Supporting Information to

Characterization of fresh PM deposits on calcareous stone surfaces: seasonality, source apportionment and soiling potential

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Table S1: List of PM samples and the corresponding meteorological parameters obtained from the nearest station in Ljubljana, Slovenia. Meteorological data was acquired from the Slovenian Environment Agency (ARSO), as well as $PM_{2.5}$ concentration marked with asterisk (*).

Sampling date (start)	Sample name	Sampling time	Average T [°C]	Max. T [°C]	Min. T [°C]	Precip. [mm]	Sun [h]	Cloudy [%]	$PM_{2.5}$* [μg m^{-3}]	$PM_{2.5}$ [μg m^{-3}]	$PM_{0.8}$ [μg m^{-3}]
Summer 2020											
03.08.2020	PM2.5(Q)-20200803	12:00-12:00	19.0	23.7	17.8	17.5	0.5	97	9	16	n.a.
04.08.2020	PM2.5(Q)-20200804	12:00-12:00	17.9	24.7	16.5	21.2	2.2	83	5	14	n.a.
05.08.2020	PM2.5(Q)-20200805	12:00-12:00	17.2	22.1	14.9	2.8	3.5	80	4	14	1
06.08.2020	PM2.5(Q)-20200806	12:00-12:00	21.4	26.5	17.0	0.9	7.3	50	8	21	3
07.08.2020	PM2.5(Q)-20200807	12:00-12:00	24.2	30.7	17.1	0	10.6	40	16	29	7
08.08.2020	PM2.5(Q)-20200808	12:00-12:00	25.1	31.5	16.8	0	13.3	3	17	31	9
09.08.2020	PM2.5(Q)-20200809	12:00-12:00	25.1	32.2	17.5	0	12.2	27	20	28	10
10.08.2020	PM2.5(PC)-20200810	12:00-12:00	25.6	32.0	18.6	0	11.3	23	21	22	11
11.08.2020	PM2.5(Q)-20200811	12:00-12:00	22.8	28.7	19.2	0	3.6	53	17	22	8
12.08.2020	PM2.5(Q)-20200812	12:00-12:00	24.1	30.0	17.3	0.3	7.0	47	17	19	8
13.08.2020	PM2.5(Q)-20200813	12:00-12:00	25.4	32.0	18.7	0	9.8	47	16	27	8
14.08.2020	PM2.5(Q)-20200814	12:00-12:00	21.3	29.3	17.6	0	7.0	70	10	21	5
15.08.2020	PM2.5(Q)-20200815	12:00-12:00	22.3	28.0	17.0	25.9	7.1	73	9	21	4
16.08.2020	PM2.5(Q)-20200815	12:00-12:00	23.4	29.0	18.8	0	7.9	60	11	10	5
17.08.2020	PM2.5(Q)-20200817	12:00-12:00	22.1	31.4	18.5	0	5.9	73	9	14	4
Autumn 2020											
09.11.2020	PM2.5(Q)-20201109	15:10-15:10	4.4	6.0	1.9	0	0	100	27	26	n.a.
10.11.2020	PM2.5(Q)-20201110	17:10-24:00	5.0	5.6	4.2	0.2	0	100	17	n.a.	n.a.
11.11.2020	PM2.5(Q)-20201111	00:00-24:00	5.7	7.2	5.0	0	0	100	12	n.a.	11
12.11.2020	PM2.5(Q)-20201112	00:00-24:00	6.5	8.2	4.5	1.2	0.3	87	22	27	10
13.11.2020	PM2.5(Q)-20201113	00:00-24:00	6.8	11.6	4.5	0.3	4.0	67	29	36	12
14.11.2020	PM2.5(Q)-20201114	00:00-24:00	8.0	13.5	3.5	0	6.0	70	29	33	18
15.11.2020	PM2.5(Q)-20201115	00:00-24:00	7.7	9.8	3.2	0	1.2	80	31	32	22
16.11.2020	PM2.5(Q)-20201116	00:00-24:00	7.9	10.8	7.5	0	0	100	14	14	22
17.11.2020	PM2.5(Q)-20201117	00:00-24:00	7.7	13.0	4.5	42.9	5.3	53	12	14	9
18.11.2020	PM2.5(Q)-20201118	00:00-24:00	5.2	10.0	3.2	0	5.0	67	25	24	10

19.11.2020	PM2.5(Q)-20201119	00:00-24:00	3.6	4.4	2.5	0.3	0	100	23	22	21
20.11.2020	PM2.5(Q)-20201120	00:00-24:00	6.0	9.2	3.6	1.8	1.1	73	7	8	6
21.11.2020	PM2.5(Q)-20201121	00:00-24:00	2.1	7.5	-0.7	0.3	8.0	7	10	22	8
22.11.2020	PM2.5(Q)-20201122	00:00-24:00	0.2	7.5	-4.6	0	7.8	13	29	32	27
Winter 2021											
15.02.2021	PM2.5(Q)-20210215	8:30-8:30	-0.3	6.6	-7.5	0	8.6	7	15	13	12
16.02.2021	PM2.5(Q)-20210216	8:30-8:31	4.4	10.0	0.1	0	7.9	63	19	27	13
17.02.2021	PM2.5(Q)-20210217	8:30-8:32	5.4	11.1	2.0	0	4.9	27	24	34	19
18.02.2021	PM2.5(Q)-20210218	8:30-8:33	5.6	9.8	-1.5	0.2	3.5	93	33	28	25
19.02.2021	PM2.5(Q)-20210219	8:30-8:34	8.3	10.5	5.0	0	0.5	97	31	20	20
20.02.2021	PM2.5(Q)-20210220	8:30-8:35	8.9	13.9	4.5	0	5.7	70	26	16	20
21.02.2021	PM2.5(Q)-20210221	8:30-8:36	9.6	13.4	6.5	0	2.0	80	28	23	17
22.02.2021	PM2.5(Q)-20210222	8:30-8:37	10.6	14.2	7.2	0	6.4	27	15	6	8
23.02.2021	PM2.5(Q)-20210223	8:30-8:38	8.8	19.8	0.9	0	8.7	0	29	28	22
24.02.2021	PM2.5(Q)-20210224	8:30-8:39	9.7	19.6	1.7	0	8.5	0	38	29	26
25.02.2021	PM2.5(Q)-20210225	8:30-8:40	9.6	20.6	2.3	0	8.8	0	37	n.a.	24
26.02.2021	PM2.5(Q)-20210226	8:30-8:41	10.0	21.7	1.5	0	9.1	0	37	41	23
27.02.2021	PM2.5(Q)-20210227	8:30-8:42	7.0	12.6	2.8	0	4.4	47	19	6	20
28.02.2021	PM2.5(Q)-20210228	8:30-8:43	5.3	12.2	0.0	0	9.7	0	10	5	n.a.
Spring 2021											
24.05.2021	PM2.5(Q)-20210524	9:10-9:10	10.4	11.5	7.8	3.6	0	100	5	n.a.	4
25.05.2021	PM2.5(Q)-20210525	9:45-9:45	11.6	20.0	8.4	17.9	2.9	87	6	11	4
26.05.2021	PM2.5(Q)-20210526	9:45-9:45	14.9	21.5	6.5	7.2	11.5	37	5	n.a.	4
27.05.2021	PM2.5(Q)-20210527	9:45-9:45	15.4	24.5	8.6	0	9.6	73	7	n.a.	4
28.05.2021	PM2.5(Q)-20210528	9:45-9:45	16.0	21.6	9.6	2.7	9.7	60	4	9	3
29.05.2021	PM2.5(Q)-20210529	9:45-9:45	13.4	20.9	10.3	0	4.7	87	6	n.a.	4
30.05.2021	PM2.5(Q)-20210530	8:43-8:43	13.3	18.9	8.7	5.1	8.1	60	6	n.a.	4
31.05.2021	PM2.5(Q)-20210531	8:43-8:43	15.4	20.7	6.3	0.4	11.6	43	6	n.a.	4
01.06.2021	PM2.5(Q)-20210601	8:43-8:43	17.5	22.8	9.0	0	12.7	33	8	n.a.	5
02.06.2021	PM2.5(Q)-20210602	8:43-8:43	18.9	25.0	10.3	0	13.9	30	9	17	6
03.06.2021	PM2.5(Q)-20210603	8:43-8:43	19.5	27.5	11.0	0	12.8	30	11	7	6

04.06.2021	PM2.5(Q)-20210604	8:43-8:43	20.3	28.7	12.8	0	12.7	37	13	n.a.	8
05.06.2021	PM2.5(Q)-20210605	8:43-8:43	22.5	28.5	14.2	0	12.5	33	12	4	7
06.06.2021	PM2.5(Q)-20210606	8:43-8:43	19.3	22.7	17.0	2.3	1.5	87	9	n.a.	6
Summer 2021											
21.06.2021	PM2.5(Q)-20210621	8:45-8:45	26.7	32.8	20.3	0	13.2	17	23	12	4
22.06.2021	PM2.5(Q)-20210622	8:45-8:45	26.3	32.6	18.8	0	14.4	10	17	4	4
23.06.2021	PM2.5(Q)-20210623	8:45-8:45	28.0	34.4	17.3	0	14.3	27	19	13	5
24.06.2021	PM2.5(Q)-20210624	8:45-8:45	27.7	34.0	20.3	0	13.8	17	21	15	6
25.06.2021	PM2.5(Q)-20210625	8:45-8:45	24.9	31.2	19.2	0	14.2	33	8	n.a.	4
26.06.2021	PM2.5(Q)-20210626	8:45-8:45	24.9	30.9	17.8	0	13.8	17	8	n.a.	4
27.06.2021	PM2.5(Q)-20210627	8:45-8:45	25.8	31.2	18.0	0	13	33	10	n.a.	6
28.06.2021	PM2.5(Q)-20210628	8:45-8:45	27.5	35.2	18.6	0	14.6	10	13	n.a.	6
29.06.2021	PM2.5(Q)-20210629	8:45-8:45	27.1	34.5	18.4	0	13.3	27	15	16	5
30.06.2021	PM2.5(Q)-20210630	8:45-8:45	25.2	31.3	19.0	0	13.2	27	10	n.a.	3
01.07.2021	PM2.5(Q)-20210701	8:45-8:45	17.4	24.6	14.5	6.6	8.6	50	5	n.a.	3
02.07.2021	PM2.5(Q)-20210702	8:45-8:45	20.7	26.8	13.5	4.5	8.7	60	6	n.a.	3
03.07.2021	PM2.5(Q)-20210703	8:45-8:45	23.9	30.4	14.3	0	13.7	40	7	n.a.	n.a.
04.07.2021	PM2.5(Q)-20210704	8:45-8:45	20.3	25.7	18.6	0	0.7	97	9	n.a.	n.a.

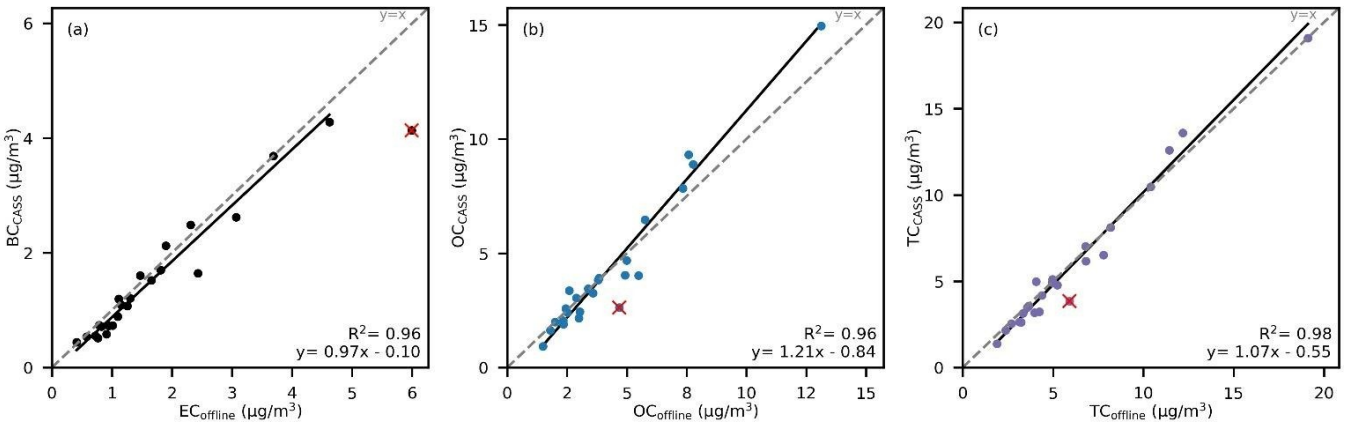


Figure S1: Correlation between online (CASS) and offline a) black carbon/elemental carbon (BC/EC), b) organic carbon (OC), and c) total carbon (TC) measurements. Outliers with the absolute studentized residual higher than or equal to 3 are marked with red crosses and were eliminated from the regression calculation.

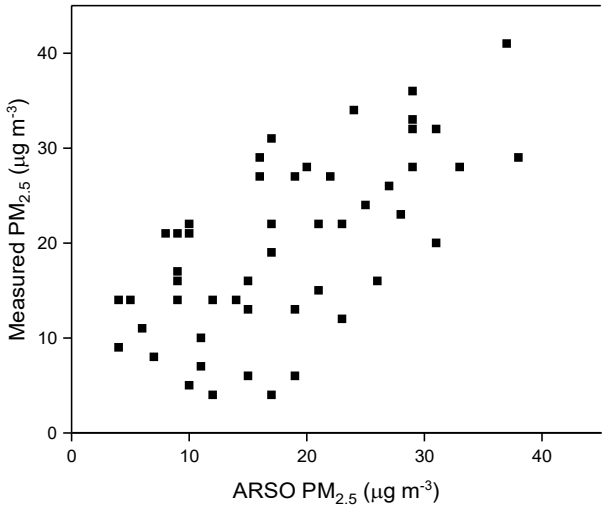


Figure S2: Correlation between measured $PM_{2.5}$ concentrations and those reported by the Slovenian Environment Agency (ARSO) for the nearest sampling site (Lj-Bežigrad, approx. 5 km air distance).

Table S2: Airborne anion concentrations in $PM_{2.5}$ from Ljubljana, Slovenia. MS stands for methanesulfonate.

Sample name	Fluoride [μg m^{-3}]	Lactate [μg m^{-3}]	MS [μg m^{-3}]	Chloride [μg m^{-3}]	Nitrite [μg m^{-3}]	Nitrate [μg m^{-3}]	Sulfate [μg m^{-3}]	Oxalate [μg m^{-3}]	Phosphate [μg m^{-3}]
Summer 2020									
PM2.5(Q)-20200803	0	0	0.0338	0.0253	0.0882	0.2387	0.9273	0.1591	0
PM2.5(Q)-20200804	0	0	0	0.0239	0.0868	0.1074	0.3432	0.0788	0.0031
PM2.5(Q)-20200805	0	0	0	0.0241	0.0908	0.1267	0.5833	0.0864	0
PM2.5(Q)-20200806	0	0	0.0741	0.0210	0.0853	0.3411	1.6370	0.1891	0
PM2.5(Q)-20200807	0.0217	0.2747	0.0731	0.0302	0.0389	0.3187	3.5266	0.2919	0.0132
PM2.5(Q)-20200808	0.0187	0.1924	0.0783	0.0324	0.0451	0.3964	3.5916	0.3510	0.0131
PM2.5(Q)-20200809	0.0180	0	0.0470	0.0245	0.1070	0.4881	4.3142	0.3587	0.0159
PM2.5(PC)-20200810	0	0.4801	0.0263	0.0322	0.0162	0.4725	3.7466	0.3408	0.0118
PM2.5(Q)-20200811	0.0224	0	0.0378	0.0382	0.1046	0.3700	2.1111	0.2633	0.0089
PM2.5(Q)-20200812	0	0	0.0320	0.0381	0.0242	0.6507	2.6736	0.3644	0.0110
PM2.5(Q)-20200813	0.0185	0.2309	0.0419	0.0252	0.0183	0.3431	3.0136	0.2614	0.0113
PM2.5(Q)-20200814	0	0.1419	0.0113	0.0278	0.1628	0.6066	1.7636	0.1622	0
PM2.5(Q)-20200815	0	0	0.0184	0.0322	0.0162	0.3225	1.7310	0.1968	0
PM2.5(Q)-20200816	0.0164	0.1662	0.0242	0.0224	0.0158	0.2869	1.3817	0.1473	0
PM2.5(Q)-20200817	0	0	0	0.0270	0.0859	0.3421	1.2730	0.1420	0
Autumn 2020									
PM2.5(Q)-20201109	0.0384	0	0	0.1125	0.2430	1.9516	6.4270	0.2404	0
PM2.5(Q)-20201110	0	0	0	0	0	0	0	0	0
PM2.5(Q)-20201111	0	0	0.0246	0.1479	0	3.3756	4.6971	0.1706	0
PM2.5(Q)-20201112	0	0	0.0471	0.2869	0.0827	4.0138	2.9200	0.2134	0.0160
PM2.5(Q)-20201113	0	0	0	0.3455	0.0713	3.4843	1.6754	0.2047	0.0123
PM2.5(Q)-20201114	0	0	0	0.4621	0.0543	3.0857	2.2120	0.2481	0.0112
PM2.5(Q)-20201115	0.0129	0	0	0.1086	0.0668	3.0729	0.9480	0.1610	0
PM2.5(Q)-20201116	0.0239	0	0	0.1741	0.1087	1.3308	0.6561	0.1258	0
PM2.5(Q)-20201117	0	0	0	0.5157	0	1.9317	1.1310	0.1342	0.0099
PM2.5(Q)-20201118	0.0174	0	0	0.2973	0.0534	1.8653	1.9102	0.1378	0
PM2.5(Q)-20201119	0	0	0	0.0492	0.0563	0.9913	0.7167	0.0983	0

PM2.5(Q)-20201120	0	0	0	0.0408	0.0924	0.7710	0.6991	0.0745	0
PM2.5(Q)-20201121	0.0207	0	0	0.4542	0.0246	3.4719	1.2865	0.1547	0.0180
Winter 2021									
PM2.5(Q)-20210215	0	0	0	0.0861	0	4.1059	0.5174	0.1236	0.0063
PM2.5(Q)-20210216	0	0	0.0258	0.1101	0.0178	9.2775	0.8049	0.1722	0.0067
PM2.5(Q)-20210217	0	0	0.0102	0.3464	0.0156	5.7740	1.1492	0.1798	0.0172
PM2.5(Q)-20210218	0	0	0.0259	0.1471	0	15.265	1.9324	0.3008	0
PM2.5(Q)-20210219	0	0	0.0281	0.2032	0.0205	16.016	2.0344	0.2993	0
PM2.5(Q)-20210220	0	0	0	0.2666	0.0881	8.3124	1.6997	0.2870	0.0136
PM2.5(Q)-20210221	0	0	0.0318	0.1427	0.0813	10.223	2.1101	0.2829	0
PM2.5(Q)-20210222	0	0	0	0.1846	0.0925	2.8643	1.8905	0.2670	0.0070
PM2.5(Q)-20210223	0.0243	0	0.0268	0.2527	0.1103	3.4439	2.0008	0.2217	0.0190
PM2.5(Q)-20210224	0	0	0.0170	0.2013	0.0197	2.1125	1.3412	0.1700	0.0275
PM2.5(Q)-20210225	0	0	0.0172	0.2126	0.0270	2.0921	1.3699	0.1901	0.0208
PM2.5(Q)-20210226	0.0330	0	0.0119	0.2223	0.1028	1.8930	1.2954	0.1821	0.0264
PM2.5(Q)-20210227	0	0	0	0.1256	0.0131	1.6292	1.0524	0.1232	0
PM2.5(Q)-20210228	0.0197	0	0	0.1357	0.0911	2.1751	1.2911	0.1588	0.0186
Spring 2021									
PM2.5(Q)-20210524	0.0167	0	0	0.0374	0.0931	1.7574	0.5270	0.1059	0
PM2.5(Q)-20210525	0.0148	0	0	0.0387	0.0702	0.5719	0.5242	0.0883	0
PM2.5(Q)-20210526	0	0	0.0261	0.0451	0	0.3666	0.6692	0.1060	0
PM2.5(Q)-20210527	0.0192	0	0.0192	0.0374	0.1057	0.4397	0.9807	0.1304	0
PM2.5(Q)-20210528	0.0141	0	0.0318	0.0298	0.0814	0.4761	1.1940	0.1408	0
PM2.5(Q)-20210529	0.0054	0.0846	0.0150	0.0318	0.0161	0.3023	0.6182	0.1021	0
PM2.5(Q)-20210530	0.0165	0	0.0540	0.0346	0.0879	0.3485	1.3618	0.1224	0
PM2.5(Q)-20210531	0.0187	0	0.0432	0.0340	0.0978	0.3391	1.1633	0.1482	0
PM2.5(Q)-20210601	0.0183	0	0.0644	0.0337	0.0984	0.4034	1.6247	0.1878	0.1225
PM2.5(Q)-20210602	0	0	0.0430	0.0337	0	0.4126	1.6174	0.1476	0
PM2.5(Q)-20210603	0.0185	0	0.0566	0.3198	0.3336	0.7221	1.6982	0.2380	0.0123
PM2.5(Q)-20210604	0.0247	0	0.0593	0.0765	0.1183	0.6522	2.2461	0.2906	0
PM2.5(Q)-20210605	0	0	0.0643	0.0323	0	0.5069	2.4968	0.2444	0

PM2.5(Q)-20210606	0.0188	0	0	0.0380	0.1012	0.3386	1.2260	0.1707	0
Summer 2021									
PM2.5(Q)-20210621	0	0	0.0532	0.1220	0	0.2705	2.1445	0.2163	0
PM2.5(Q)-20210622	0.0161	0.1067	0.0827	0.0278	0.0754	0.3225	1.5969	0.172	0
PM2.5(Q)-20210623	0	0	0.0729	0.0575	0	0.3365	1.8289	0.3008	0
PM2.5(Q)-20210624	0	0	0.0756	0.0379	0.0655	0.2602	2.1631	0.2388	0
PM2.5(Q)-20210625	0	0	0.0603	0.0803	0.0711	0.3906	1.4617	0.1965	0
PM2.5(Q)-20210626	0	0	0	0.0386	0.0711	0.2348	0.7527	0.1902	0
PM2.5(Q)-20210627	0.0075	0.1489	0.0257	0.0351	0.0244	0.3187	0.9283	0.2416	0
PM2.5(Q)-20210628	0.0210	0	0.0414	0.0262	0.0760	0.3271	1.2216	0.2273	0
PM2.5(Q)-20210629	0	0	0.0474	0.0370	0.0180	0.2739	1.7775	0.2302	0.0077
PM2.5(Q)-20210630	0.0209	0	0.0669	0.0296	0.0771	0.3189	0.7623	0.1367	0
PM2.5(Q)-20210701	0	0	0	0.0363	0.0264	0.3116	0.3222	0.0928	0
PM2.5(Q)-20210702	0.0091	0.1821	0.0081	0.0501	0.0274	0.2930	0.3101	0.1109	0
PM2.5(Q)-20210703	0	0.4585	0.0468	0.1550	0.0838	0.3155	1.0364	0.1923	0.0149
PM2.5(Q)-20210704	0.0180	0	0.0579	0.0270	0.0919	0.2920	1.0377	0.2090	0

Table S3: LA-ICPMS operation parameters during the analysis of PM filters, NIST glass SRM, and stone surface deposits. Fluence with 10x demagnification was always used.

Parameter	**PM samples**	**NIST glass SRM**	**Stones**
Beam size	128 µm (square mask)	56 µm (square mask)	128 µm (square mask)
Scan speed	256 µm s^{-1}	112 µm s^{-1}	256 µm s^{-1}
Fluence	1.5 J cm^{-2}	4 J cm^{-2}	0.43 J cm^{-2}
Repetition rate	20 Hz	20 Hz	20 Hz
Dosage	10	10	10
Total acquisition time	0.5 s	0.5 s	0.5 s
Ablated area	3 × 12 mm^2	0.168 mm^2	5 × 3.84 mm^2
Carrier gas	Helium; cup flow 0.5 L min^{-1}; cell flow 0.3 L min^{-1}		
Makeup gas	Argon; 0.8 L min^{-1}		

Table S4: Airborne element concentrations in $PM_{2.5}$ from Ljubljana, Slovenia; a) Al, As, Ba, Cd, Co, Cr, Cu, Fe; b) Mn, Mo, Ni, Pb, Rb, Sb, V, Zn.

a) Sample name	^{27}Al [ng m^{-3}]	^{75}As [ng m^{-3}]	^{137}Ba [ng m^{-3}]	^{111}Cd [ng m^{-3}]	^{59}Co [ng m^{-3}]	^{52}Cr [ng m^{-3}]	^{63}Cu [ng m^{-3}]	^{56}Fe [ng m^{-3}]
Summer 2020								
PM2.5(Q)-20200803	33.9	0.031	3.68	0.013	0.0475	0.45	10.84	281
PM2.5(Q)-20200804	21.5	0.026	3.22	0.010	0.0450	0.37	7.87	266
PM2.5(Q)-20200805	19.4	0.035	3.38	0.018	0.0450	0.45	7.99	271
PM2.5(Q)-20200806	42.7	0.093	4.34	0.045	0.0514	0.80	11.20	305
PM2.5(Q)-20200807	125.2	0.227	6.92	0.151	0.0798	2.02	11.64	413
PM2.5(Q)-20200808	143.9	0.264	6.06	0.216	0.0724	1.64	63.39	393
PM2.5(Q)-20200809	162.4	0.281	6.88	0.125	0.0793	2.03	9.73	394
PM2.5(PC)-20200810	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.
PM2.5(Q)-20200811	81.8	0.196	5.37	0.106	0.0664	2.25	20.76	354
PM2.5(Q)-20200812	99.8	0.193	5.29	0.205	0.0686	1.77	41.58	354
PM2.5(Q)-20200813	177.3	0.216	5.80	0.117	0.0813	3.26	15.35	386
PM2.5(Q)-20200814	196.6	0.132	4.59	0.034	0.0690	1.38	8.61	349
PM2.5(Q)-20200815	149.4	0.128	4.93	0.048	0.0609	1.06	42.45	340
PM2.5(Q)-20200816	101.4	0.089	4.58	0.032	0.0556	1.13	14.10	314
PM2.5(Q)-20200817	103.9	0.073	3.99	0.017	0.0573	1.16	8.59	298
Autumn 2020								
PM2.5(Q)-20201109	239.8	1.471	42.0	1.937	0.5570	8.07	97.24	3271
PM2.5(Q)-20201110	45.8	0.142	10.4	0.056	0.1500	1.25	26.71	912
PM2.5(Q)-20201112	33.5	0.153	4.07	0.075	0.0544	2.19	11.51	293
PM2.5(Q)-20201113	104.4	0.324	7.83	0.225	0.1697	28.63	16.43	577
PM2.5(Q)-20201114	89.7	0.396	7.68	0.409	0.0922	7.80	13.67	415
PM2.5(Q)-20201115	29.0	0.165	3.86	0.266	0.0634	3.43	15.19	294
PM2.5(Q)-20201116	37.0	0.137	3.64	0.140	0.0662	4.22	9.17	293
PM2.5(Q)-20201117	19.7	0.075	3.51	0.103	0.0481	1.38	19.29	277
PM2.5(Q)-20201118	24.1	0.154	4.75	0.278	0.0681	7.22	23.97	337
PM2.5(Q)-20201119	20.0	0.190	3.85	0.211	0.0678	7.83	38.30	311
PM2.5(Q)-20201120	16.4	0.061	3.01	0.056	0.0444	0.62	8.05	256

PM2.5(Q)-20201121	21.2	0.090	3.30	0.085	0.0449	0.69	7.97	261
PM2.5(Q)-20201122	60.1	0.407	7.37	0.748	0.1028	6.70	31.94	503
Winter 2021								
PM2.5(Q)-20210215	24.4	0.108	3.52	0.084	0.0494	1.03	8.29	278
PM2.5(Q)-20210216	23.3	0.161	3.84	0.087	0.0558	1.84	8.41	283
PM2.5(Q)-20210217	20.8	0.203	5.13	0.304	0.0662	3.12	20.42	356
PM2.5(Q)-20210218	14.3	0.223	4.78	0.166	0.0666	2.83	9.57	318
PM2.5(Q)-20210219	19.9	0.249	4.50	0.141	0.1121	13.76	10.30	343
PM2.5(Q)-20210220	81.3	0.275	6.08	0.248	0.0888	5.66	13.00	388
PM2.5(Q)-20210221	67.0	0.162	4.28	0.097	0.0633	1.96	8.44	305
PM2.5(Q)-20210222	111.4	0.157	4.58	0.141	0.1748	2.55	8.88	342
PM2.5(Q)-20210223	937.3	0.675	18.57	0.459	0.2134	9.07	21.43	1196
PM2.5(Q)-20210224	2208.9	0.868	23.47	1.214	0.4855	14.59	26.95	1893
PM2.5(Q)-20210225	1914.9	0.811	21.27	1.101	0.4175	11.25	23.07	1687
PM2.5(Q)-20210226	2601.5	0.819	24.63	1.031	0.5400	12.99	27.90	2016
PM2.5(Q)-20210227	103.5	0.158	3.64	0.084	0.0522	1.11	7.83	285
PM2.5(Q)-20210228	13.9	0.453	4.24	0.196	0.0443	1.33	9.55	278
Spring 2021								
PM2.5(Q)-20210524	15.7	0.047	3.67	0.025	0.0771	1.14	8.74	285
PM2.5(Q)-20210525	13.8	0.033	3.44	0.015	0.0485	1.09	8.37	280
PM2.5(Q)-20210526	21.7	0.066	3.75	0.050	0.0503	2.32	11.53	283
PM2.5(Q)-20210527	22.8	0.034	3.51	0.015	0.0491	1.46	8.21	276
PM2.5(Q)-20210528	15.2	0.047	3.48	0.028	0.0473	1.00	8.37	272
PM2.5(Q)-20210529	15.9	0.057	3.87	0.029	0.0459	0.83	9.65	277
PM2.5(Q)-20210530	16.5	0.057	3.55	0.016	0.0472	1.07	8.29	280
PM2.5(Q)-20210531	23.0	0.083	3.78	0.028	0.0484	0.82	9.84	281
PM2.5(Q)-20210601	48.4	0.240	6.80	0.068	0.0601	1.12	9.22	306
PM2.5(Q)-20210602	53.7	0.151	4.42	0.048	0.0527	0.97	8.77	288
PM2.5(Q)-20210603	41.1	0.121	4.66	0.069	0.0533	1.53	16.21	300
PM2.5(Q)-20210604	44.3	0.111	4.37	0.057	0.0533	1.16	12.44	289
PM2.5(Q)-20210605	103.0	0.081	3.97	0.019	0.0549	0.87	7.71	293

PM2.5(Q)-20210606	42.3	0.048	3.36	0.026	0.0473	0.77	7.56	268
				Summer 2021				
PM2.5(Q)-20210621	1199.8	0.368	11.05	0.045	0.2823	4.23	8.29	985
PM2.5(Q)-20210622	697.1	0.250	7.74	0.040	0.1726	2.78	8.35	666
PM2.5(Q)-20210623	1127.2	0.411	11.15	0.112	0.2558	4.42	39.24	947
PM2.5(Q)-20210624	965.9	0.344	9.51	0.397	0.2024	3.33	8.81	749
PM2.5(Q)-20210625	266.7	0.067	130.75	0.042	0.0531	0.87	44.02	275
PM2.5(Q)-20210626	44.4	0.081	4.04	0.025	0.0544	0.91	9.42	290
PM2.5(Q)-20210627	42.9	0.100	4.44	0.032	0.0532	0.99	9.45	298
PM2.5(Q)-20210628	40.3	0.141	4.48	0.041	0.0578	1.31	8.97	311
PM2.5(Q)-20210629	691.7	0.300	8.28	0.058	0.1722	3.41	9.71	653
PM2.5(Q)-20210630	38.5	0.060	3.48	0.018	0.0517	0.86	8.02	278
PM2.5(Q)-20210701	16.2	0.081	3.65	0.028	0.0454	0.80	8.20	272
PM2.5(Q)-20210702	13.5	0.070	3.60	0.032	0.0455	1.04	10.62	273
PM2.5(Q)-20210703	16.1	0.085	3.75	0.028	0.0497	1.31	8.59	278
PM2.5(Q)-20210704	13.6	0.049	3.34	0.026	0.0582	2.20	8.22	270

b)	^{55}Mn	^{95}Mo	^{60}Ni	^{208}Pb	^{85}Rb	^{121}Sb	^{51}V	^{66}Zn
Sample name	[ng m^{-3}]	[ng m^{-3}]	[ng m^{-3}]	[ng m^{-3}]	[ng m^{-3}]	[ng m^{-3}]	[ng m^{-3}]	[ng m^{-3}]
				Summer 2020				
PM2.5(Q)-20200803	0.55	0.206	0.535	1.93	0.156	0.506	0.218	10.2
PM2.5(Q)-20200804	0.39	0.214	0.515	0.29	0.139	0.466	0.194	10.1
PM2.5(Q)-20200805	0.49	0.288	0.525	0.50	0.143	0.486	0.191	10.8
PM2.5(Q)-20200806	1.48	0.270	0.584	5.75	0.193	1.704	0.240	12.5
PM2.5(Q)-20200807	3.62	0.372	0.754	2.53	0.512	0.819	0.458	15.1
PM2.5(Q)-20200808	3.02	0.332	0.713	21.12	0.505	4.291	0.448	17.0
PM2.5(Q)-20200809	3.91	0.316	0.813	4.51	0.551	0.841	0.489	15.9
PM2.5(PC)-20200810	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.
PM2.5(Q)-20200811	3.14	0.473	0.837	43.57	0.398	1.937	0.318	16.2
PM2.5(Q)-20200812	2.94	0.374	0.716	37.44	0.452	1.841	0.366	17.1
PM2.5(Q)-20200813	3.01	0.479	1.073	8.67	0.461	0.936	0.477	15.0

PM2.5(Q)-20200814	2.06	0.292	0.720	1.36	0.346	0.538	0.425	13.0
PM2.5(Q)-20200815	1.63	0.283	0.620	15.37	0.279	1.487	0.344	12.4
PM2.5(Q)-20200816	1.44	0.247	0.661	7.92	0.280	1.197	0.294	11.3
PM2.5(Q)-20200817	1.35	0.255	0.702	1.05	0.258	0.522	0.312	10.3
Autumn 2020								
PM2.5(Q)-20201109	21.431	3.899	7.040	37.505	3.114	8.397	1.960	292.2
PM2.5(Q)-20201110	0.813	0.821	1.861	1.048	0.536	1.409	0.507	33.3
PM2.5(Q)-20201112	2.01	0.358	0.969	3.26	0.305	0.851	0.193	14.0
PM2.5(Q)-20201113	10.66	2.207	12.352	5.57	0.634	1.136	0.350	24.7
PM2.5(Q)-20201114	6.46	1.854	3.722	9.46	0.785	1.087	0.378	28.4
PM2.5(Q)-20201115	2.41	0.797	2.090	7.54	0.543	0.954	0.228	23.0
PM2.5(Q)-20201116	3.16	1.273	2.210	6.65	0.290	0.548	0.214	27.4
PM2.5(Q)-20201117	2.61	0.397	0.654	4.48	0.339	5.785	0.159	16.1
PM2.5(Q)-20201118	4.03	0.693	2.015	9.98	0.738	8.089	0.187	19.7
PM2.5(Q)-20201119	4.55	0.620	2.456	29.24	0.544	4.392	0.161	23.0
PM2.5(Q)-20201120	1.32	0.284	0.549	0.82	0.248	0.451	0.149	14.1
PM2.5(Q)-20201121	1.16	0.278	0.539	1.09	0.424	0.484	0.166	12.5
PM2.5(Q)-20201122	8.82	1.839	1.968	15.60	2.024	4.730	0.298	38.1
Winter 2021								
PM2.5(Q)-20210215	2.20	0.390	0.638	1.60	0.504	0.555	0.213	17.6
PM2.5(Q)-20210216	2.66	0.745	0.893	2.92	0.350	0.656	0.281	22.7
PM2.5(Q)-20210217	4.18	0.738	0.913	36.58	1.080	2.326	0.242	32.5
PM2.5(Q)-20210218	5.97	0.963	1.074	6.98	0.434	0.943	0.270	34.9
PM2.5(Q)-20210219	7.41	2.075	4.651	4.98	0.348	0.930	0.284	29.7
PM2.5(Q)-20210220	6.19	1.784	2.310	6.57	0.908	2.385	0.443	33.3
PM2.5(Q)-20210221	3.56	0.655	0.994	4.19	0.385	0.713	0.428	24.2
PM2.5(Q)-20210222	3.62	0.537	1.178	5.08	0.527	0.927	0.517	27.8
PM2.5(Q)-20210223	15.19	1.737	1.839	20.66	3.445	1.933	1.550	29.8
PM2.5(Q)-20210224	39.05	2.225	3.633	11.02	6.065	3.177	3.095	81.6
PM2.5(Q)-20210225	32.04	2.234	2.823	16.91	5.394	2.855	2.744	78.6
PM2.5(Q)-20210226	40.31	2.017	3.134	15.86	6.032	1.804	3.715	71.5

PM2.5(Q)-20210227	1.24	0.237	0.629	1.68	0.383	0.668	0.290	14.0
PM2.5(Q)-20210228	1.22	0.366	0.630	5.81	0.652	0.795	0.193	21.5
Spring 2021								
PM2.5(Q)-20210524	1.07	0.331	0.700	0.79	0.197	0.599	0.169	11.1
PM2.5(Q)-20210525	0.67	0.344	0.745	0.48	0.159	0.545	0.175	11.1
PM2.5(Q)-20210526	1.69	0.324	0.860	2.73	0.186	1.460	0.193	11.2
PM2.5(Q)-20210527	0.79	0.285	0.836	0.51	0.158	0.553	0.183	9.9
PM2.5(Q)-20210528	0.69	0.472	0.802	1.17	0.163	0.588	0.174	11.5
PM2.5(Q)-20210529	0.74	0.278	0.608	2.03	0.174	0.665	0.176	11.8
PM2.5(Q)-20210530	0.67	0.320	0.755	0.67	0.158	0.622	0.188	10.0
PM2.5(Q)-20210531	0.96	0.286	0.636	0.82	0.172	1.234	0.215	11.1
PM2.5(Q)-20210601	1.77	0.342	0.631	2.94	0.255	0.642	0.296	13.6
PM2.5(Q)-20210602	1.35	0.266	0.617	3.08	0.252	0.662	0.269	12.6
PM2.5(Q)-20210603	1.55	0.325	0.741	4.61	0.229	1.088	0.228	11.9
PM2.5(Q)-20210604	1.59	0.370	0.765	5.83	0.220	1.007	0.251	13.3
PM2.5(Q)-20210605	1.07	0.268	0.682	1.04	0.238	0.516	0.331	10.0
PM2.5(Q)-20210606	0.77	0.294	0.566	1.13	0.163	0.484	0.196	12.1
Summer 2021								
PM2.5(Q)-20210621	13.44	0.284	1.378	1.88	1.947	0.554	2.588	14.4
PM2.5(Q)-20210622	7.83	0.290	1.106	1.92	1.101	0.781	1.617	13.9
PM2.5(Q)-20210623	12.70	0.434	1.450	13.22	1.780	8.706	2.375	20.1
PM2.5(Q)-20210624	9.44	0.357	1.245	2.45	1.293	0.769	1.978	16.2
PM2.5(Q)-20210625	0.90	0.293	0.702	2.96	0.263	0.915	0.252	11.5
PM2.5(Q)-20210626	1.29	0.306	0.636	1.50	0.219	0.743	0.236	10.7
PM2.5(Q)-20210627	1.19	0.320	0.645	3.65	0.205	0.688	0.236	11.1
PM2.5(Q)-20210628	2.73	0.425	0.760	0.99	0.214	0.758	0.316	12.9
PM2.5(Q)-20210629	7.59	0.525	1.244	2.01	1.146	1.014	1.647	13.8
PM2.5(Q)-20210630	0.90	0.316	0.645	0.48	0.188	0.532	0.243	10.6
PM2.5(Q)-20210701	0.87	0.289	0.607	1.33	0.148	0.647	0.171	14.4
PM2.5(Q)-20210702	0.76	0.327	0.623	3.47	0.151	1.629	0.169	12.5
PM2.5(Q)-20210703	1.37	0.363	0.831	1.30	0.149	0.736	0.211	12.1

PM2.5(Q)-20210704	1.18	0.605	1.472	1.04	0.132	0.557	0.174	11.9

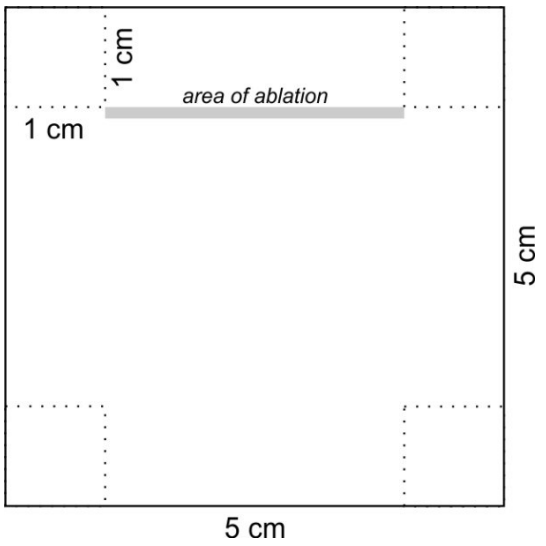


Figure S3: Schematic representation of the laser-ablation area on stone samples.

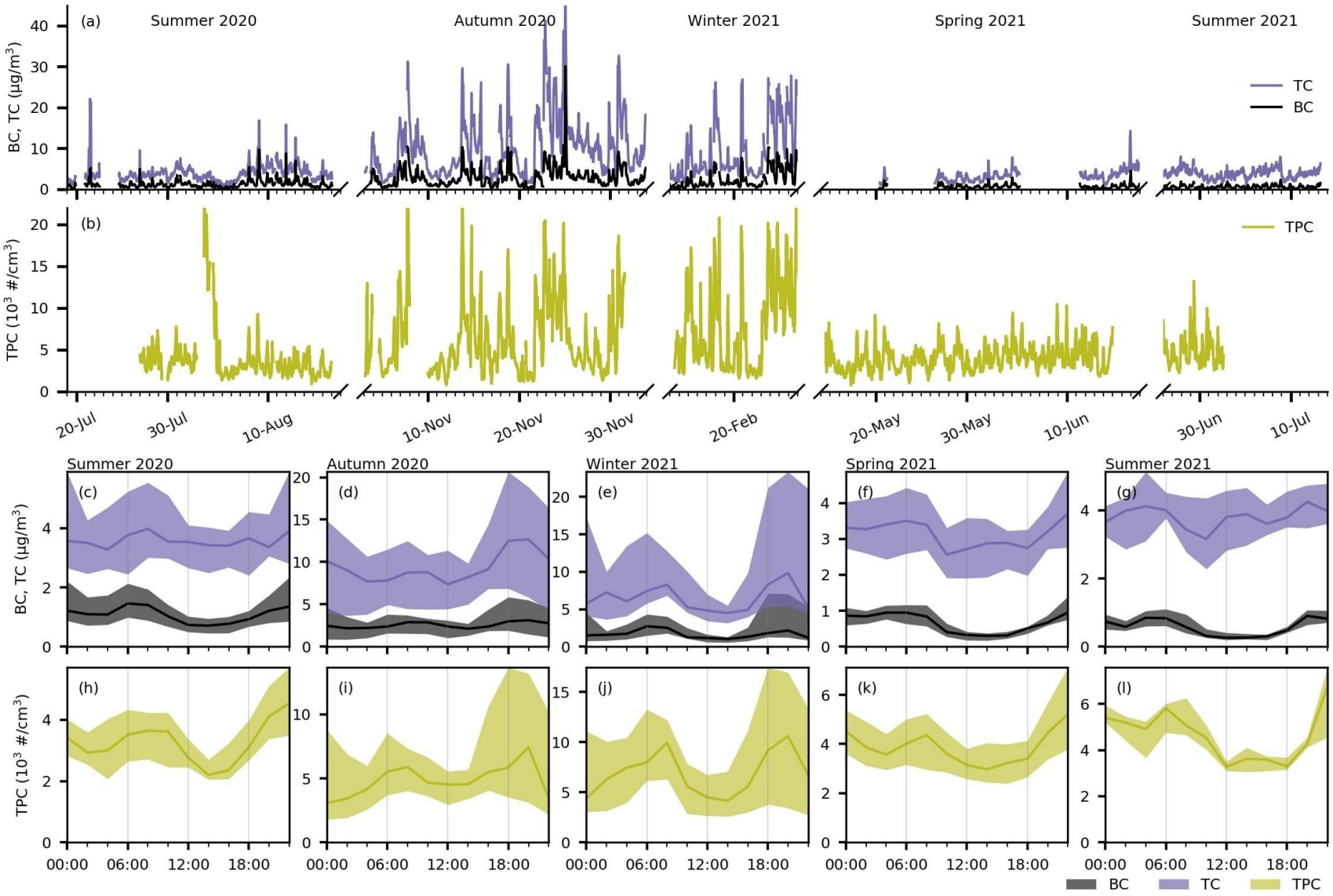


Figure S4: Time series of a) total carbon (TC) and black carbon (BC), and b) total particle concentration (TPC, 14–750 nm) in Ljubljana, Slovenia in different seasons from July 2020 to July 2021; c–l) mean diurnal profiles in different seasons: median, Q1 and Q3.

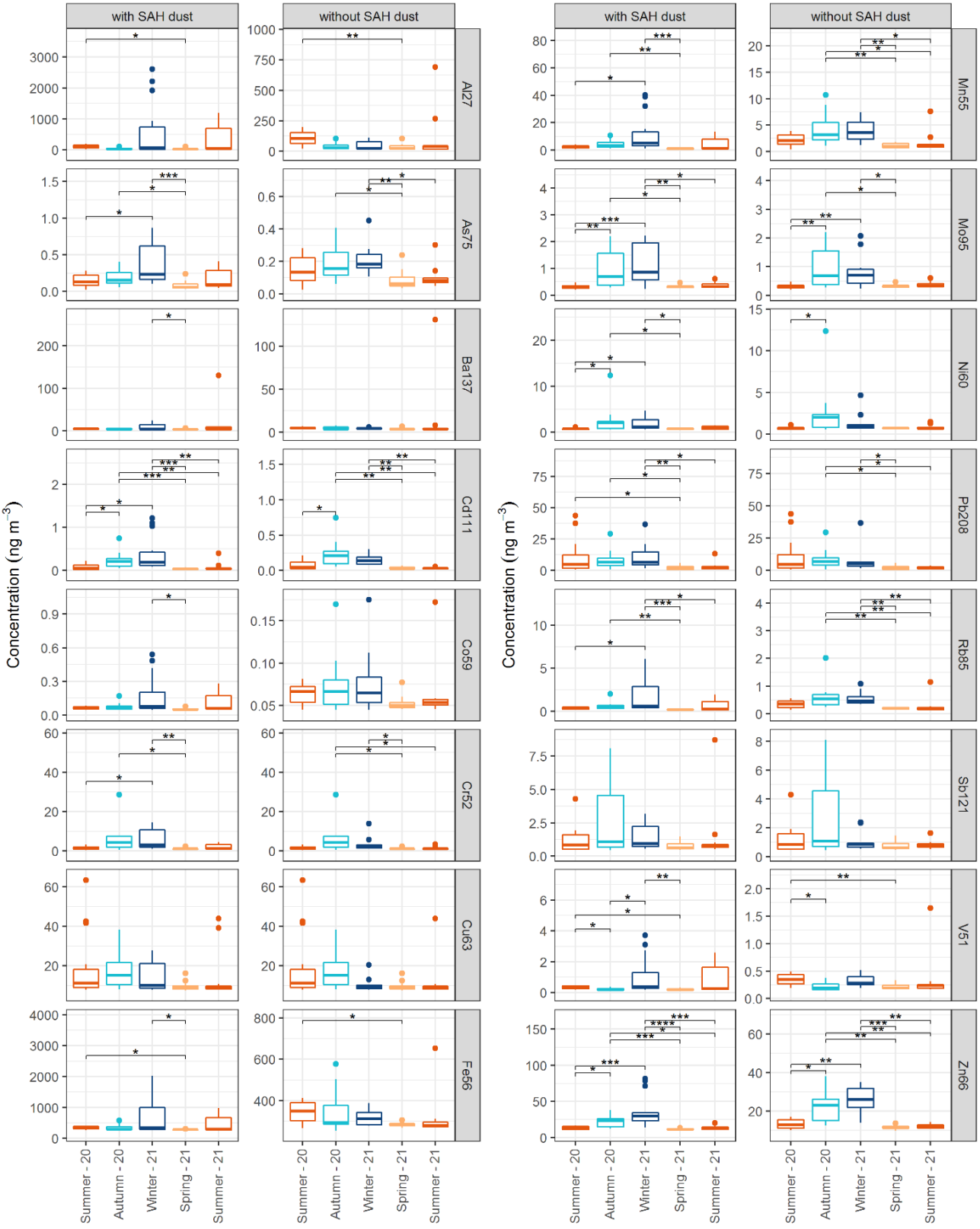


Figure S5: Airborne element concentrations in $PM_{2.5}$ from Ljubljana, Slovenia including Saharan dust events and without Saharan dust days for five seasonal campaigns conducted between July 2020 and July 2021. Statistically significant differences between campaigns ($p<0.05$) are denoted with asterisks (the more asterisks the more significant the difference).

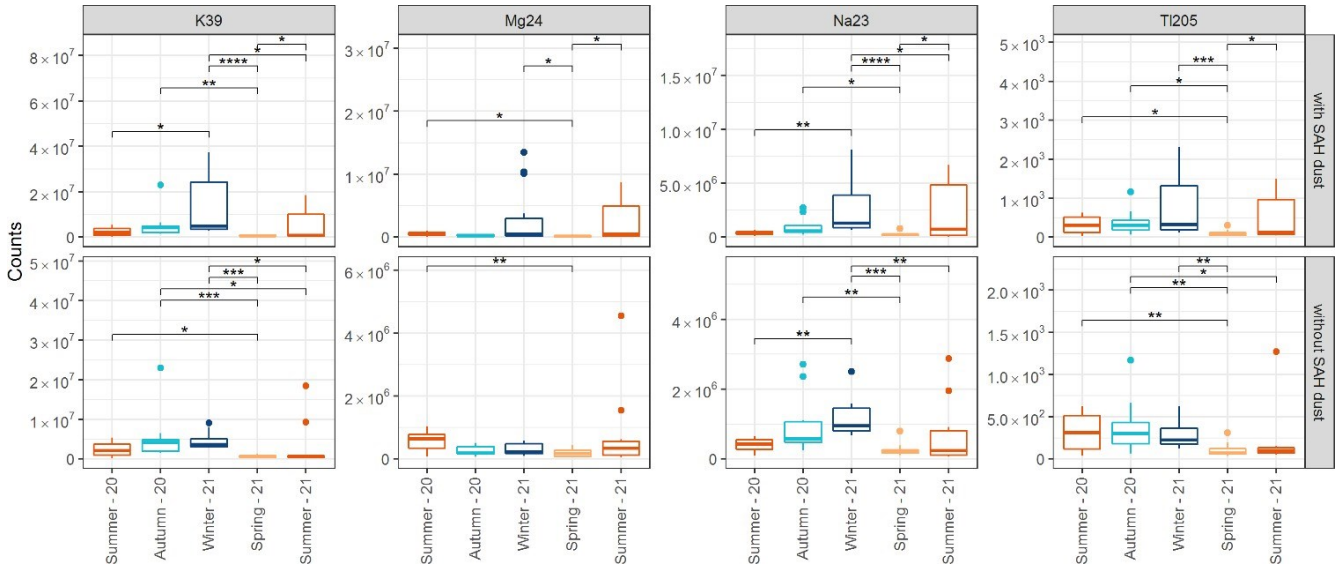


Figure S6: Elemental composition of $PM_{2.5}$ from Ljubljana, Slovenia including Saharan dust events and without Saharan dust days for unquantified elements for five seasonal campaigns conducted between July 2020 and July 2021. Statistically significant differences between campaigns ($p<0.05$) are denoted with asterisks (the more asterisks the more significant the difference).

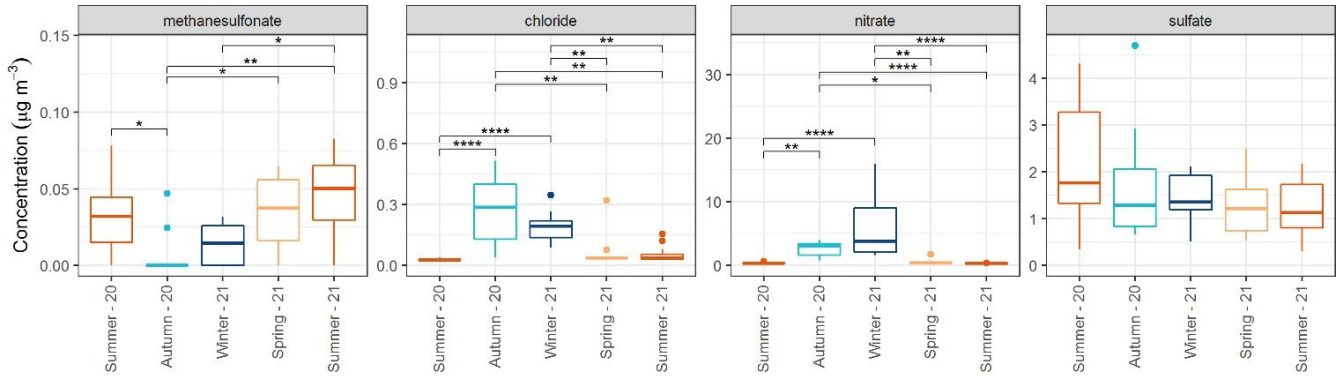


Figure S7: Airborne concentrations of selected anionic species in $PM_{2.5}$ from Ljubljana, Slovenia in different seasons from July 2020 until July 2021: methanesulfonate (MS), chloride, nitrate, and sulphate. Statistically significant differences between campaigns ($p<0.05$) are denoted with asterisks (the more asterisks the more significant the difference).

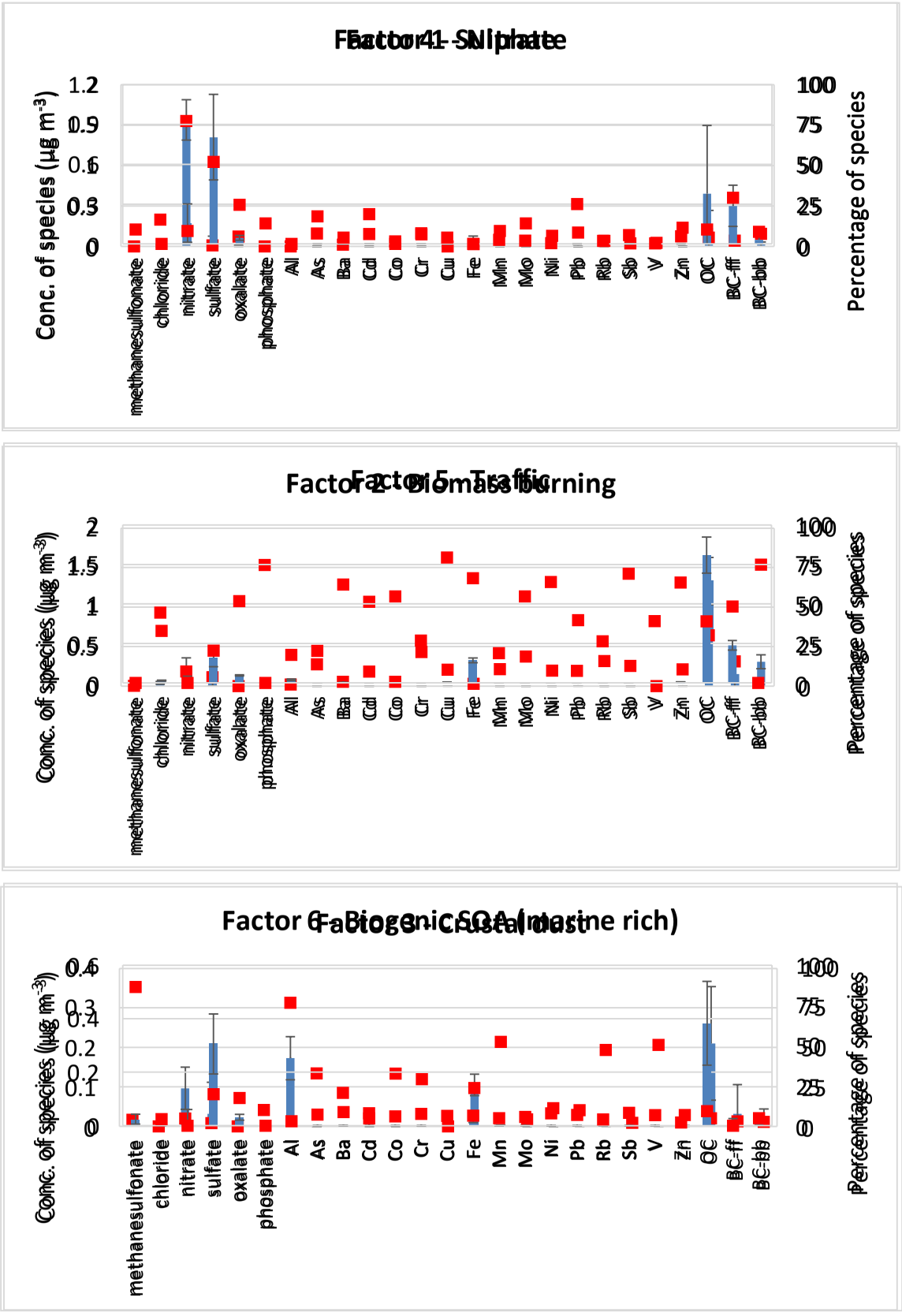
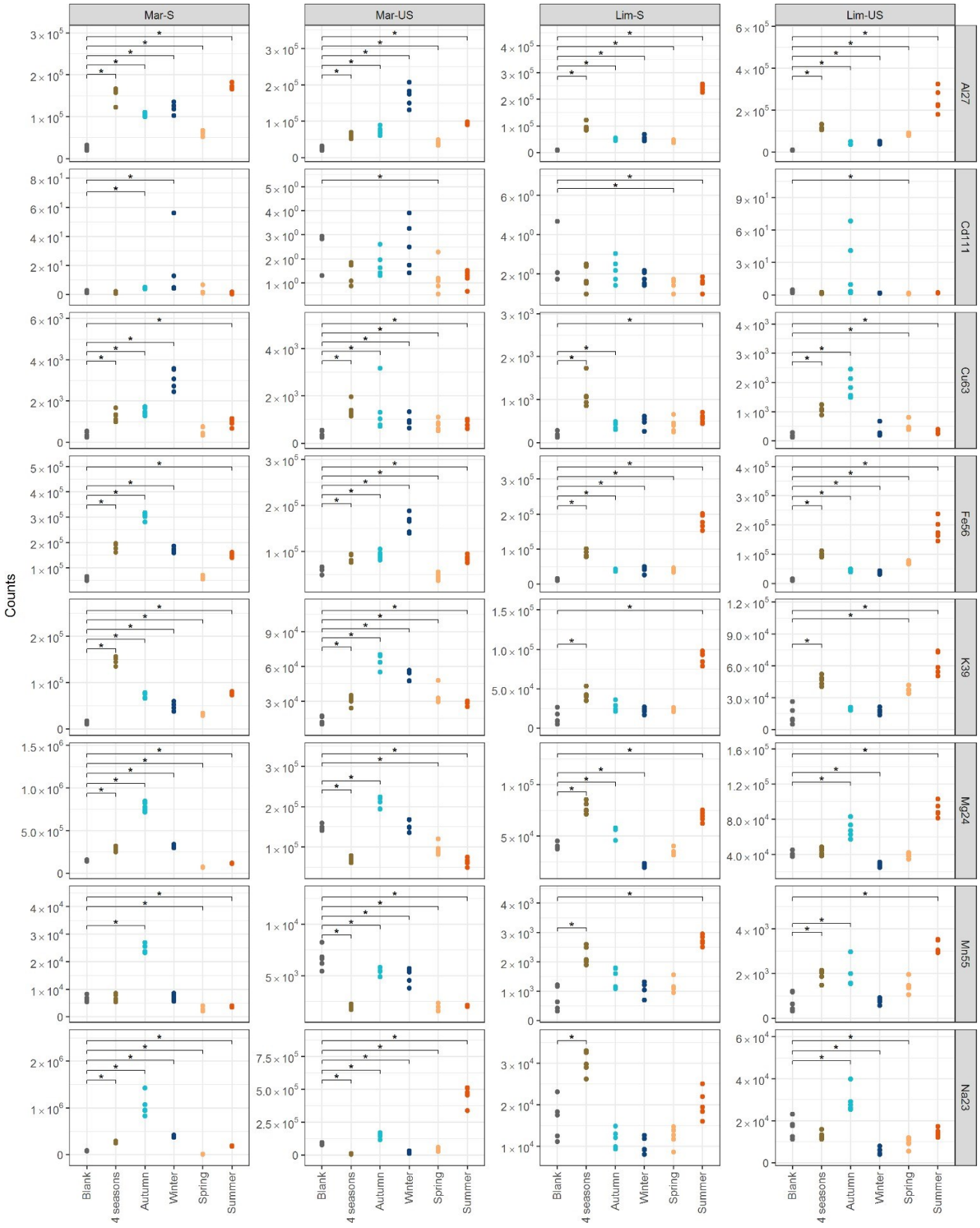


Figure S8: Profiles of the factors corresponding to specific PM sources as obtained with the PMF analysis of acquired data for seasonal $PM_{2.5}$ from Ljubljana, Slovenia. Absolute concentrations are depicted with columns and percent contributions with red squares.


Mar-S
Mar-US
Lim-S
Lim-US
Al27
Cd111
Cu63
Fe56
K39
Mg24
Mn55
Na23
Counts
Blank
4 seasons
Autumn
Winter
Spring
Summer

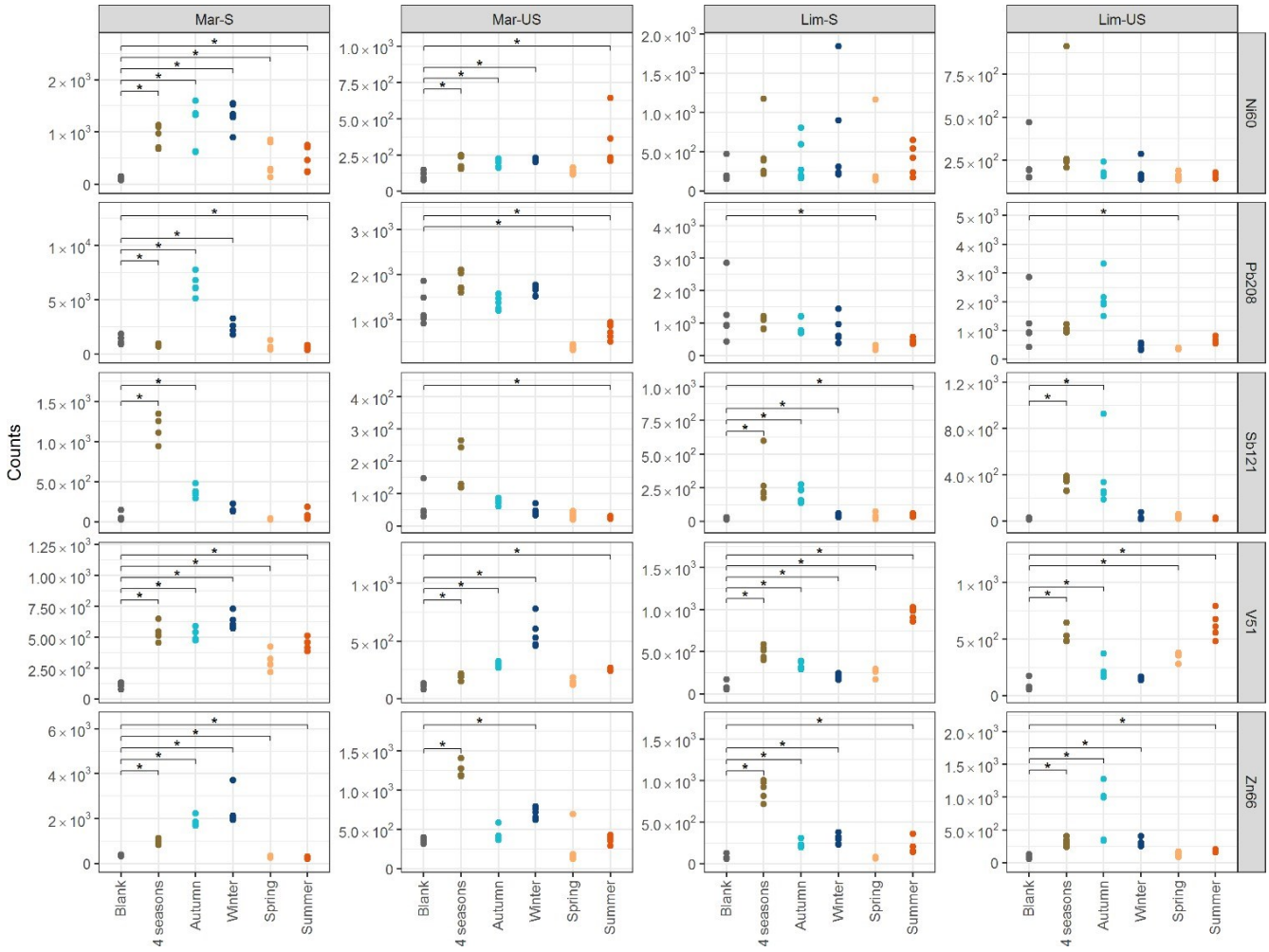


Figure S9: Comparison of marble (Mar) and limestone (Lim) samples, sheltered (S) and unsheltered (US), exposed in five seasonal campaigns from July 2020 until July 2021, with blanks. Statistically significant differences between blank and exposed samples ($p<0.05$) are denoted with asterisks (the more asterisks the more significant the difference); ns stands for nonsignificant.

Table S5: Comparison between sheltered and unsheltered marble (Mar) and limestone (Lim) samples exposed in five seasonal campaigns: C1 – four-season exposure, C2 – autumn 2020, C3 – winter 2021, C4 – spring 2021, C5 – summer 2021. In brackets, adjusted p-values according to Benjamini & Hochberg are reported; cases with no evidence of statistical deference (p>0.05) are bolded.

		Al27	Cd111	Cu63	Fe56	K39	Mg24	Mn55	Na23	Ni60	Pb208	Sb121	V51	Zn66
Mar	C1	0.008	0.750	0.690	0.008	0.008	0.008	0.008	0.008	0.008	0.008	0.008	0.008	0.008
		(0.009)	**(0.750)**	**(0.748)**	(0.009)	(0.009)	(0.009)	(0.009)	(0.009)	(0.009)	(0.009)	(0.009)	(0.009)	(0.009)
	C2	0.008	0.008	0.222	0.008	0.222	0.008	0.008	0.008	0.008	0.008	0.008	0.008	0.008
		(0.009)	(0.009)	**(0.222)**	(0.009)	**(0.222)**	(0.009)	(0.009)	(0.009)	(0.009)	(0.009)	(0.009)	(0.009)	(0.009)
	C3	0.016	0.016	0.008	0.548	0.310	0.008	0.016	0.008	0.008	0.016	0.008	0.310	0.008
		(0.021)	(0.021)	(0.017)	**(0.548)**	**(0.336)**	(0.017)	(0.021)	(0.017)	(0.017)	(0.021)	(0.017)	**(0.336)**	(0.017)
	C4	0.008	0.248	0.095	0.008	0.421	0.008	0.016	0.008	0.056	0.016	0.841	0.008	0.151
		(0.021)	**(0.293)**	**(0.138)**	(0.021)	**(0.456)**	(0.021)	(0.030)	(0.021)	(0.090)	(0.030)	**(0.841)**	(0.021)	**(0.196)**
	C5	0.008	0.600	0.310	0.008	0.008	0.008	0.008	0.008	0.222	0.421	0.008	0.008	0.016
		(0.013)	**(0.600)**	**(0.366)**	(0.013)	(0.013)	(0.013)	(0.013)	(0.013)	**(0.289)**	**(0.456)**	(0.013)	(0.013)	(0.023)
Lim	C1	0.095	0.916	0.841	0.095	0.421	0.008	0.421	0.008	0.421	1.000	0.310	0.690	0.008
		(0.248)	**(0.992)**	**(0.992)**	**(0.248)**	**(0.608)**	(0.034)	**(0.608)**	(0.034)	**(0.608)**	**(1.000)**	**(0.608)**	**(0.897)**	(0.034)
	C2	0.056	0.056	0.008	0.032	0.008	0.032	0.548	0.008	0.151	0.008	0.222	0.056	0.008
		(0.072)	(0.072)	(0.021)	(0.059)	(0.021)	(0.059)	**(0.548)**	(0.021)	**(0.178)**	(0.021)	**(0.241)**	(0.072)	(0.021)
	C3	0.095	0.709	0.286	0.310	0.095	0.008	0.421	0.008	0.032	0.095	0.151	0.032	0.690
		(0.177)	**(0.709)**	**(0.403)**	**(0.403)**	**(0.177)**	**(0.052)**	**(0.498)**	**(0.052)**	**(0.103)**	**(0.177)**	**(0.245)**	**(0.103)**	**(0.709)**
	C4	0.008	0.671	0.421	0.008	0.008	0.056	0.310	0.151	0.310	0.008	0.841	0.016	0.008
		(0.021)	**(0.727)**	**(0.498)**	(0.021)	(0.021)	**(0.103)**	**(0.403)**	**(0.245)**	**(0.403)**	(0.021)	**(0.841)**	(0.034)	(0.021)
	C5	0.841	0.512	0.008	1.000	0.008	0.008	0.016	0.016	0.032	0.016	0.008	0.008	0.690
		(0.911)	**(0.666)**	(0.021)	**(1.000)**	(0.021)	(0.021)	(0.026)	(0.026)	(0.046)	(0.026)	(0.021)	(0.021)	**(0.815)**


Mar-S
Mar-US
Lim-S
Lim-US
Al27
Cd111
Cu63
Fe56
K39
Mg24
Mn55
Na23
Counts
4 seasons
Autumn
Winter
Spring
Summer

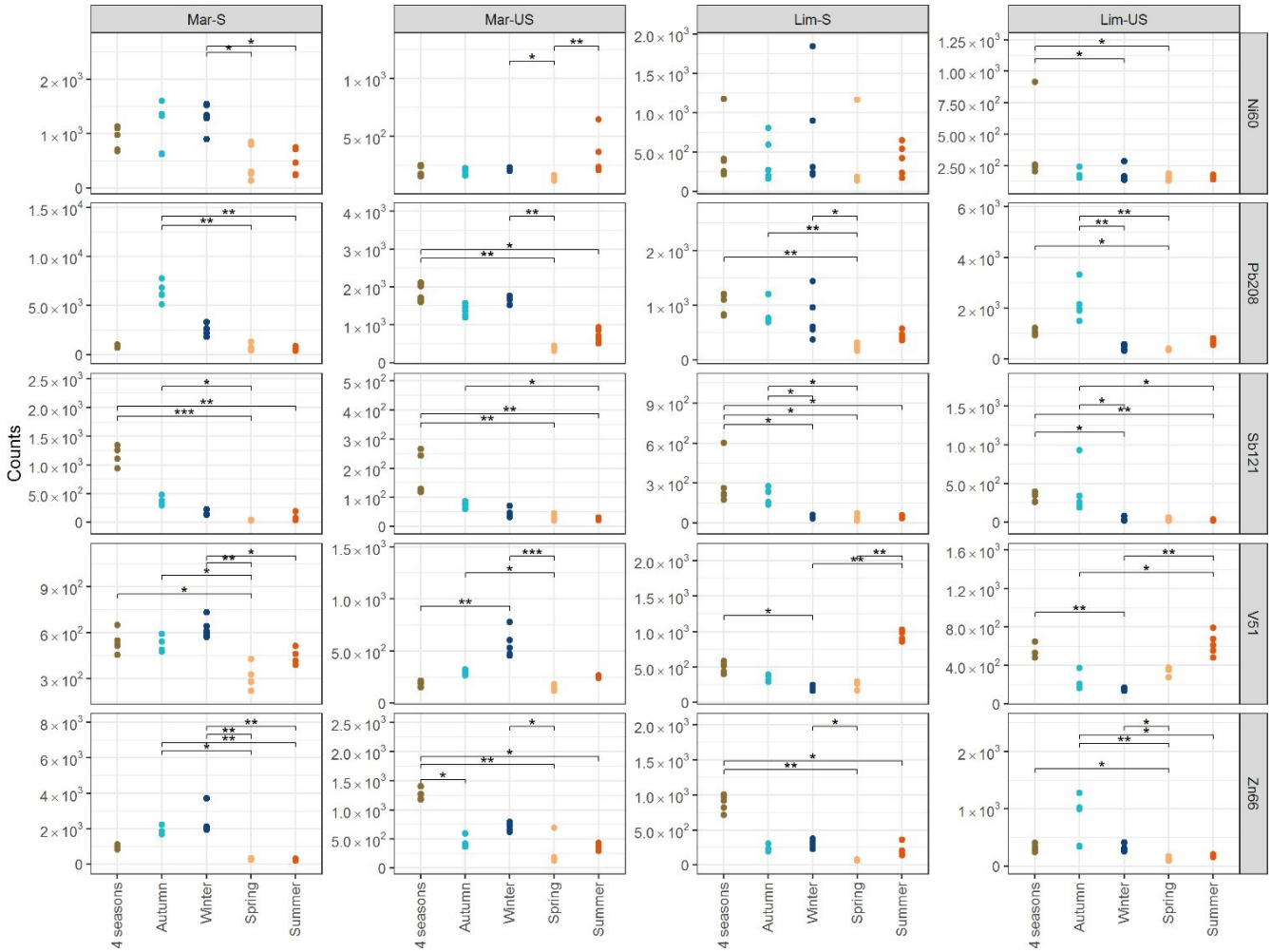


Figure S10: Stone surface elemental profiles for marble (Mar) and limestone (Lim) samples, sheltered (S) and unsheltered (US), exposed in five seasonal campaigns from July 2020 until July 2021. Statistically significant differences between the exposed samples ($p<0.05$) are denoted with asterisks (the more asterisks the more significant the difference).

Table S6: Colour data from measured reflectance for marble (Mar) and limestone (Lim) samples before and after the exposure, sheltered (S) and unsheltered (US), in four seasonal campaigns: C2 – autumn 2020, C3 – winter 2020, C4 – spring 2021, C5 – summer 2021. A unit change of every parameter is considered significant.

	Sample name	Exposure days	L^*	a^*	b^*	C^*	ΔE^*	ΔL^*
Mar	C2-US	0	72.6	-1.3	-2.5	2.8	2.6	2.6
		31	75.2	-1.3	-2.4	2.7		
	C2-S	0	72.6	-1.3	-2.5	2.8	0.3	0.0
		31	72.6	-1.1	-2.3	2.5		
	C3-US	0	75.4	-1.2	-2.4	2.7	0.5	0.5
		29	75.9	-1.3	-2.6	2.9		
	C3-S	0	73.0	-1.3	-2.7	3.0	0.4	0.0
		29	73.0	-1.4	-3.1	3.4		
	C4-US	0	72.7	-1.2	-2.4	2.7		

		35	74.5	-1.1	-2.5	2.7	1.7	1.7
	C4-S	0	71.4	-1.3	-2.8	3.1	0.3	-0.3
		35	71.1	-1.2	-2.7	3.0		
	C5-US	0	72.9	-1.0	-2.0	2.2	1.2	1.2
		30	74.1	-1.1	-2.3	2.5		
	C5-S	0	73.0	-1.2	-2.1	2.4	1.7	1.6
		30	74.6	-1.2	-2.4	2.7		
Lim	C2-US	0	40.5	-0.9	-3.2	3.4	3.8	3.8
		31	44.3	-0.8	-2.9	3.0		
	C2-S	0	37.3	-0.9	-3.3	3.4	4.2	4.1
		31	41.4	-0.7	-2.6	2.6		
	C3-US	0	42.4	-0.7	-3.0	3.1	3.3	3.3
		29	45.6	-0.5	-2.4	2.5		
	C3-S	0	40.0	-0.7	-3.1	3.2	3.7	3.6
		29	43.6	-0.6	-2.5	2.6		
	C4-US	0	43.3	-0.9	-3.4	3.5	5.7	5.7
		35	49.0	-0.8	-3.7	3.8		
	C4-S	0	42.0	-0.9	-3.4	3.5	0.2	0.0
		35	42.0	-0.7	-3.3	3.4		
	C5-US	0	40.8	-0.8	-3.5	3.6	3.8	3.8
		30	44.6	-0.7	-3.9	4.0		
	C5-S	0	42.6	-0.9	-3.3	3.4	4.0	4.0
		30	46.5	-0.9	-3.7	3.8		

[a]Chromaticity (C) is calculated as $C^* = \sqrt{a^{*2} + b^{*2}}$

[b]Colour difference formula: $\Delta E^* = \sqrt{(L^*_2 - L^*_1)^2 + (a^*_2 - a^*_1)^2 + (b^*_2 - b^*_1)^2}$

Table S7: Colour data from measured reflectance for marble (Mar) and limestone (Lim) samples, sheltered (S) and unsheltered (US), exposed in all four seasonal campaigns from July 2020 until June 2021. A unit change of every parameter is considered significant.

	Exposure days	L^*	a^*	b^*	C^*	ΔE^*	ΔL^*
Mar-US	0	73.0	-1.3	-2.3	2.6		
	36	77.9	-1.3	-2.4	2.7	3.2	4.9
	67	82.1	-1.1	-1.5	1.9	7.4	9.2
	96	79.3	-1.3	-2.2	2.5	4.6	6.3
	131	78.7	-1.3	-2.1	2.5	3.9	5.7
Mar-S	0	73.0	-1.3	-2.3	2.6		
	36	75.8	-1.1	-2.3	2.6	1.1	2.9
	67	77.2	-1.2	-2.4	2.7	2.5	4.2
	96	74.6	-1.2	-2.8	3.0	0.5	1.7
	131	73.4	-1.1	-2.6	2.8	1.4	0.4
Lim-US	0	41.1	-0.9	-3.1	3.2		
	36	44.0	-0.7	-3.6	3.7	2.9	2.9
	67	43.9	-0.7	-3.8	3.8	3.1	2.7
	96	42.0	-0.8	-4.4	4.5	5.1	0.9
	131	42.9	-0.9	-5.7	5.8	4.8	1.7
Lim-S	0	41.1	-0.9	-3.1	3.2		

36	43.0	-0.9	-3.7	3.8	3.9	1.9
67	42.9	-0.8	-3.2	3.3	4.0	1.8
96	44.3	-1.0	-4.3	4.4	2.9	3.2
131	45.0	-1.0	-4.8	4.8	2.6	3.8