.05		

Table S2: The descriptive statistics of pseudo-total element concentrations (mg kg^{-1}) (M−Median, Min−Minimum, Max−Maximum, Mean, SD−Standard Deviation, RSD%−Relative Standard Deviation, MAC−Maximum Allowable Concentrations) of 23 elements (Al, As, B, Ba, Be, Ca, Cd, Co, Cr, Cu, Fe, K, Mg, Mn, Na, Ni, P, Pb, S, Sb, Sr, V and Zn) measured in the topsoil and the subsoil samples (n=182) collected through the grapevine season

	Al	As	B	Ba	Be	Ca	Cd	Co	Cr	Cu	Fe	K	Mg	Mn	Na	Ni	P	Pb	S	Sb	Sr	V	Zn
April (0-30 cm)																							
M	66429	13.2	40.5	227	1.87	7933	2.14	24.7	138.7	45.1	41517	10691	8746	1181	685	111.5	317	16.8	58.7	2.8	38.5	61.4	84.1
Min	56451	10.6	30.8	187	1.67	5013	2.00	19.7	89.3	36.1	37535	9273	7158	885	624	61.2	243	13.2	17.5	2.4	31.3	54.4	68.7
Max	75969	15.2	59.9	251	2.11	47888	3.10	33.0	264.8	117.8	58350	11991	11670	1625	875	188.4	480	27.1	92.0	4.5	87.6	71.0	179.1
April (30-60 cm)																							
M	65071	17.2	24.1	294	1.73	7293	0.28	23.5	164.9	47.2	43082	9198	9207	1041	595	141.5	328	36.0	122.1	5.7	93.5	115.2	82.3
Min	52445	11.9	13.7	202	1.33	5193	0.20	18.9	95.0	37.2	39115	8078	7277	750	496	82.8	190	23.9	60.4	4.1	60.8	96.3	73.2
Max	75762	19.1	61.3	335	1.99	83337	0.37	28.9	223.8	111.6	48020	10284	12042	1470	670	223.7	539	49.7	539.2	6.7	133.3	124.1	85.7
May (0-30 cm)																							
M	67933	13.8	41.7	237	1.91	7654	2.21	26.2	132.9	44.4	42023	10351	8724	1090	675	110.0	380	20.1	96.8	3.1	40.7	63.9	85.8
Min	54581	9.9	30.1	189	1.60	4764	1.94	19.4	118.5	36.7	38752	8563	7176	899	536	61.1	258	13.2	73.0	2.2	22.7	53.7	76.2
Max	137629	28.3	77.0	515	4.15	43391	4.43	50.6	284.1	123.8	85368	20743	16245	2619	1522	212.7	813	36.3	266.3	6.2	85.5	131.9	182.4
May (30-60 cm)																							
M	66044	16.5	21.0	306	1.80	6835	0.27	24.0	160.0	48.8	43194	9254	8816	1106	691	132.4	361	35.8	108.9	5.1	62.5	109.8	79.9
Min	44414	11.6	15.8	160	1.13	4420	0.18	15.9	98.8	39.5	34007	6654	6942	808	430	69.0	244	19.6	75.4	4.5	86.8	83.8	65.8
Max	75431	20.5	77.0	348	2.19	118262	0.45	28.9	523.0	87.1	47903	10929	11154	1597	866	277.2	494	45.0	751.0	8.3	43.6	126.0	90.7
June (0-30 cm)																							
M	66812	14.3	37.8	250	1.95	7492	2.18	25.2	140.5	41.7	42516	10393	8432	1170	687	106.1	332	18.6	104.4	3.0	43.7	62.8	79.9
Min	57661	13.4	34.2	196	1.73	5437	2.14	22.4	123.3	39.2	40651	9723	7702	871	592	63.1	306	13.1	60.4	2.7	36.2	57.1	74.0
Max	73511	16.6	50.3	265	2.10	51011	2.35	29.3	180.8	72.6	45122	11797	9644	1477	773	188.1	486	29.8	126.7	3.8	86.6	67.6	86.0
June (30-60 cm)																							
M	61786	16.8	17.8	291	1.80	6931	0.34	22.3	144.6	43.6	42292	8434	7872	1163	724	128.7	361	35.7	124.2	5.0	87.2	111.1	79.7
Min	30793	10.0	14.5	124	0.87	5003	0.24	15.6	94.4	28.2	29682	5542	6962	776	592	88.3	235	10.8	80.2	3.8	70.1	49.4	45.5
Max	70495	20.5	31.6	328	1.98	189642	0.39	23.9	172.6	99.6	46426	10863	10380	1503	802	169.2	515	53.5	1060	6.7	130.3	119.7	104.8
July (0-30 cm)																							
M	67305	13.4	37.2	241	1.91	7098	2.18	25.3	124.4	43.9	42813	12104	8925	1104	675	106.5	362	17.7	102.4	3.1	40.8	63.2	81.0
Min	55298	9.8	29.8	188	1.62	5093	1.92	20.2	96.6	37.1	39704	10255	7174	875	492	61.6	232	10.7	59.2	2.4	27.8	53.6	68.7
Max	78817	16.5	64.5	370	2.10	65679	2.42	31.4	200.5	138.6	48140	9263	12553	3261	844	197.8	487	25.7	124.2	3.9	94.5	69.7	92.4
July (30-60 cm)																							
M	70186	15.6	38.0	303	2.63	7127	1.19	22.9	149.3	40.7	44353	9629	9022	991	802	129.6	324	27.1	53.8	1.2	52.8	61.9	87.6
Min	44538	10.4	17.9	188	1.84	5278	0.26	16.5	39.9	32.1	37807	7508	7501	701	573	69.4	22	18.1	10.7	0.3	31.7	38.5	63.4
Max	81124	19.0	52.9	345	3.10	113533	2.07	177.1	219.4	112.0	46864	11142	13023	1488	1005	373.6	464	59.9	141.1	38.2	173.0	180.6	98.2
August (0-30 cm)																							
M	71509	13.7	41.6	262	2.00	8298	3.91	25.9	137.8	43.2	42299	11402	9006	1084	756	106.2	336	16.7	91.2	3.2	42.6	66.8	82.2
Min	60784	8.6	33.7	215	1.70	5342	3.56	21.3	93.9	38.1	40138	10304	7731	904	701	62.0	241	13.0	52.5	2.6	28.5	57.4	71.5
Max	81568	17.6	59.8	284	2.41	48874	4.40	33.6	221.0	82.9	47782	13109	12152	1681	866	199.3	553	27.7	145.7	4.2	93.2	71.7	101.1
August (30-60 cm)																							
M	68017	15.5	39.8	298	2.18	7281	1.25	25.6	145.3	38.2	43092	9421	8833	1106	766	130.3	333	23.8	71.4	4.8	48.2	96.6	82.9
Min	8536	1.2	11.0	129	0.16	4764	0.44	17.4	34.4	26.9	10236	1634	6390	767	395	32.6	47	11.5	21.9	1.6	36.4	0.9	55.8
Max	81384	19.0	96.3	424	2.84	96473	1.39	31.2	256.8	90.2	52079	12646	12629	1755	1000	298.1	536	54.4	142.1	6.9	147.7	107.7	131.2
Local back ground (C)																							
Mean	69453	14.47	27.4	315	1.97	5796	0.24	19	70	46.6	42692	10040	8538	1013	685	56.9	347.3	30.6	87.3	4.6	46.5	105	83.1
SD	3806	1.43	9.0	30	0.12	211	0.03	4.0	19	6.4	878	1336	307	95	92	2.3	1.3	2.6	1.1	1	3.1	13	2
RSD %	5.5	9.9	32.0	9.4	5.9	3.6	9.4	20.5	27.0	14.0	2.1	13.3	3.6	9.4	13.5	3.9	0.4	8.4	1.2	21.7	6.5	12.1	2.4
MAC		25	50				3		100	100						50		100					

Table S3: The descriptive statistics of element concentrations (mg kg^{-1}) (Median-M, Minimum-Min, and Maximum-Max) extracted from the topsoil and the subsoil samples by deionised H_2O during 2 h extraction

	Al	As	B	Ba	Be	Ca	Cd	Co	Cr	Cu	Fe	K	Li	Mg	Mn	Na	Ni	P	Pb	S	Sb	Sr	V	Zn
April 0-30 cm																								
M	9.0	<DL	0.51	0.66	0.0037	137	0.0007	0.0068	0.02	0.30	2.67	5.99	0.007	15.0	0.35	11.7	0.042	1.04	0.034	10.0	<DL	0.249	0.009	0.68
Min	2.5	/	0.08	0.26	0.0008	97	0.0004	0.0001	0.005	0.03	0.57	3.12	0.004	3.5	0.12	7.9	0.012	0.57	0.010	2.7	/	0.119	0.007	0.07
Max	24.9	/	4.75	2.14	0.0101	412	0.0015	0.0242	0.07	1.48	7.64	11.44	0.015	64.6	1.75	46.3	0.194	2.21	0.086	20.2	/	4.689	0.025	2.49
April 30-60 cm																								
M	10.7	<DL	0.59	0.93	0.0037	162	0.0008	0.0050	0.01	0.64	1.53	3.91	0.006	12.1	0.45	17.1	0.038	0.70	0.055	10.7	<DL	0.286	0.010	0.44
Min	3.5	/	0.11	0.28	0.0018	68	0.0004	0.0006	0.004	0.22	0.52	2.18	0.004	3.7	0.10	8.6	0.005	0.37	0.014	4.0	/	0.137	0.007	0.12
Max	41.0	/	14.17	2.45	0.0117	380	0.0014	0.0337	0.03	8.30	10.76	7.45	0.018	39.2	1.23	54.3	0.224	1.65	0.355	18.0	/	2.653	0.075	1.86
Maj 0-30 cm																								
M	8.6	<DL	0.11	0.51	0.0034	127	0.0009	0.0095	0.01	0.70	2.03	4.94	0.034	11.3	0.34	6.0	0.085	1.24	0.028	25.2	<DL	0.113	0.017	1.20
Min	0.4	/	0.10	0.05	0.0032	42	0.0008	0.0088	0.003	0.08	0.25	2.11	0.013	4.1	0.04	3.0	0.035	0.04	0.017	3.4	/	0.055	0.016	0.08
Max	37.6	/	14.47	1.58	0.0065	226	0.0017	0.0337	0.30	1.38	13.94	14.54	0.079	25.3	1.80	17.9	0.230	5.54	0.086	38.0	/	0.456	0.035	4.81
May 30-60 cm																								
M	8.9	<DL	0.19	0.68	0.0035	45	0.0009	0.0095	0.01	0.78	1.86	2.06	0.028	11.6	0.39	6.3	0.085	0.63	0.035	27.9	<DL	0.096	0.018	1.16
Min	1.2	/	0.10	0.04	0.0032	200	0.0008	0.0086	0.00	0.12	0.54	0.87	0.013	3.4	0.04	3.7	0.015	0.04	0.017	4.8	/	0.058	0.016	0.07
Max	71.9	/	29.02	1.46	0.0035	225	0.0009	0.0236	0.41	1.13	32.85	6.58	0.071	19.3	1.33	35.9	0.197	2.89	0.092	40.3	/	0.318	0.036	5.30
June 0-30 cm.																								
M	27.8	<DL	0.01	0.49	0.0024	82	0.0100	0.0092	0.05	0.67	11.27	4.88	0.026	11.4	0.36	5.7	0.110	0.91	0.029	10.2	<DL	0.083	0.036	0.45
Min	14.3	/	0.00	0.09	0.0024	52	0.0044	0.0007	0.02	0.33	3.53	3.00	0.014	4.3	0.19	3.6	0.047	0.63	0.007	3.3	/	0.053	0.016	0.003
Max	52.7	/	0.24	1.09	0.0024	178	0.0235	0.0149	0.12	0.93	25.05	18.37	0.045	19.4	1.18	13.1	0.210	1.80	0.051	18.9	/	0.175	0.063	1.02
June 30-60 cm																								
M	32.8	<DL	0.00	0.62	0.0024	83	0.0092	0.0076	0.07	0.75	12.55	5.51	0.026	11.1	0.55	7.6	0.148	0.62	0.039	12.3	<DL	0.078	0.035	0.94
Min	13.0	/	0.00	0.15	0.0023	40	0.0036	0.0016	0.02	0.30	3.77	0.03	0.017	4.7	0.22	4.6	0.050	0.40	0.017	5.6	/	0.038	0.016	0.16
Max	93.5	/	0.18	1.35	0.0024	589	0.0195	0.0175	0.18	4.79	42.32	12.88	0.111	22.8	1.20	12.2	0.451	1.61	0.105	21.8	/	1.007	0.098	4.64
July 0-30 cm																								
M	10.0	<DL	0.41	0.54	0.0023	78	0.0096	0.0064	0.02	0.53	3.49	6.32	0.021	10.3	0.42	4.8	0.100	1.03	0.041	9.7	<DL	0.074	0.022	0.13
Min	4.8	/	0.10	0.11	0.0003	35	0.0022	0.0005	0.01	0.36	1.52	2.25	0.008	4.0	0.06	2.0	0.032	0.45	0.010	5.4	/	0.031	0.006	0.003
Max	44.9	/	1.92	1.07	0.0024	150	0.0165	0.0142	0.12	1.10	20.26	9.82	0.032	15.5	1.05	18.6	0.231	2.15	0.307	17.2	/	0.114	0.059	2.23
July 30-60 cm																								
M	18.1	<DL	0.41	1.40	0.0024	58	0.0059	0.0059	0.04	0.78	6.98	3.61	0.023	8.1	0.19	7.3	0.084	0.38	0.023	14.2	<DL	0.063	0.026	0.24
Min	6.0	/	0.14	0.27	0.0023	15	0.0011	0.0011	0.01	0.34	2.31	0.69	0.007	3.4	0.12	2.4	0.012	0.05	0.001	8.7	/	0.016	0.004	0.003
Max	46.7	/	0.98	3.65	0.0024	196	0.0126	0.0126	0.11	1.34	20.52	5.95	0.044	14.5	0.82	47.6	0.840	1.08	0.175	27.1	/	0.113	0.056	3.89
August 0-30 cm																								
M	11.1	<DL	0.01	0.62	0.0038	139	0.0011	0.0065	0.02	0.63	3.17	5.44	0.003	11.9	0.30	6.9	0.053	1.10	0.042	9.3	<DL	0.251	0.016	0.37
Min	1.2	/	0.00	0.29	0.0017	44	0.0007	0.0011	0.003	0.11	0.53	1.38	0.0004	5.6	0.12	4.1	0.014	0.32	0.004	4.1	/	0.118	0.007	0.00
Max	110.8	/	1.04	2.49	0.0110	256	0.0098	0.0217	0.33	1.23	72.76	15.04	0.053	31.9	0.71	21.9	0.429	2.98	1.355	15.0	/	0.517	0.134	2.12
August 30-60 cm																								
M	19.4	<DL	0.01	0.96	0.0053	132	0.0015	0.0098	0.03	0.62	4.92	3.68	0.005	13.4	0.32	8.5	0.068	0.62	0.075	10.0	<DL	0.266	0.018	0.86
Min	5.2	/	0.00	0.26	0.0017	49	0.0007	0.0023	0.02	0.32	1.33	1.30	0.0002	4.1	0.18	4.4	0.027	0.19	0.004	5.8	/	0.087	0.006	0.23
Max	65.3	/	20.73	3.24	0.0152	248	0.0080	0.0239	0.22	3.83	39.01	9.25	0.032	24.9	1.19	69.3	1.293	1.91	0.544	17.7	/	0.391	0.079	6.84

Table S4: The descriptive statistics of element concentrations (mg kg^{-1}) (Median-M, Minimum-Min, and Maximum-Max) extracted from the topsoil and the subsoil samples by deionised H_2O during 16 h extraction

	Al	As	B	Ba	Be	Ca	Cd	Co	Cr	Cu	Fe	K	Li	Mg	Mn	Na	Ni	P	Pb	S	Sb	Sr	V	Zn
April0-30 cm																								
M	8.5	<DL	0.43	0.79	0.0041	193	0.002	0.005	0.009	0.44	2.11	7.1	0.006	18.7	0.67	31.7	0.06	0.82	0.024	11.3	<DL	0.310	0.014	0.26
Min	1.8	/	0.18	0.16	0.00027	112	0.001	0.003	0.004	0.19	0.53	4.3	0.003	5.5	0.09	19.6	0.01	0.37	0.002	6.7	/	0.212	0.001	0.07
Max	60.4	/	6.48	3.09	0.01218	288	0.005	0.015	0.184	2.42	48.89	15.5	0.050	35.1	3.46	90.2	0.24	2.57	0.093	17.8	/	0.484	0.114	0.92
April 30-60 cm																								
M	12.6	<DL	0.53	1.04	0.00531	189	0.002	0.003	0.012	0.48	2.35	4.8	0.006	15.1	0.80	32.4	0.07	0.59	0.020	12.0	<DL	0.287	0.017	0.30
Min	3.0	/	0.18	0.25	0.00073	81	0.001	0.003	0.002	0.22	0.73	2.9	0.003	5.7	0.13	25.3	0.01	0.16	0.010	3.8	/	0.192	0.001	0.00
Max	60.7	/	12.28	3.53	0.01601	241	0.004	0.016	0.144	5.03	38.71	14.6	0.036	31.7	2.23	164.3	0.22	1.27	0.108	23.1	/	0.438	0.079	0.78
Maj 0-30 cm																								
M	8.7	<DL	0.39	0.50	0.00339	150	0.001	0.011	0.015	0.35	1.40	5.6	0.037	14.8	0.34	5.6	0.07	0.87	0.048	3.5	<DL	0.190	0.017	0.63
Min	1.0	/	0.10	0.04	0.00328	23	0.001	0.006	0.003	0.01	0.09	0.8	0.002	2.3	0.01	2.2	0.02	0.06	0.018	0.1	/	0.002	0.015	0.10
Max	34.3	/	17.10	3.23	0.00724	252	0.002	0.270	0.358	1.23	13.68	21.1	0.099	38.8	2.25	14.3	0.28	3.68	0.096	25.1	/	0.429	0.036	1.61
May 30-60 cm																								
M	10.1	<DL	0.42	0.71	0.0034	144	0.001	0.011	0.020	0.29	2.72	2.3	0.039	15.5	0.38	6.2	0.07	0.30	0.028	2.9	<DL	0.155	0.017	0.58
Min	1.1	/	0.10	0.02	0.00325	20	0.001	0.009	0.003	0.00	0.17	0.5	0.009	2.5	0.01	1.9	0.00	0.02	0.015	0.2	/	0.008	0.016	0.07
Max	65.9	/	7.65	4.35	0.00844	219	0.003	0.064	0.474	1.22	9.37	13.9	0.053	37.9	3.23	35.9	0.27	3.09	0.148	24.7	/	0.438	0.047	1.83
June 0-30 cm.																								
M	13.1	<DL	0.08	0.76	0.00237	56	0.050	0.400	0.024	0.75	5.25	4.6	0.013	14.7	0.53	3.4	0.22	0.42	0.033	14.1	<DL	0.112	0.029	0.89
Min	8.0	/	0.02	0.05	0.00158	3.8	0.004	0.332	0.005	0.61	1.27	3.0	0.008	6.1	0.06	1.9	0.02	0.14	0.014	9.8	/	0.083	0.015	0.51
Max	34.7	/	0.64	1.36	0.0024	107	0.051	0.711	0.033	1.37	6.75	10.3	0.024	20.3	1.01	8.7	0.57	0.94	0.110	21.1	/	0.156	0.049	1.56
June 30-60 cm																								
M	14.7	<DL	0.14	0.62	0.00236	36	0.050	0.518	0.021	0.99	4.53	3.3	0.015	11.2	0.43	3.9	0.19	0.28	0.054	13.6	<DL	0.102	0.030	0.89
Min	5.0	/	0.01	0.08	0.00169	2.6	0.004	0.255	0.005	0.51	0.98	2.2	0.001	5.3	0.11	2.1	0.01	0.10	0.020	7.9	/	0.058	0.009	0.00
Max	55.0	/	0.34	1.55	0.00241	311	0.051	4.235	0.064	8.42	15.56	5.5	0.054	22.0	1.27	7.5	0.86	0.67	0.148	18.1	/	0.502	0.051	3.07
July 0-30 cm																								
M	21.5	<DL	0.40	0.81	0.00699	85	0.011	0.462	0.033	0.91	7.35	6.4	0.026	16.8	0.59	3.1	0.21	0.85	0.097	14.6	<DL	0,185	0.023	0.54
Min	5.6	/	0.19	0.11	0.0023	10	0.003	0.330	0.017	0.65	3.65	2.3	0.012	6.9	0.04	0.6	0.01	0.36	0.042	9.8	/	0,084	0.006	0.08
Max	54.7	/	0.65	1.85	0.01339	235	0.019	1.918	0.063	2.76	15.03	10.1	0.043	24.5	1.63	18.4	0.80	2.11	0.234	24.4	/	0,481	0.049	1.99
July 30-60 cm																								
M	22.8	<DL	0.28	0.93	0.00918	88	0.008	0.425	0.036	0.84	8.03	4.3	0.026	18.0	0.40	3.6	0.13	0.44	0.096	12.0	<DL	0.181	0.030	0.64
Min	8.1	/	0.07	0.09	0.00384	28	0.003	0.232	0.018	0.46	4.02	2.0	0.014	6.7	0.07	0.6	0.02	0.14	0.020	7.5	/	0.090	0.012	0.03
Max	61.5	/	1.01	3.00	0.01445	269	0.030	9.276	0.070	18.54	15.59	8.5	0.055	34.0	2.24	60.6	0.73	2.06	1.631	19.5	/	0.391	0.060	2.52
August 0-30 cm																								
M	10.3	<DL	0.01	0.27	0.00027	42	0.001	0.027	0.038	0.44	3.78	3.4	0.009	19.3	0.26	2.8	0.10	0.40	0.065	9.0	<DL	0.138	0.004	0.16
Min	1.2	/	0.01	0.08	0.00026	7	0.001	0.003	0.013	0.06	1.33	0.5	0.001	6.5	0.03	1.9	0.01	0.02	0.012	3.4	/	0.051	0.004	0.00
Max	46.1	/	3.61	1.80	0.0078	196	0.003	0.074	0.272	1.13	43.48	9.5	0.057	43.7	0.71	35.6	5.14	1.46	0.189	13.0	/	0.381	0.043	1.27
August 30-60 cm																								
M	12.6	<DL	0.06	0.45	0.00027	36	0.001	0.026	0.054	0.48	8.18	2.0	0.011	12.4	0.22	3.1	0.09	0.18	0.067	7.6	<DL	0.108	0.004	0.13
Min	1.0	/	0.00	0.07	0.00026	6	0.000	0.003	0.004	0.14	0.78	0.2	0.001	5.6	0.06	2.0	0.01	0.01	0.001	1.6	/	0.043	0.004	0.00
Max	49.4	/	10.36	2.76	0.00261	61	0.005	0.070	0.224	3.05	38.02	12.9	0.054	44.4	1.30	41.2	1.03	0.89	0.183	16.2	/	0.263	0.044	1.83

Table S5: The descriptive statistics of element concentrations (mg kg^{-1}) (Median-M, Minimum-Min, and Maximum-Max) extracted from the topsoil and the subsoil samples by 0.01 $CaCl_2$ during 3 h extraction

	Al	As	B	Ba	Be	Cd	Co	Cr	Cu	Fe	K	Mg	Mn	Na	Ni	P	Pb	S	Sb	Sr	V	Zn
April0-30 cm																						
M	1.61	<DL	0.21	1.95	0.0008	0.0020	0.0050	0.0051	0.176	1.50	32.3	224	1.72	9.5	0.043	0.36	<DL	1.61	<DL	2.96	0.008	0.138
Min	1.07	/	0.01	0.84	0.0002	0.0011	0.0011	0.0036	0.034	1.11	17.8	56	0.19	6.2	0.003	0.14	/	1.07	/	2.14	0.004	0.002
Max	3.79	/	6.38	4.75	0.0024	0.0112	0.1026	0.0138	1.093	2.57	47.0	365	27.32	53.7	0.751	0.59	/	3.79	/	3.95	0.014	0.466
April 30-60 cm																						
M	1.46	<DL	0.82	2.22	0.0007	0.0022	0.0056	0.0071	0.105	1.67	17.5	256	2.24	14.1	0.072	0.24	<DL	1.46	<DL	3.23	0.006	0.070
Min	0.93	/	0.08	0.77	0.0002	0.0011	0.0009	0.0009	0.006	0.91	10.2	44	0.09	9.6	0.003	0.09	/	0.93	/	1.73	0.003	0.002
Max	4.01	/	18.05	4.30	0.0022	0.0127	0.0782	0.0677	0.501	14.90	45.1	356	29.62	95.6	0.835	0.66	/	4.01	/	3.96	0.015	0.759
Maj 0-30 cm																						
M	4.27	<DL	0.31	1.15	0.0010	0.0009	0.0050	0.0094	0.022	2.50	26.1	156	2.51	7.1	0.059	0.27	<DL	4.55	<DL	1.99	0.005	0.192
Min	0.62	/	0.02	0.55	0.0000	0.0005	0.0008	0.0014	0.006	0.30	11.3	47	0.03	2.1	0.000	0.10	/	1.75	/	0.82	0.000	0.001
Max	5.99	/	16.06	3.53	0.0030	0.0075	0.1442	0.4113	0.261	3.61	65.3	405	26.17	27.9	11.691	1.95	/	12.32	/	3.75	0.023	16.883
May 30-60 cm																						
M	3.32	<DL	0.43	1.14	0.0010	0.0009	0.0054	0.0085	0.008	1.91	11.5	164	3.29	10.1	0.114	0.12	<DL	4.42	<DL	1.85	0.005	0.012
Min	1.83	/	0.01	0.48	0.0001	0.0006	0.0001	0.0023	0.006	0.86	3.5	38	0.00	2.8	0.004	0.01	/	1.26	/	0.81	0.000	0.001
Max	6.89	/	30.95	2.91	0.0028	0.0133	0.1100	0.4076	0.686	5.47	28.4	254	21.75	88.9	2.383	0.67	/	7.86	/	2.57	0.012	4.157
June 0-30 cm.																						
M	8.29	<DL	0.63	1.94	0.0008	0.0011	0.0064	0.0210	0.042	4.11	27.3	222	2.49	14.9	0.104	0.44	<DL	4.07	<DL	0.00	0.010	0.119
Min	4.28	/	0.01	0.84	0.0001	0.0009	0.0005	0.0048	0.006	2.17	11.9	71	0.10	8.9	0.011	0.14	/	1.62	/	0.00	0.002	0.002
Max	10.86	/	2.37	4.30	0.0040	0.0125	0.1112	0.0542	13.682	5.23	52.6	373	26.41	22.7	0.864	0.98	/	8.91	/	0.00	0.022	0.960
June 30-60 cm																						
M	8.32	<DL	0.82	1.62	0.0008	0.0010	0.0063	0.0174	0.006	4.10	14.6	223	1.37	18.6	0.058	0.39	<DL	2.64	<DL	0.00	0.009	0.099
Min	2.30	/	0.08	0.65	0.0003	0.0009	0.0017	0.0042	0.006	1.44	7.9	42	0.18	6.9	0.017	0.12	/	0.18	/	0.00	-0.002	0.002
Max	11.27	/	1.19	5.11	0.0034	0.0180	0.1120	0.0312	3.404	5.92	30.3	385	45.62	35.4	1.148	0.42	/	4.82	/	0.16	0.016	0.279
July 0-30 cm																						
M	8.11	<DL	0.01	2.12	0.0008	0.0010	0.0062	0.0147	0.183	3.16	30.6	235	3.75	18.3	0.186	0.56	<DL	3.61	<DL	0.18	0.008	0.674
Min	5.88	/	0.01	0.81	0.0000	0.0003	0.0023	0.0079	0.006	2.33	14.7	55	0.32	-0.1	0.008	0.95	/	2.38	/	0.00	0.002	0.002
Max	11.98	/	11.22	4.90	0.0042	0.0285	0.0991	0.0311	770.117	5.14	45.8	352	34.26	64.8	0.895	2.87	/	7.08	/	0.99	0.015	1.996
July 30-60 cm																						
M	9.23	<DL	0.01	2.61	0.0008	0.0009	0.0058	0.0188	0.154	3.50	19.5	269	1.92	26.9	0.124	0.37	<DL	3.71	<DL	0.04	0.008	0.548
Min	5.39	/	0.01	0.67	0.0001	0.0002	0.0003	0.0034	0.006	2.74	10.2	55	0.24	3.3	0.005	0.18	/	1.68	/	0.00	0.000	0.120
Max	12.52	/	0.62	5.05	0.0047	0.0141	0.1162	0.0484	6.760	48.14	33.1	368	24.08	142.9	1.170	0.87	/	7.86	/	1.51	0.016	1.472
August 0-30 cm																						
M	1.73	<DL	0.01	1.80	0.0010	0.0031	0.0022	0.0059	0.130	1.58	29.4	0	194.32	0.6	9.322	0.04	0.271494	1.73	<DL	2.93	0.006	0.002
Min	0.80	/	0.01	0.77	0.0002	0.0011	0.0009	0.0007	0.005	0.74	9.1	0	59.36	0.0	3.922	0.00	0.051933	0.80	/	1.25	0.003	0.002
Max	3.19	/	4.68	4.26	0.0025	0.0123	0.0335	0.0110	3.042	2.46	46.9	0	336.86	12.8	36.391	0.96	0.79131	3.19	/	3.89	0.013	0.150
August 30-60 cm																						
M	1.88	<DL	0.01	1.72	0.0007	0.0028	0.0030	0.0053	0.051	1.55	17.5	0	234.36	0.3	13.405	0.03	0.138636	1.88	<DL	2.98	0.006	0.002
Min	0.98	/	0.01	0.77	0.0002	0.0011	0.0009	0.0007	0.001	0.86	6.8	0	44.22	0.0	5.633	0.00	0.06977	0.98	/	1.43	0.003	0.002
Max	4.72	/	24.68	5.49	0.0078	0.0205	0.1136	0.0856	0.295	4.08	46.6	0	356.84	26.1	41.01	1.61	0.438309	4.72	/	4.11	0.011	0.643

Table S6: The descriptive statistics of element concentrations (mg kg^{-1}) (Median-M, Minimum-Min, and Maximum-Max) extracted from the topsoil and the subsoil samples by 0.1 mol L^{-1} NH_4NO_3 during 2 h extraction

	Al	As	B	Ba	Be	Ca	Cd	Co	Cr	Cu	Fe	K	Mg	Mn	Na	Ni	P	Pb	S	Sb	Sr	V	Zn
April0-30 cm																							
M	2.08	<DL	0.0031	36.0	0.0008	3234	<DL	0.0023	0.006	0.027	1.85	134	310	3.15	9.2	0.063	0.096	<DL	0.921	<DL	6.79	0.006	0.026
Min	0.79	/	0.0030	23.9	0.0002	2229	/	0.0004	0.003	0.007	0.86	86	78	0.52	6.3	0.010	0.021	/	0.452	/	5.28	0.001	0.001
Max	6.47	/	1.1099	57.0	0.0157	3765	/	0.1342	0.017	0.274	4.94	174	498	41.37	47.8	1.293	0.147	/	2.401	/	8.80	0.017	0.538
April 30-60 cm																							
M	3.17	<DL	0.0222	36.9	0.0012	3207	<DL	0.0041	0.008	0.025	1.92	99	346	4.02	11.9	0.076	0.052	<DL	0.947	<DL	6.72	0.008	0.024
Min	0.47	/	0.0031	24.3	0.0002	2109	/	0.0009	0.002	0.002	0.79	71	63	0.19	8.3	0.007	0.021	/	0.368	/	4.96	0.001	0.001
Max	7.53	/	5.9736	58.8	0.0114	3920	/	0.1023	0.024	0.126	6.05	158	535	47.44	79.6	1.471	0.271	/	2.217	/	8.18	0.018	0.376
Maj 0-30 cm																							
M	4.26	<DL	0.8745	25.9	0.0020	1739	<DL	0.0098	0.016	0.037	1.88	18	77	2.96	9.0	0.117	0.047	0.0212	1.141	<DL	3.17	0.089	0.243
Min	2.74	0	0.0336	8.4	0.0001	1160	/	0.0021	0.008	0.003	0.53	4	16	0.02	5.0	0.018	0.044	0.0004	0.003	/	1.47	0.024	0.002
Max	8.03	0	11.0044	41.7	0.0021	2320	/	0.1971	0.218	46.951	5.94	36	141	25.20	30.6	2.346	0.225	0.0549	3.977	/	5.18	0.200	1.949
May 30-60 cm																							
M	4.16	<DL	0.7074	25.7	0.0020	1729	<DL	0.0097	0.019	0.038	2.04	32	68	2.28	7.7	0.164	0.046	0.0216	1.497	<DL	3.16	0.088	0.260
Min	2.75	0	0.0017	8.1	0.0019	1112	/	0.0003	0.009	0.007	0.75	12	19	0.09	4.6	0.017	0.004	0.0024	0.003	/	1.47	0.000	0.002
Max	9.59	0	4.7722	54.1	0.0039	2980	/	0.2765	0.214	0.829	6.75	64	155	30.60	21.0	1.741	0.949	0.0428	6.378	/	5.52	0.177	2.311
June 0-30 cm.																							
M	2.27	0.016566	0.0067	24.2	0.0198	28459	<DL	0.0067	0.007	0.005	0.83	82	146	1.82	15.2	0.110	0.043	0.0141	0.081	<DL	7.44	0.123	0.215
Min	1.89	0.016377	0.0066	12.2	0.0091	14955	/	0.0049	0.003	0.004	0.39	49	20	0.05	8.5	0.008	0.043	0.0115	0.080	/	4.81	0.063	0.001
Max	4.94	0.016734	0.0068	32.8	0.0239	35567	/	0.1022	0.017	1.098	3.62	133	222	20.59	23.0	0.929	0.044	0.0142	0.082	/	10.72	0.647	0.865
June 30-60 cm																							
M	1.84	0.016442	0.0067	23.2	0.0244	28512	<DL	0.0062	0.005	0.004	0.53	56	122	0.83	16.2	0.052	0.043	0.0140	0.080	<DL	6.30	0.097	0.048
Min	1.51	0.015961	0.0065	6.3	0.0148	18480	/	0.0048	0.003	0.001	0.19	18	8	0.08	9.2	0.005	0.042	0.0136	0.078	/	1.51	0.061	0.001
Max	3.04	0.01666	0.0068	30.1	0.0298	40857	/	0.1323	0.010	0.250	0.97	85	205	42.93	25.5	1.560	0.043	0.0142	0.081	/	9.45	1.179	0.959
July 0-30 cm																							
M	2.04	0.016388	0.0067	18.4	0.0221	27148	<DL	0.0053	0.005	0.026	0.59	92	101	2.22	14.1	0.124	0.043	0.0139	0.080	<DL	7.45	0.136	0.134
Min	1.57	0.016054	0.0065	13.1	0.0117	19271	/	0.0048	0.003	0.004	0.18	52	21	0.14	10.0	0.006	0.042	0.0136	0.078	/	3.85	0.062	0.001
Max	3.85	0.016606	0.0067	32.9	0.0287	37025	/	0.1534	0.015	0.476	1.37	129	232	39.27	20.9	1.294	0.043	0.0141	0.081	/	10.07	1.114	0.796
July 30-60 cm																							
Median	2.36	0.016462	0.0067	24.1	0.0191	27148	<DL	0.0050	0.006	0.004	0.83	71	141	1.17	18.2	0.117	0.043	0.0140	0.080	<DL	7.89	0.070	0.003
Min	1.42	0.015661	0.0064	9.1	0.0060	19271	/	0.0048	0.000	0.001	0.14	35	14	0.05	6.3	0.000	0.041	0.0133	0.076	/	2.30	0.062	0.001
Max	7.49	0.045142	0.0068	34.8	0.0317	37025	/	0.1799	0.020	0.143	2.76	92	271	26.77	25.5	2.060	0.043	0.1284	0.081	/	10.61	0.856	0.891
August 0-30 cm																							
M	1.96	<DL	0.0031	37.2	0.0012	2991	<DL	0.0018	0.005	0.041	1.56	126	281	1.08	9.1	0.051	0.065	<DL	1.693	<DL	5.96	0.006	0.052
Min	0.45	/	0.0030	20.0	0.0002	2029	/	0.0001	0.001	0.005	0.58	74	67	0.07	5.2	0.003	0.020	/	0.897	/	3.80	0.002	0.001
Max	6.06	/	0.4391	66.9	0.0078	3902	/	0.0408	0.013	4.151	3.66	181	558	21.21	30.9	1.713	0.293	/	5.440	/	7.82	0.010	0.434
August 30-60 cm																							
M	2.63	<DL	0.0031	40.7	0.0013	3064	<DL	0.0018	0.006	0.067	1.71	91	331	0.67	13.6	0.045	0.040	<DL	1.705	<DL	6.43	0.006	0.045
Min	0.35	/	0.0030	21.3	0.0002	1960	/	0.0001	0.001	0.002	0.72	68	57	0.05	5.4	0.004	0.020	/	0.477	/	4.37	0.003	0.001
Max	24.69	/	7.7141	56.5	0.0342	4513	/	0.1432	0.017	43.849	4.42	163	553	33.31	86.3	2.475	0.210	/	4.117	/	9.12	0.012	0.800

Table S7: The descriptive statistics of element concentrations (mg kg^{-1}) (Median-M, Minimum-Min, and Maximum-Max) extracted from the topsoil and the subsoil samples by 0.05 mol L^{-1} Na_2EDTA during 1 h extraction

	Al	As	B	Ba	Be	Ca	Cd	Co	Cr	Cu	Fe	K	Li	Mg	Mn	Na	Ni	P	Pb	S	Sb	Sr	V	Zn
April 0-30 cm																								
M	318	0.141	0.300	14.9	0.0038	3121	0.075	5.56	0.029	7.1	167	44	0.082	199	308	/	7.6	7.2	8.5	0.5	<DL	5.1	1.1	3.44
Min	76	0.040	0.017	4.4	0.0001	1932	0.044	2.16	0.005	4.7	68	26	0.053	78	205	/	3.9	2.4	4.5	0.4	/	3.6	0.5	0.95
Max	437	0.312	9.560	20.0	0.0231	16035	0.106	6.83	0.268	36.7	289	74	0.100	272	423	/	16.3	29.4	11.1	13.2	/	18.0	2.6	6.28
April 30-60 cm																								
M	313	0.121	0.125	15.6	0.0067	2834	0.057	5.56	0.020	6.1	155	23	0.078	201	291	/	6.5	1.5	7.4	0.5	<DL	4.8	1.0	1.81
Min	26	0.028	0.017	3.6	0.0019	1797	0.016	0.27	0.005	1.7	26	14	0.065	63	61	/	1.0	0.1	2.6	0.4	/	2.8	0.1	0.34
Max	469	0.309	25.726	19.4	0.0241	15950	0.135	8.70	0.213	33.2	466	62	0.114	267	504	/	21.8	22.6	15.2	15.1	/	16.4	3.8	4.11
Maj 0-30 cm																								
M	560	0.137	0.316	21.8	0.0043	3329	0.075	7.35	0.302	9.3	7	56	0.107	229	408	/	9.2	25.7	11.8	30.1	<DL	5.4	2.5	6.42
Min	222	0.082	0.031	8.9	0.0000	1880	0.024	5.51	0.117	4.6	6	27	0.052	93	252	/	4.1	7.3	7.5	4.2	/	4.0	1.5	0.79
Max	1149	0.347	17.099	51.3	0.0362	24613	0.150	18.17	10.979	50.0	18	119	0.201	531	909	/	22.9	135.3	20.0	48.6	/	28.8	4.9	11.37
May 30-60 cm																								
M	561	0.105	0.347	21.9	0.0097	3073	0.055	7.29	0.275	7.2	373	31	0.111	237	381	/	8.2	13.6	9.6	28.4	<DL	5.8	2.1	2.07
Min	71	0.085	0.004	7.0	0.0022	1963	0.025	2.90	0.060	2.9	74	7	0.048	84	246	/	5.1	0.2	4.9	5.7	/	3.7	0.3	0.26
Max	745	0.247	32.655	29.9	0.0287	28469	0.122	13.62	4.701	32.8	802	78	0.169	321	683	/	17.0	55.5	14.0	46.0	/	38.1	4.7	8.19
June 0-30 cm.																								
M	233	0.077	0.017	9.5	0.0018	2507	0.033	3.45	0.005	4.1	129	21	0.045	196	210	/	4.0	4.4	4.7	0.4	<DL	5.7	0.7	1.92
Min	57	0.041	0.016	3.9	0.0003	1451	0.002	1.33	0.003	0.0	48	9	0.019	43	134	/	2.6	1.3	2.9	0.4	/	0.0	0.3	0.004
Max	291	0.220	4.722	15.4	0.0160	17010	0.079	6.34	0.063	15.6	247	64	0.061	264	341	/	7.8	20.3	8.3	16.7	/	17.0	2.0	5.03
June 30-60 cm																								
M	182	0.040	0.017	8.1	0.0021	2209	0.005	3.38	0.009	0.0	98	6	0.033	87	170	/	3.7	1.0	2.7	0.4	<DL	3.9	0.5	0.004
Min	8.6	0.013	0.016	2.3	0.0001	517	0.0001	0.13	0.005	0.0	30	0	0.002	70	33	/	0.3	0.1	0.1	0.4	/	0.0	0.1	0.004
Max	477	0.086	1.818	10.6	0.0188	14421	0.050	5.69	0.059	14.1	301	20	0.055	209	351	/	6.0	8.9	7.2	12.6	/	16.9	1.8	5.25
July 0-30 cm																								
M	240	0.052	0.016	9.4	0.0018	2216	0.036	4.00	0.005	4.4	143	26	0.054	159	218	/	4.8	7.8	4.2	0.4	<DL	5.5	0.8	3.27
Min	0.53	0.006	0.016	0.001	0.0014	1.88	0.002	0.002	0.003	0.0	0	3	0.0001	1	0	/	0.0	0.1	0.03	0.4	/	0.01	0.0	0.004
Max	541	1.332	1.501	16.9	0.0179	13884	0.077	6.86	0.078	27.1	329	49	0.100	258	408	/	13.9	38.4	9.7	14.2	/	13.4	2.3	6.64
July 30-60 cm																								
M	275	0.051	0.016	8.6	0.0045	2543	0.020	3.74	0.005	2.4	131	16	0.070	181	178	/	4.1	2.1	4.2	0.4	<DL	5.3	0.7	2.23
Min	23	0.013	0.016	3.2	0.0013	1091	0.002	0.32	0.004	0.0	32	1	0.020	76	65	/	1.1	0.1	1.3	0.4	/	2.9	0.1	0.004
Max	457	0.196	0.017	18.3	0.0285	17412	0.089	5.25	0.020	14.3	239	30	0.088	311	392	/	10.0	15.2	6.5	21.5	/	16.7	1.6	6.04
August 0-30 cm																								
M	322	0.182	0.017	14.7	0.0025	3976	0.077	6.03	0.005	6.5	179	37	0.079	209	351	/	8.3	9.8	7.4	0.5	<DL	4.9	1.2	2.34
Min	62	0.003	0.016	4.4	0.0018	1926	0.038	1.58	0.005	4.1	63	10	0.060	68	207	/	3.2	2.6	3.8	0.4	/	3.1	0.5	0.89
Max	461	0.366	17.382	20.2	0.0286	19065	0.228	7.94	0.038	23.9	283	68	0.096	300	486	/	13.6	72.4	10.7	17.9	/	19.2	2.6	5.95
August 30-60 cm																								
M	306	0.160	0.017	15.1	0.0043	3216	0.058	5.73	0.005	5.6	170	19	0.076	206	292	/	6.6	3.2	6.5	0.5	<DL	4.4	1.0	1.17
Min	28	0.006	0.015	4.3	0.0009	2140	0.007	0.22	0.004	1.4	26	3	0.051	60	47	/	1.4	0.1	2.1	0.4	/	1.6	0.1	0.005
Max	507	0.313	34.067	21.2	0.0338	19240	0.144	8.80	0.750	394.1	335	59	0.101	331	538	/	13.6	33.6	11.3	11.3	/	21.3	2.5	7.77

Table S8: The descriptive statistics of element concentrations (mg kg^{-1}) (Median-M, Minimum-Min, and Maximum-Max) extracted from the topsoil and the subsoil samples by 0.11 mol L^{-1} CH_3COOH during 16 h extraction

	Al	As	B	Ba	Be	Ca	Cd	Co	Cr	Cu	Fe	K	Mg	Mn	Na	Ni	P	Pb	S	Sb	Sr	V	Zn
April0-30 cm																							
M	45.6	<DL	0.90	21.8	0.146	1310	0.013	0.300	0.094	0.010	13.1	90.6	339	49.6	11.3	6.9	4.8	0.035	0.27	<DL	5.8	0.014	0.744
Min	35.7	/	0.13	13.7	0.039	826	0.005	0.070	0.035	0.010	8.1	50.8	155	37.4	7.1	4.6	0.9	0.008	0.26	/	4.9	0.003	0.002
Max	64.1	/	12.19	25.9	0.199	3345	0.042	0.559	0.165	3.025	15.7	129.1	421	94.6	57.2	29.6	17.3	0.088	2.41	/	48.2	0.024	1.818
April 30-60 cm																							
M	42.1	<DL	1.26	22.7	0.160	1167	0.014	0.264	0.108	0.010	12.7	61.6	322	48.8	13.5	8.0	1.2	0.042	0.27	<DL	5.9	0.019	0.426
Min	17.3	/	0.15	12.2	0.003	702	0.002	0.027	0.053	0.010	9.7	46.0	128	18.5	7.1	4.8	0.2	0.035	0.27	/	3.9	0.001	0.002
Max	114.0	/	27.70	27.1	0.213	3579	0.050	0.592	0.274	3.495	16.9	105.1	424	120.5	109.0	55.3	3.7	0.158	0.56	/	81.0	0.043	1.349
Maj 0-30 cm																							
Mn	35.9	0.053	0.88	19.6	0.150	1094	0.015	0.506	0.076	0.011	9.3	90.2	316	62.3	6.2	2.8	7.5	0.043	11.32	<DL	5.5	0.020	0.928
Min	14.9	0.001	0.15	15.2	0.019	617	0.000	0.119	0.025	0.010	1.2	51.4	150	45.3	0.3	1.8	1.4	0.020	2.73	/	4.1	0.003	0.002
Max	82.6	0.164	22.18	40.5	0.337	6472	0.053	1.842	4.483	3.486	28.6	244.9	659	181.4	29.6	6.5	48.0	0.149	23.66	/	47.4	0.067	5.980
May 30-60 cm																							
M	26.7	0.038	1.02	20.0	0.160	1086	0.009	0.363	0.079	0.011	5.9	52.5	319	49.8	9.2	2.4	1.5	0.033	9.57	<DL	5.5	0.010	0.464
Min	6.3	0.008	0.10	12.6	0.014	662	0.001	0.022	0.009	0.010	0.4	23.4	141	31.4	4.0	1.5	0.1	0.002	0.74	/	3.8	0.000	0.002
Max	54.6	0.148	40.14	23.9	0.197	11451	0.079	0.838	2.106	3.355	17.9	149.3	468	352.3	109.0	4.5	11.3	0.162	22.14	/	87.4	0.068	4.918
June 0-30 cm.																							
M	41.6	0.045	0.03	18.1	0.013	28459	0.022	0.297	0.121	0.414	8.1	83.1	286	38.2	10.6	2.0	0.1	0.033	0.33	<DL	3.6	0.019	0.692
Min	33.7	0.011	0.03	12.0	0.002	14955	0.013	0.022	0.057	0.010	5.9	55.5	123	27.8	2.0	1.3	0.1	0.001	0.31	/	2.8	0.018	0.006
Max	50.4	0.047	0.03	20.3	0.022	35567	0.063	0.543	0.165	4.907	12.5	155.3	349	107.3	19.0	3.2	0.1	0.096	0.34	/	35.5	0.030	2.340
June 30-60 cm																							
M	39.9	0.045	0.03	18.3	0.013	28512	0.018	0.309	0.108	0.063	8.4	52.7	271	40.5	9.6	2.0	0.1	0.034	0.33	<DL	3.3	0.019	0.652
Min	12.1	0.011	0.03	5.0	0.001	18480	0.010	0.112	0.055	0.009	6.9	40.7	92	20.1	6.1	1.1	0.1	0.019	0.31	/	2.2	0.018	0.005
Max	58.6	0.046	0.48	21.3	0.037	40857	0.033	0.557	0.183	15.595	15.2	93.2	358	107.1	28.0	3.1	0.1	0.239	0.34	/	70.8	0.052	3.972
July 0-30 cm																							
M	48.4	0.051	0.03	17.9	0.013	27148	0.031	0.320	0.118	0.220	10.0	94.2	276	36.3	8.7	2.1	0.1	0.032	17.40	<DL	3.5	0.019	1.601
Min	24.8	0.005	0.03	8.9	0.000	19271	0.018	0.013	0.053	0.009	5.9	27.9	114	23.6	1.0	1.2	0.1	0.002	10.28	/	0.9	0.018	0.093
Max	59.3	0.102	0.03	22.2	0.046	37025	0.178	0.710	0.243	12.38	11.5	144.0	374	118.1	43.2	3.6	0.1	0.394	21.99	/	33.5	0.048	4.134
July 30-60 cm																							
M	43.8	0.044	0.03	18.8	0.015	27148	0.026	0.266	0.124	0.238	9.4	69.4	302	31.6	17.4	1.6	0.1	0.034	6.60	<DL	3.7	0.019	1.199
Min	13.8	0.0002	0.03	10.4	0.003	19271	0.006	0.026	0.059	0.010	4.4	53.5	96	14.0	4.1	1.1	0.1	0.001	1.96	/	2.4	0.018	0.006
Max	66.2	0.083	0.03	23.1	0.049	37025	0.063	0.621	0.209	4.620	12.6	84.8	413	124.6	154.8	2.8	0.1	0.261	13.34	/	59.0	0.045	4.007
August 0-30 cm																							
M	44.2	<DL	0.06	22.0	0.158	1394	0.011	0.283	0.077	0.010	11.5	93.3	323	37.2	15.9	9.1	4.6	0.036	14.76	<DL	5.6	0.024	0.845
Min	26.7	/	0.01	14.6	0.035	762	0.002	0.030	0.039	0.010	5.7	42.6	156	21.6	11.3	6.9	0.7	0.015	5.98	/	4.1	0.009	0.130
Max	53.5	/	9.17	26.3	0.208	6442	0.048	0.427	0.150	19.26	15.4	138.8	421	111.4	43.5	22.7	31.1	0.254	24.35	/	54.9	0.042	3.029
August 30-60 cm																							
M	38.3	<DL	0.10	21.7	0.169	1192	0.010	0.267	0.076	0.010	10.3	61.9	323	35.1	21.8	12.1	0.9	0.036	13.77	<DL	5.7	0.022	0.646
Min	15.2	/	0.05	12.3	0.002	717	0.001	0.019	0.035	0.003	4.6	32.5	121	17.1	14.1	7.7	0.0	0.034	0.27	/	3.8	0.002	0.002
Max	57.8	/	36.00	27.1	0.214	6659	0.045	0.497	0.878	6.885	16.3	113.5	463	175.3	125.6	63.4	10.9	0.108	21.07	/	76.1	0.046	3.820

Table S9: The descriptive statistics of contamination factor (CF) (-) calculated for PTEs and representing the comparison the element concentration in the vineyard according to the local background concentration (C)

	Al	As	B	Ba	Be	Cd	Co	Cr	Cu	Fe	Mn	Ni	P	Pb	S	Sb	Sr	V	Zn
April 0-30 cm																			
M	0.96	0.91	1.48	0.72	0.95	8.93	1.28	1.99	0.99	0.97	1.17	1.96	0.91	0.55	0.67	0.62	0.83	0.59	1.01
Min	0.81	0.73	1.13	0.59	0.85	8.35	1.02	1.28	0.79	0.88	0.87	1.08	0.70	0.43	0.20	0.52	0.67	0.52	0.83
Max	1.09	1.05	2.19	0.80	1.07	12.94	1.71	3.79	2.58	1.37	1.60	3.31	1.38	0.89	1.05	0.98	1.88	0.68	2.15
April 30-60 cm																			
M	0.94	1.19	0.88	0.93	0.88	1.17	1.22	2.36	1.04	1.01	1.03	2.49	0.94	1.18	1.40	1.25	2.01	1.10	0.99
Min	0.76	0.82	0.50	0.64	0.68	0.86	0.98	1.36	0.82	0.92	0.74	1.45	0.55	0.78	0.69	0.89	1.31	0.92	0.88
Max	1.09	1.32	2.24	1.06	1.01	1.53	1.50	3.21	2.45	1.12	1.45	3.93	1.55	1.62	6.18	1.47	2.87	1.18	1.03
May 0-30 cm																			
M	0.98	0.95	1.52	0.75	0.97	9.21	1.36	1.90	0.97	0.98	1.08	1.93	1.10	0.66	1.11	0.68	0.87	0.61	1.03
Min	0.79	0.68	1.10	0.60	0.81	8.09	1.01	1.70	0.80	0.91	0.89	1.07	0.74	0.43	0.84	0.48	0.49	0.51	0.92
Max	1.98	1.96	2.81	1.63	2.11	18.51	2.62	4.07	2.71	2.00	2.58	3.73	2.34	1.19	3.05	1.35	1.84	1.26	2.19
May 30-60 cm																			
M	0.95	1.14	0.77	0.97	0.92	1.12	1.25	2.29	1.07	1.01	1.09	2.33	1.04	1.17	1.25	1.11	1.87	1.05	0.96
Min	0.64	0.80	0.58	0.51	0.57	0.76	0.83	1.42	0.87	0.80	0.80	1.21	0.70	0.64	0.86	0.99	0.94	0.80	0.79
Max	1.09	1.42	2.82	1.10	1.11	1.89	1.50	7.49	1.91	1.12	1.58	4.87	1.42	1.47	8.60	1.80	3.09	1.20	1.09
June 0-30 cm																			
M	0.96	0.99	1.38	0.79	0.99	9.08	1.31	2.01	0.91	1.00	1.15	1.86	0.96	0.61	1.20	0.65	0.94	0.60	0.96
Min	0.83	0.93	1.25	0.62	0.88	8.92	1.16	1.77	0.86	0.95	0.86	1.11	0.88	0.43	0.69	0.58	0.78	0.55	0.89
Max	1.06	1.15	1.84	0.84	1.07	9.82	1.52	2.59	1.59	1.06	1.46	3.30	1.40	0.98	1.45	0.83	1.86	0.65	1.03
June 30-60 cm																			
M	0.89	1.16	0.65	0.92	0.92	1.41	1.16	2.07	0.96	0.99	1.15	2.26	1.04	1.17	1.42	1.10	1.88	1.06	0.96
Min	0.44	0.69	0.53	0.39	0.44	1.02	0.81	1.35	0.62	0.70	0.77	1.55	0.68	0.35	0.92	0.82	1.51	0.47	0.55
Max	1.02	1.42	1.15	1.04	1.01	1.61	1.24	2.47	2.18	1.09	1.48	2.97	1.48	1.75	12.14	1.47	2.80	1.14	1.26
July 0-30 cm																			
M	0.97	0.93	1.36	0.77	0.97	9.12	1.31	1.78	0.96	1.00	1.09	1.87	1.04	0.58	1.17	0.69	0.88	0.60	0.97
Min	0.80	0.68	1.09	0.60	0.82	8.03	1.05	1.38	0.81	0.93	0.86	1.08	0.67	0.35	0.68	0.53	0.60	0.51	0.83
Max	1.13	1.14	2.36	1.18	1.07	10.09	1.63	2.87	3.04	1.13	3.22	3.47	1.40	0.84	1.42	0.85	2.03	0.67	1.11
July 30-60 cm																			
M	1.01	1.08	1.39	0.96	1.34	4.96	1.19	2.14	0.89	1.04	0.98	2.28	0.93	0.89	0.62	0.26	1.14	0.59	1.05
Min	0.64	0.72	0.65	0.60	0.94	1.10	0.86	0.57	0.70	0.89	0.69	1.22	0.06	0.59	0.12	0.06	0.68	0.37	0.76
Max	1.17	1.31	1.93	1.10	1.57	8.64	9.19	3.14	2.46	1.10	1.47	6.56	1.34	1.96	1.62	8.32	3.72	1.72	1.18
August 0-30 cm																			
M	1.03	0.95	1.52	0.83	1.02	16.34	1.34	1.97	0.95	0.99	1.07	1.87	0.97	0.54	1.05	0.69	0.92	0.64	0.99
Min	0.88	0.60	1.23	0.68	0.86	14.85	1.10	1.35	0.84	0.94	0.89	1.09	0.69	0.42	0.60	0.57	0.61	0.55	0.86
Max	1.17	1.22	2.19	0.90	1.22	18.35	1.74	3.17	1.82	1.12	1.66	3.50	1.59	0.91	1.67	0.92	2.00	0.68	1.22
August 30-60 cm																			
M	0.98	1.07	1.46	0.95	1.11	5.21	1.33	2.08	0.84	1.01	1.09	2.29	0.96	0.78	0.82	1.04	1.04	0.92	1.00
Min	0.12	0.09	0.40	0.41	0.08	1.82	0.90	0.49	0.59	0.24	0.76	0.57	0.14	0.37	0.25	0.34	0.78	0.01	0.67
Max	1.17	1.31	3.52	1.34	1.45	5.78	1.62	3.68	1.98	1.22	1.73	5.23	1.54	1.78	1.63	1.50	3.18	1.03	1.58

Table S10: The mobility factor (MF%) (Median-M values) (-) representing the efficiency of the extractants for each of the elements

	Al	As	B	Ba	Be	Ca	Cd	Co	Cr	Cu	Fe	K	Mg	Mn	Na	Ni	P	Pb	S	Sb	Sr	V	Zn
											0-30 cm 2 h H_2O												
M	0.016	/	0.38	0.226	0.16	1.162	0.043	0.028	0.017	1.239	0.008	14.7	0.13	0.030	0.93	0.068	0.31	0.18	11.2	/	0.30	0.029	0.72
											30-60 cm 2 h H_2O												
M	0.022	/	0.65	0.304	0.19	1.140	0.333	0.033	0.020	1.537	0.011	0.037	0.11	0.030	1.11	0.066	0.19	0.11	15	/	0.175	0.017	0.73
											0-30 cm 16 h H_2O												
M	0.017	0.16	0.63	0.260	0.177	0.918	0.06	0.104	0.020	1.38	0.010	14.4	0.18	0.030	0.61	0.094	0.19	0.30	13	/	0.41	0.028	0.56
											30-60 cm 16 h H_2O												
M	0.022	0.13	0.83	0.261	0.186	0.724	0.415	0.107	0.021	1.335	0.013	0.036	0.16	0.031	0.81	0.070	0.11	0.19	12.08	/	0.23	0.017	0.47
											0-30 cm $CaCl_2$												
M	0.01	/	0.16	0.68	0.04	/	0.06	0.02	0.01	0.23	0.01	77	2.2	0.10	1.6	0.06	0.11	/	3.44	/	4.40	0.01	0.13
											30-60 cm $CaCl_2$												
M	0.0060	/	0.54	0.58	0.043	/	0.34383	0.02387	0.00712	0.14	0.0061	0.19	2.30	0.10	2.47	0.049	0.067	/	2.93	/	2.11	0.0067	0.094
											0-30 cm NH_4NO_3												
M	0.0036	/	0.02	11.8	0.11	39.8	/	0.0213	0.0049	0.078	0.0029	240	1.43	0.16	1.48	0.077	0.014	0.088	0.97	/	14.01	0.10	0.10
											30-60 cm NH_4NO_3												
M	0.0047	/	0.03	9.92	0.12	40.7	/	0.0304	0.0057	0.058	0.0032	0.7507	1.40	0.12	2.04	0.053	0.013	0.052	0.68	/	8.25	0.06	0.11
											0-30 cm Na_2EDTA												
M	0.47	0.86	0.05	5.78	0.18	42.2	2.37	21.94	0.00	14.6	0.32	94	2.11	26.7	/	6.67	3.08	40.0	0.58	/	13.33	1.96	3.75
											30-60 cm Na_2EDTA												
M	0.46	0.58	0.08	4.37	0.33	41.0	7.19	21.38	0.01	12.1	0.38	0.21	2.08	25.4	/	4.74	1.02	19.8	0.76	/	8.12	1.32	2.18
											0-30 cm CH_3COOH												
M	0.06	0.33	0.25	8.04	6.77	18.57	0.82	1.24	0.066	0.026	0.024	239	3.297	3.827	1.594	2.925	0.727	0.223	11.721	1.360	12.902	0.032	1.247
											30-60 cm CH_3COOH												
M	0.058	0.28	0.69	6.70	6.02	17.38	2.94	1.19	0.061	0.026	0.022	0.63	3.539	3.511	2.384	2.256	0.156	0.135	7.989	0.802	8.078	0.020	0.833

Table S11: The descriptive statistics of environmental risk assessment index for the elements (As, Cd, Cr, Cu, Ni, Pb, and Zn) (-) and ecological risk (RI) (-) calculated for the topsoil and the subsoil samples

Eri	As	Cd	Cr	Cu	Ni	Pb	Zn	RI=ΣEri
April 0-30 cm								
M	4.56	268	3.97	4.95	9.79	2.74	1.01	295
Min	3.65	250	2.56	3.96	5.38	2.16	0.83	269
Max	5.25	388	7.59	12.92	16.55	4.44	2.15	437
April 30-60 cm								
M	5.93	35	4.73	5.18	12.43	5.88	0.99	70
Min	4.10	26	2.72	4.08	7.27	3.91	0.88	49
Max	6.61	46	6.41	12.23	19.65	8.12	1.03	100
May 0-30 cm								
M	4.76	276	3.81	4.87	9.66	3.28	1.03	304
Min	3.42	243	3.40	4.02	5.36	2.15	0.92	262
Max	9.79	555	8.14	13.57	18.67	5.93	2.19	613
May 30-60 cm								
M	5.70	34	4.58	5.35	11.63	5.86	0.96	68
Min	4.01	23	2.83	4.34	6.06	3.21	0.79	44
Max	7.09	57	14.98	9.55	24.34	7.36	1.09	121
June 0-30 cm								
M	4.96	272	4.03	4.57	9.31	3.04	0.96	299
Min	4.63	268	3.53	4.30	5.54	2.15	0.89	289
Max	5.74	295	5.18	7.95	16.52	4.88	1.03	336
June 30-60 cm								
M	5.82	42	4.14	4.78	11.30	5.83	0.96	75
Min	3.47	31	2.70	3.09	7.75	1.77	0.55	50
Max	7.08	48	4.95	10.92	14.86	8.75	1.26	96
July 0-30 cm								
M	4.65	273	3.56	4.82	9.35	2.90	0.97	300
Min	3.38	241	2.77	4.07	5.41	1.75	0.83	259
Max	5.72	303	5.74	15.20	17.36	4.20	1.11	352
July 30-60 cm								
M	5.38	149	4.28	4.46	11.38	4.43	1.05	180
Min	3.58	33	1.14	3.51	6.10	2.95	0.76	51
Max	6.55	259	6.29	12.28	32.80	9.79	1.18	328
August 0-30 cm								
M	4.74	490	3.95	4.73	9.33	2.72	0.99	517
Min	2.98	445	2.69	4.18	5.45	2.12	0.86	464
Max	6.09	551	6.33	9.09	17.50	4.53	1.22	595
August 30-60 cm								
M	5.37	156	4.16	4.19	11.44	3.90	1.00	186
Min	0.43	55	0.99	2.95	2.86	1.87	0.67	64
Max	6.57	173	7.36	9.89	26.17	8.89	1.58	234

Table S12: The descriptive statistics of biogeochemical absorption coefficient (BGI) (-) representing the element absorptions in the topsoil indicating the anthropogenic element origin

BGI	Al	As	B	Ba	Be	Ca	Cd	Co	Cr	Cu	Fe	K	Mg	Mn	Na	Ni	P	Pb	S	Sb	Sr	V	Zn
April																							
M	1.01	0.81	1.74	0.79	1.09	0.94	7.81	1.04	0.88	0.93	0.97	0.00	0.95	1.02	1.19	0.83	1.05	0.49	0.41	0.51	0.46	0.56	1.03
Min	0.84	0.72	0.98	0.70	0.97	0.16	5.57	0.90	0.67	0.79	0.81	0.00	0.85	0.74	0.99	0.64	0.68	0.38	0.08	0.40	0.34	0.49	0.88
Max	1.15	0.89	2.25	1.01	1.25	1.35	11.06	1.57	1.47	1.37	1.35	0.00	1.06	1.59	1.41	0.93	1.58	0.67	1.31	0.80	0.66	0.62	2.17
May																							
M	1.03	0.86	1.77	0.82	1.09	1.09	8.05	1.08	0.82	0.96	0.98	0.00	1.00	1.02	0.98	0.86	1.11	0.58	0.90	0.59	0.48	0.58	1.10
Min	0.85	0.76	1.00	0.62	0.87	0.16	4.90	0.87	0.38	0.69	0.87	0.00	0.90	0.76	0.79	0.46	0.54	0.40	0.21	0.46	0.26	0.47	0.93
Max	2.35	1.82	4.44	1.77	2.46	2.73	16.96	2.27	2.25	2.57	2.27	0.01	2.30	2.24	2.31	2.03	2.19	1.03	2.00	1.36	1.05	1.28	2.58
June																							
M	1.06	0.86	2.01	0.84	1.11	1.02	6.47	1.14	0.92	1.00	0.99	0.00	1.02	1.08	0.96	0.82	1.00	0.56	0.82	0.55	0.52	0.58	1.01
Min	0.97	0.79	1.36	0.80	0.98	0.27	5.54	1.02	0.82	0.40	0.91	0.00	0.89	0.88	0.84	0.63	0.78	0.45	0.09	0.50	0.39	0.54	0.82
Max	1.87	1.41	3.24	1.58	1.98	2.01	8.98	1.77	1.92	1.96	1.52	0.01	1.25	1.48	1.11	1.14	1.33	1.22	1.41	1.01	0.72	1.16	1.78
July																							
M	0.98	0.86	1.04	0.80	0.70	1.01	1.83	1.07	0.85	1.07	0.97	0.00	0.98	1.10	0.85	0.84	1.10	0.59	1.80	2.82	0.83	1.02	0.96
Min	0.88	0.79	0.89	0.72	0.55	0.13	0.99	0.13	0.66	0.49	0.87	0.00	0.87	0.90	0.66	0.36	0.80	0.24	0.77	0.09	0.27	0.30	0.84
Max	1.24	1.00	1.98	1.23	1.08	2.18	7.71	1.38	3.55	1.39	1.12	0.01	1.12	3.34	1.17	1.11	11.28	0.86	9.56	10.11	1.11	1.73	1.09
August																							
M	1.03	0.88	1.02	0.86	0.91	1.06	3.16	1.05	0.85	1.11	0.97	1.17	0.99	0.97	0.99	0.80	1.08	0.73	1.31	0.64	0.89	0.68	0.98
Min	0.94	0.82	0.62	0.57	0.83	0.09	2.87	0.82	0.56	0.50	0.81	1.00	0.91	0.66	0.82	0.67	0.71	0.42	0.68	0.55	0.47	0.62	0.69
Max	1.22	1.10	1.13	0.94	1.09	1.38	3.83	1.30	1.21	1.50	1.12	1.43	1.16	1.34	1.29	1.13	1.50	1.26	3.31	0.77	1.05	0.75	1.36

Table S13: The descriptive statistics of bioavailability risk assessment (-) calculated for topsoil and subsoil samples using element concentrations extracted by Na_2EDTA (regular equation) and CH_3COOH (modify equation); The BRAIprobable (using concentrations of As, Cd, Cr, Cu, Ni, Pb and Zn) and BRAIapparent (using concentrations of As, Cd, Co, Cr, Cu, Mn, Ni, Pb, V and Zn) for both regular and modify equations are presented:

	BRAIEDTAprobbable	BRAIEDTAapparent	BRAICH3COOHprobbable	BRAICH3COOHapparent
April 0-30 cm				
M	1.49	1.55	1.00	1.00
Min	1.33	1.18	1.00	1.00
Max	1.76	1.68	1.40	1.13
April 30-60 cm				
M	1.80	1.92	1.00	1.00
Min	1.16	1.27	1.00	1.00
Max	2.66	2.79	2.06	1.72
May 0-30 cm				
M	1.59	1.79	1.00	1.00
Min	1.40	1.53	1.00	1.00
Max	1.87	1.97	1.00	1.00
May30-60 cm				
M	2.04	2.22	1.00	1.00
Min	1.48	1.63	1.00	1.00
Max	2.66	3.08	1.46	1.40
June 0-30 cm				
M	1.24	1.30	1.00	1.00
Min	1.09	1.06	1.00	1.00
Max	1.65	1.91	1.00	1.00
June 30-60				
M	1.09	1.25	1.00	1.00
Min	1.00	1.00	1.00	1.00
Max	1.55	1.81	1.46	1.44
July 0-30 cm				
M	1.25	1.28	1.00	1.00
Min	1.00	1.00	1.00	1.00
Max	1.50	1.67	1.00	1.00
July 30-60 cm				
M	1.09	1.28	1.00	1.00
Min	1.00	1.00	1.00	1.00
Max	1.17	1.52	1.00	1.00
August 0-30 cm				
M	1.41	1.59	1.09	1.05
Min	1.17	1.09	1.00	1.00
Max	1.87	1.82	2.00	1.36
August 30-60				
M	1.24	1.51	1.04	1.00
Min	1.09	1.03	1.00	1.00
Max	1.87	1.82	2.00	1.36

Table 14: The descriptive statistic (M−Median, Min−Minimum, Max−Maximum) of 21 element concentrations (mg kg^{-1}) (Al, As, B, Ba, Be, Ca, Cd, Co Cr, Cu, Fe, K, Mg, Mn, Na, Ni, Pb, Sb, Sr, V and Zn) and SIRM values (µA m^2 kg^{-1}) in the leaf samples (n=75) collected through the grapevine season

	Al	As	B	Ba	Be	Ca	Cd	Co	Cr	Cu	Fe	K	Mg	Mn	Na	Ni	Pb	Sb	Sr	V	Zn	SIRM
May																						
M	138	0.043	30.5	7.6	0.00371	11371	0.0074	0.037	0.140	9.53	134	6595	1302	49	42	3.10	0.28	0.00	11	0.153	21.1	37.9
Min	98	0.016	14.2	3.2	0.00155	6625	0.0007	0.024	0.025	6.32	85	4757	1007	22	8	0.03	0.01	0.00	3	0.066	13.3	22.4
Max	186	0.105	74.6	17.2	0.01150	25175	0.0780	0.110	0.770	15.83	253	9590	1831	274	103	9.69	1.90	0.66	38	0.239	44.8	80.4
June																						
M	62	0.042	41.8	12.3	0.00245	20719	0.0020	0.031	0.078	8.67	115	7612	1983	41	121	1.40	0.14	0.01	26	0.055	23.1	39.3
Min	44	0.016	22.9	5.2	0.000004	14465	0.0004	0.019	0.045	5.41	103	6843	1701	25	10	0.03	0.01	0.00	13	0.001	16.3	30.6
Max	83	0.088	57.4	27.1	0.01620	27603	0.1653	0.039	0.175	14.47	137	10071	2651	234	191	2.09	3.91	0.01	40	0.095	30.1	87.8
July																						
M	91	0.094	30.6	13.8	0.00263	24198	0.0026	0.070	0.351	5.22	131	7762	2242	59	101	2.26	0.44	0.02	30	0.108	13.3	47.2
Min	67	0.035	16.6	5.4	0.00046	17848	0.0001	0.034	0.205	4.23	88	5716	1495	26	25	0.03	0.11	0.00	13	0.052	9.1	26.0
Max	139	0.205	47.9	28.3	0.01673	35906	0.0139	0.214	1.793	13.44	367	12095	3883	274	136	6.59	1.22	0.24	45	0.439	20.6	201.5
August																						
M	53	0.075	20.3	17.3	0.00236	29917	0.0019	0.074	0.203	4.40	102	7215	2816	55	54	1.76	0.15	0.01	45	0.052	15.7	53.1
Min	22	0.031	14.2	8.3	0.00025	25411	0.0004	0.033	0.049	3.21	56	4587	1954	27	5	0.03	0.01	0.00	18	0.000	11.2	29.0
Max	89	0.310	47.5	37.2	0.01919	39573	0.0108	0.249	2.835	6.36	228	9238	3993	185	120	6.02	0.70	0.44	64	0.136	29.2	145.7

Table S15: The descriptive statistics of biological accumulation concentration (BAC) (-) representing bioaccumulation the elements from topsoil and subsoil to the leaves through the entire grapevine season

	Al	As	B	Ba	Be	Ca	Cd	Co	Cr	Cu	Fe	K	Mg	Mn	Na	Ni	Pb	Sb	Sr	V	Zn
BAC 0-30 cm																					
May																					
M	0.0022	0.0031	0.70	0.0312	0.0019	1.58	0.0034	0.0014	0.0011	0.19	0.0033	0.64	0.15	0.0439	0.07	0.0293	0.0153	0.0013	0.29	0.002376	0.24
Min	0.0008	0.0006	0.22	0.0142	0.0010	0.30	0.0003	0.0007	0.0001	0.06	0.0012	0.33	0.07	0.0120	0.01	0.0002	0.0003	0.0005	0.07	0.000996	0.11
Max	0.0030	0.0081	1.62	0.0660	0.0061	3.69	0.0392	0.0040	0.0059	0.39	0.0064	0.93	0.22	0.1547	0.18	0.0792	0.0892	0.1913	1.17	0.003799	0.46
June																					
M	0.0009	0.0028	1.04	0.0479	0.0012	2.72	0.0009	0.0011	0.0006	0.19	0.0027	0.70	0.25	0.0366	0.17	0.0137	0.0069	0.0019	0.51	0.000910	0.29
Min	0.0006	0.0011	0.67	0.0237	0.0000	0.50	0.0002	0.0008	0.0003	0.13	0.0023	0.62	0.22	0.0171	0.01	0.0002	0.0005	0.0009	0.25	0.000016	0.20
Max	0.0012	0.0062	1.58	0.1023	0.0077	4.19	0.0750	0.0016	0.0013	0.31	0.0031	0.98	0.28	0.2157	0.25	0.0318	0.1758	0.0048	0.98	0.001466	0.38
July																					
M	0.0014	0.0069	0.77	0.0569	0.0014	3.23	0.0012	0.0029	0.0028	0.12	0.0031	0.73	0.26	0.0546	0.14	0.0166	0.0288	0.0052	0.76	0.001734	0.17
Min	0.0010	0.0024	0.46	0.0281	0.0003	0.37	0.0000	0.0013	0.0018	0.04	0.0020	0.50	0.12	0.0096	0.04	0.0002	0.0079	0.0009	0.24	0.000826	0.11
Max	0.0019	0.0156	1.39	0.1117	0.0086	4.94	0.0057	0.0079	0.0142	0.28	0.0082	1.21	0.41	0.2291	0.21	0.0719	0.0647	0.0740	1.18	0.006401	0.24
August																					
M	0.0007	0.0057	0.46	0.0699	0.0012	3.58	0.0005	0.0028	0.0014	0.10	0.0025	0.61	0.31	0.0521	0.07	0.0146	0.0103	0.0026	1.00	0.000805	0.18
Min	0.0003	0.0021	0.31	0.0339	0.0001	0.81	0.0001	0.0012	0.0004	0.06	0.0013	0.40	0.16	0.0196	0.01	0.0002	0.0008	0.0008	0.42	0.000001	0.12
Max	0.0012	0.0244	1.12	0.1312	0.0093	5.88	0.0028	0.0090	0.0230	0.13	0.0051	0.85	0.49	0.1650	0.15	0.0655	0.0448	0.1482	1.50	0.002072	0.33
BAC 30-60 cm																					
May																					
M	0.0022	0.0026	1.30	0.0261	0.0021	1.73	0.0269	0.0016	0.0010	0.20	0.0030	0.71	0.16	0.0462	0.06	0.0269	0.0107	0.0009	0.14	0.001402	0.29
Min	0.0015	0.0010	0.22	0.0120	0.0012	0.11	0.0018	0.0008	0.0001	0.07	0.0018	0.55	0.11	0.0151	0.01	0.0001	0.0002	0.0004	0.03	0.000565	0.16
Max	0.0035	0.0073	2.83	0.0547	0.0064	4.21	0.3384	0.0048	0.0057	0.36	0.0061	1.37	0.23	0.2271	0.19	0.0683	0.0423	0.1150	0.75	0.002249	0.53
June																					
M	0.0010	0.0028	2.32	0.0506	0.0013	2.84	0.0065	0.0014	0.0005	0.22	0.0028	0.98	0.24	0.0400	0.16	0.0112	0.0040	0.0012	0.27	0.000506	0.33
Min	0.0006	0.0009	1.02	0.0243	0.0000	0.14	0.0012	0.0008	0.0003	0.08	0.0024	0.67	0.22	0.0215	0.02	0.0002	0.0003	0.0004	0.10	0.000009	0.18
Max	0.0023	0.0054	3.59	0.0826	0.0085	4.25	0.5260	0.0021	0.0013	0.34	0.0046	1.26	0.35	0.2040	0.28	0.0201	0.0964	0.0031	0.51	0.001114	0.41
July																					
M	0.0014	0.0063	0.84	0.0446	0.0012	3.33	0.0036	0.0031	0.0024	0.13	0.0029	0.78	0.25	0.0590	0.12	0.0137	0.0149	0.0126	0.51	0.001425	0.16
Min	0.0009	0.0023	0.51	0.0251	0.0002	0.19	0.0001	0.0006	0.0013	0.05	0.0019	0.53	0.13	0.0200	0.04	0.0002	0.0045	0.0005	0.13	0.000308	0.10
Max	0.0020	0.0133	2.62	0.0866	0.0085	5.46	0.0231	0.0095	0.0250	0.26	0.0091	1.21	0.37	0.2474	0.23	0.0637	0.0402	0.2974	1.21	0.006954	0.23
August																					
M	0.0007	0.0049	0.50	0.0621	0.0012	4.10	0.0020	0.0030	0.0015	0.12	0.0024	0.77	0.31	0.0498	0.07	0.0125	0.0075	0.0020	0.89	0.000561	0.20
Min	0.0003	0.0018	0.26	0.0215	0.0001	0.29	0.0003	0.0016	0.0002	0.06	0.0012	0.42	0.16	0.0227	0.01	0.0008	0.0004	0.0005	0.15	0.000001	0.11
Max	0.0061	0.0859	1.68	0.1169	0.0098	6.63	0.0108	0.0095	0.0201	0.16	0.0222	3.40	0.50	0.2208	0.15	0.0549	0.0238	0.1022	1.33	0.052076	0.33

Figures S1, S2, S3 and S4:

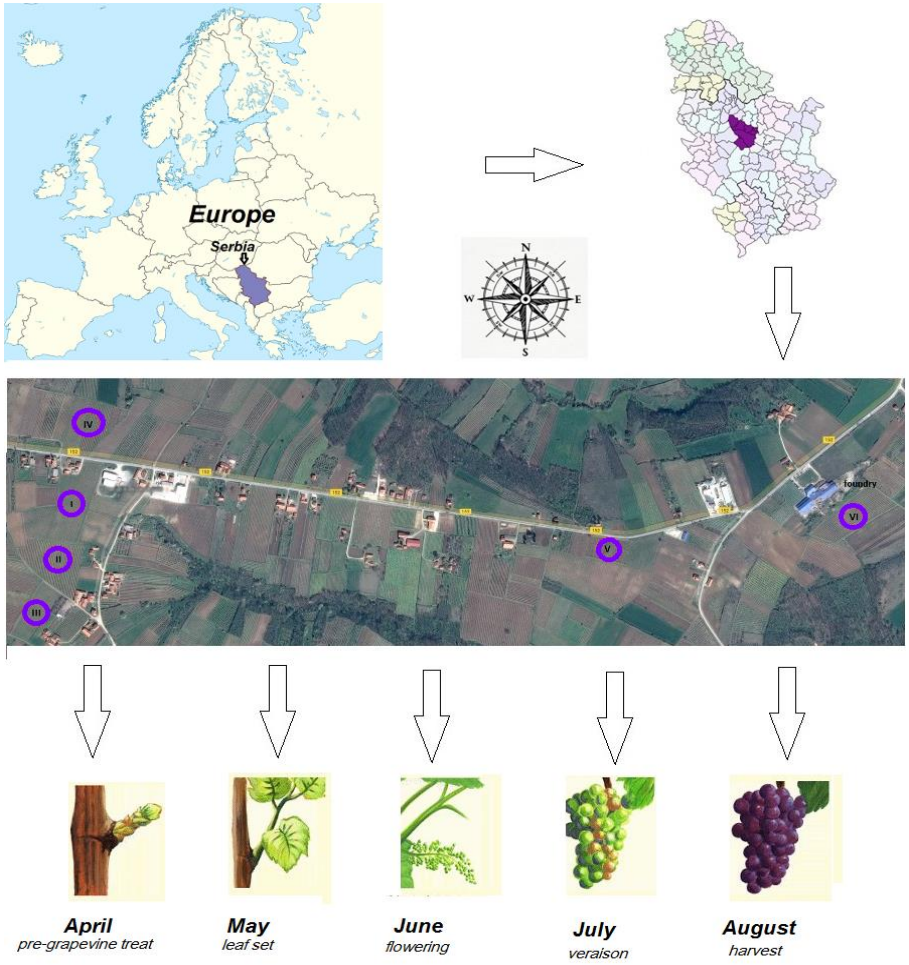

Figure S1. The location of the investigated vineyard, position of the investigated parcels and illustration of grapevine growing phases

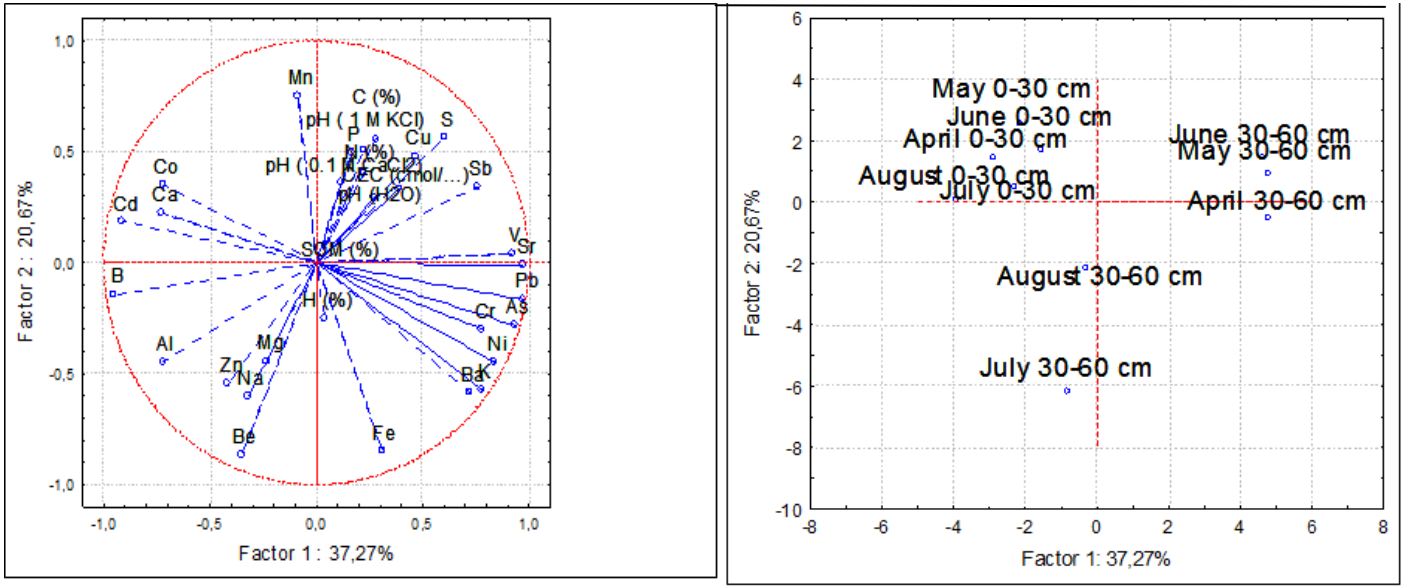

Figure S2. a) The PCA representing the relations between the element concentrations (mg kg^{-1}) and physicochemical parameters pH (-), SOM (%), CEC (cmol kg^{-}), N (%), C (%), H (%) and b) PCA Q-Q plot representing differences between the sampling phases (-) through the season

Figure S3. Environmental risk values (-) distribution through the season calculated for both soil layers for a) As; b) Cd; c) Cr; d) Cu; e) Ni; f) Pb; g) Zn and h) environmental risk (RI) (-) for the vineyard soil distribution through the season

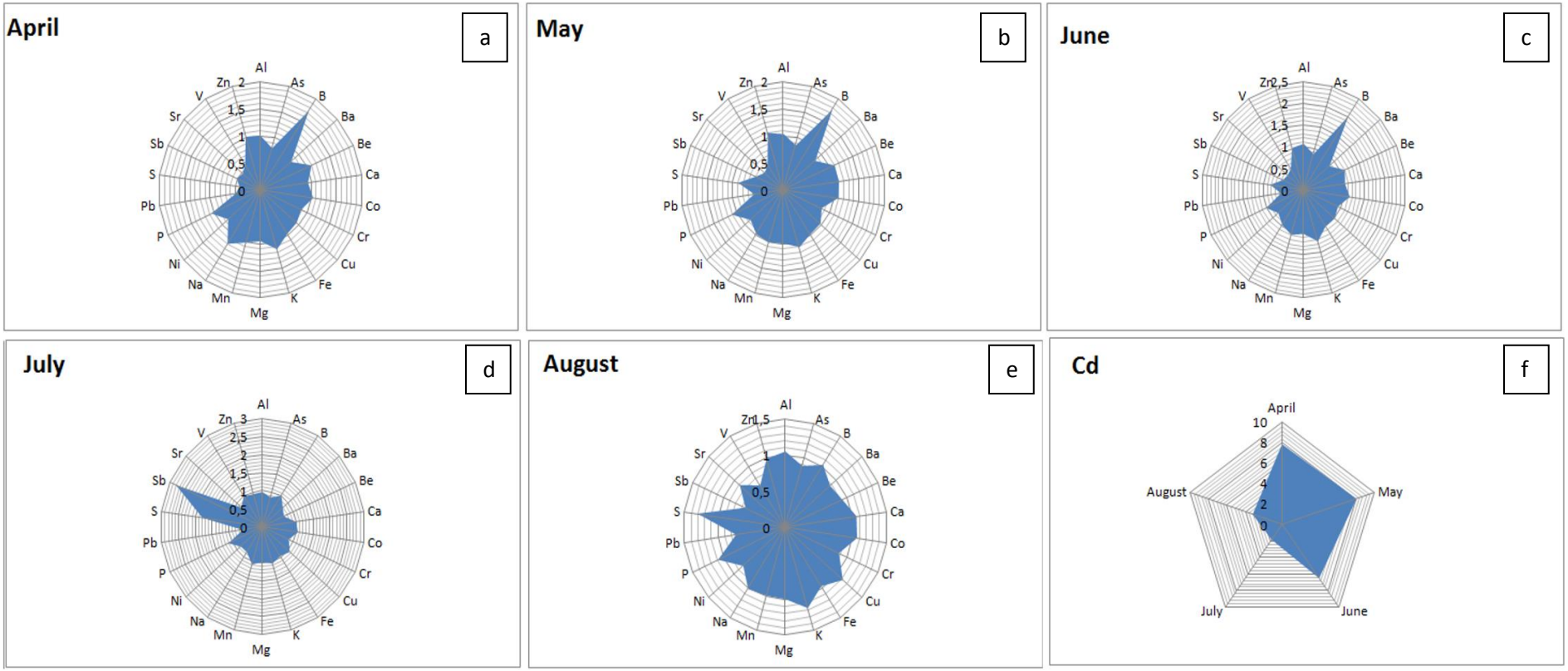

Figure S4: Biogeochemical absorption index (BGI) (-) representing the element absorption in the topsoil through the season a) April; b) May; c) June; d) July; e) August; and f) BGI (-) for Cd through the season.

Supplementary material for the section Materials and methods (theoretical part)

2.2. Chemical analyses

2.2.1. Soil preparation for element analyses

Each soil sample was air-dried in the laboratory conditions, sieved through 2 mm stainless sieves and finally ground to a fine powder in an agate mortar with a pestle. The hygroscopic moisture of the soil samples was determined by drying at 105°C until dry weight (d.w.). Acidity (pH) of the soil samples was determined in mixtures (1:5) of soil–distilled H_2O, soil–1mol L^{-1} KCl, and soil–0.1 mol L^{-1} $CaCl_2$. The organic matter was determined by the weighting the soil samples by the procedure described by Storer (1984). The cation exchange capacity (CEC) was measured applying the method recommended by the Soil Science Society of America (Sumner and Miller, 1996). Soil samples were prepared for the chemical analyses applying six single extraction procedures (deionised water during 2 and 16 h extracting, 0.01 mol L^{-1} $CaCl_2$ during 3 h extracting, 0.1 mol L^{-1} NH_4NO_3 during 2 h extracting, 0.05 mol L^{-1} Na_2EDTA during 1 h extracting and 0.11 mol L^{-1} CH_3COOH during 16 h extracting) (Ure, 1996; Quevauviller, 1998; Pueyo et al., 2004; Milićević et al., 2017a; Milićević et al., 2018) and pseudo-total digestion (35% HCl: 65% HNO_3=3:1 in microwave digester) (US EPA 3050b Method). All chemicals used for samples preparation were of analytical grade and produced by Sigma–Aldrich.

2.2.2. Leaf samples preparation for the element and magnetic particle analyses

The grapevine leaves were collected at the same sampling sites as the soil samples. Then, the leaf samples were dried in an oven at 45°C for 24 h and powdered in an agate mortar. For element determination, the homogenised leaf samples were digested with 7 mL 65% HNO_3 (Sigma–Aldrich, puriss p.a.) and 1 mL 30% H_2O_2 (Sigma–Aldrich, puriss p.a.) in a microwave digester (US EPA 3050 Method). A portion of the homogenised leaf samples (0.5 g) was tightly packed with cling film and pressed in a 10 cm^3 plastic container for the Saturation Isothermal Remanent Magnetisation (SIRM) analysis.

2.2.3. Element analyses

In the soil samples collected through the entire grapevine season from two depths, the concentrations of 23 elements (Al, As, B, Ba, Be, Ca, Cd, Co, Cr, Cu, Fe, K, Mg, Mn, Na, Ni, P, Pb, S, Sb, Sr, V and Zn) were determined using inductively coupled plasma-optical emission spectrometry (ICP-OES, Thermo Scientific iCAP 6500 Duo, Thermo Scientific, UK). For the calibration, a Multi-Element Plasma Standard Solution 4, Specpure (Alfa Aesar GmbH & Co KG, Germany) was used. This method was used for determining 13 elements (Al, B, Ba, Ca, Cu, Fe, K, Mg, Mn, Na, Ni, Sr and Zn) in the leaf samples. The concentrations of other nine elements (As, Be, Cd, Co, Cr, Pb, Sb and V) in the leaves were determined using inductively coupled plasma-mass spectrometry (ICP-MS, Thermo Scientific iCAP Q, Thermo Scientific, UK). For the ICP-OES and ICP-MS calibration, a low-level Elements Calibration Stock, EPA Method Standard (VHG Labs, Manchester) was used.

2.2.4. Magnetic particles analysis

The SIRM determination was performed by the method described by Hofman et al. (2014). Previously crushed and homogenised leaf samples were packed in the plastic containers and magnetised with a pulsed magnetic field of 1 T with a Molspin pulse magnetizer (Molspin Ltd, UK). After magnetisation, the magnetic intensity of the sample was measured using a Molspin Minispin magnetometer with high sensitivity ($\sim 0.1 \times 10^{-8}$ A m^2, Molspin Ltd, UK). The isothermal remanent magnetization (IRM) of the samples (A m^2) was measured twice. The instrument was calibrated before and after every ten measurements by means of a magnetically-stable rock specimen, as described by Mitchell et al. (2010). This IRM at 1 T is taken to be the saturation remanent magnetisation (SIRM) of each measured sample. The magnetic intensity values (mA m^{-1}), were normalised to the sampling container volume (10 cm^3) and for the leaf mass (g), and finally expressed in A m^2 kg^{-1}.

2.3. Quality of measurements and assurance

The blank samples and certified reference materials (CRMs) were analysed once every 10 samples. Four CRMs: 2711a (Montana II Soil), BCR 143R (Sewage Sludge-Amended Soil), ERM CC 135a (Contaminated Brickworks Soil) and SARM 42 (Soil) were analysed to validate the pseudo-total protocol. According to the CRMs, the recovery for pseudo-total soil analysis of the elements was ranged between 80% and 120%. For validation of the single extraction protocols, the calibrations were prepared using a low-level Elements Calibration Stock, EPA Method Standard (VHG Labs, Manchester) and for the preparation of the standard series for the calibration of every specific single extraction protocol, the matrix matching technique was applied to eliminate the problems related to the occurrence of the matrix effect during the determination of the element concentrations in the extracts. In addition, the results after the determination on were selectively chosen firstly, according to LOD of the method and secondly, according to Relative Standard Deviation (RSD<20%) of three measurements of every sample. Most of the samples were analysed in triplicates and according to results for these three analysed subsamples Standard Deviation and Relative Standard Deviation (RSD: 0.4─32%; Table S2) were calculated as one of the single extraction parameters important for the validation. In addition, for the single extraction protocols validation, BCR 483 (Sewage Sludge-Amended Soil) CRM was also used. The element recovery was from 72% to 131% (for Cd, Ni, Pb and Zn extracted by $CaCl_2$, NH_4NO_3, Na_2EDTA; and Cr and Cu extracted by NH_4NO_3, Na_2EDTA). Limits of the detection (LOD) for the elements extracted with $CaCl_2$ were: Cd (λ=226.5 nm), LOD=0.06 µg L^{-1}; Ni (λ=231.6 nm), LOD=0.24 µg L^{-1}; Pb (λ=220.3 nm), LOD=0.88 µg L^{-1}; and Zn (λ=213.8 nm), LOD=0.07 µg L^{-1}. With NH_4NO_3, LODs were: Cd (λ=226.5 nm), LOD=0.06 µg L^{-1}; Cr (λ=205.5 nm), LOD=0.16 µg L^{-1}; Cu (λ=213.5 nm), LOD=0.32 µg L^{-1}; Ni (λ=231.6 nm), LOD=0.23 µg L^{-1}; Pb (λ=220.3 nm), LOD=0.87 µg L^{-1}; Zn (λ=213.8 nm): LOD=0.08 µg L^{-1}, and with Na_2EDTA LODs were: Cd (λ=226.5 nm), LOD=0.08 µg L^{-1}; Cr (λ=205.5 nm), LOD=0.19 µg L^{-1}, Cu (λ=213.5 nm), LOD=0.41 µg L^{-1}; Ni (λ=231.6 nm). LOD=0.33 µg L^{-1}; Pb (λ=220.3 nm), LOD=1.23 µg L^{-1}; and Zn (λ=213.8 nm), LOD=0.21 µg L^{-1}. Exceptions were the recoveries for Cr and Cu extracted with $CaCl_2$ − 138%, and 139%, respectively, which were recalculated according to the value in the reference materials, and these results should be approached cautiously (LODs: Cr (λ=205.5 nm), LOD=0.18 µg L^{-1} and Cu (λ=213.5 nm), LOD=0.36 µg L^{-1}). For validation of the leaf sample protocols, the moss *Pleurozium schreberi*, M2 and M3, were used as the certified reference materials (Steinnes, 1997). Recovery of the elements in M2 ranged from 76% to 108%, except for Cr (69%). For M3, the element recoveries were ranged from 78% to 117%, except recovery of Cr which was 58%. For Cr, the measured concentrations were recalculated according to the recovery. Concentrations of Cr in analysed leaves were recalculated according to the relevant value in the reference materials.

2.4. Data processing

Statistical analyses were performed using the SPSS software version 21 for Windows and Statistic 8 (Stat Soft Inc., Tulsa, OK, USA). The normality ($p<0.05$) of the data was tested by the Kolmogorov-Smirnov test. For determining differences ($p<0.05$) in the element concentrations between two soil layers, different vineyard's parcels and the leaves from studied grapevine species, the Wilcoxon signed-rank test was applied. The principal component (PCA) and correlation analyses were used to indicate associations of the elements in the soil or leaf samples, the bioavailability of the elements and to determine the most suitable single extraction procedures for the vineyard soil. In addition, PCA, Spearman's correlation (R) and regression (R^2) analyses were used to determine associations between environmental risk assessment, biogeochemical factor, mobility factor, bioavailability risk, and bioaccumulation factor.

2.4.1. Ecological and bioavailability risk assessment

For predicting mobility of the elements throughout efficiency and selectivity of different single-extraction procedures the mobility factor (MF%) was calculated for the extracted elements according to Equation S1:

$$MF\% = (C_{se}/C_{pt}) \times 100 \qquad (S1)$$

where C_{se} represents the element concentrations (mg kg^{-1}) extracted from the soil by single extractions and C_{pt} is pseudo-total concentrations (mg kg^{-1}) of the elements.

For assessment of Contamination factor (CF) of the PTE measured in the soil, the following elements: Al, As, B, Ba, Be, Cd, Co, Cr, Cu, Fe, P, Pb, S, Sb, Sr, V and Zn were considered in the calculation. The other measured elements were not considered in the applied CF calculation because nowadays there are no proves about their hazardous or toxic effects to humans. For assessing the soil pollution during the grapevine growing period in the investigated vineyard parcels and to compare the element concentrations in the vineyard soil with the concentrations in the local background soil sample, the CF was calculated according to the Equation S2:

$$CF = C_n/B_n \qquad (S2)$$

where C_n represents an element's concentration (mg kg^{-1}) in soil from the vineyard and B_n is the local background concentration (mg kg^{-1}) of the element in the soil (Likuku et al., 2013; Milićević et al., 2017).

The ecological risk (Equations S3, S4) was calculated for assessing the environmental implications of elements in the vineyard soil samples through the entire season. The ecological risks (Eri and RI) were calculated according to the equations defined by Hakason (1980) and they were usually used in environmental studies for assessing the risk of sediments or soils (Hui-na et al., 2012; Mazurek et al., 2017; Ghasemi et al., 2018):

$$Eri=TR\times CF \quad (S3)$$

$$RI=\Sigma Eri \quad (S4)$$

where CF is contamination factor; TR is toxic response factor given for the elements (it was reported by Hakason (1980) only for As, Cd, Cr, Cu, Ni, Pb, and Zn with known values 10, 30, 2, 5, 5, 5, and 1 respectively). Eri is the potential risk of every individual element (i=As, Cd, Cr, Cu, Pb, and Zn). In Equation S4, RI represents the sum of the potential risks of individual elements.

For assessing the contamination of topsoil against subsoil the Biogeochemical index (BGI) (Mazurek et al., 2017) for all measured elements (Equation S5) was calculated:

$$BGI=C_{topsoil}/C_{subsoil} \quad (S5)$$

Bioavailability risk assessment index (BRAI) was calculated for quantification of the bioavailability risk of PTEs from the vineyard soil according to the equation proposed by Jamshidi-Zanjani et al. (2015) (Equation S6). This index was developed for element concentrations extracted by Na_2EDTA, with the main objective to assess the risk of PTEs bioavailability (Equation S6).

$$BRAI=\Sigma Bdi\ (i=1\ to\ n)/\Sigma TEi\ (i=1\ to\ n) \quad (S6)$$

where n is the number of the PTE, TE is the toxic effect of the PTE (As, Cd, Cr, Cu, Ni, Pb and Zn) derived from the effect range median (ERM) values presented by Long et al. (1995), calculated using probable effect levels (PEL) published by NOAA (2004). Bdi presents the bioavailable degree of each metal in the vineyard soil sample, which was calculated by applying following Equations S7:

$$Bdi = MF\times TEi \quad (S7)$$

where the source effect (ME) represents the release potential of the bioavailable fraction of the elements (Jamshidi-Zanjani et al., 2015). This BRAI was named as $BRAI_{probable}$. Besides $BRAI_{probable}$, $BRAI_{apparent}$ was calculated using a larger element set (As, Cd, Co, Cr, Cu, Mn, Ni, Pb, V and Zn), and for the calculation of TE values the apparent effects threshold values (AET) were used (NOAA, 2004). Thus the calculated BRAI values further will be represented as a $BRAI_{probable}$ and $BRAI_{apparent}$ (NOAA, 2004). In this study, the BRAI was calculated for the element concentrations extracted with Na_2EDTA (regular equation) and CH_3COOH (modified equation).

Bioaccumulation (Equation S8) of the elements in the grapevine leaf was evaluated by calculation of the biological absorption coefficient (BAC):

$$BAC=C_{leaf}/C_{soil} \quad (S8)$$

where C_{leaf} represent the element concentration in leaf and C_{soil} represents element concentration in topsoil or subsoil (Bravo et al., 2017).