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Metal and metalloid content in real urban synthetic surfaces made of recycled tire crumb rubber including playgrounds and football fields

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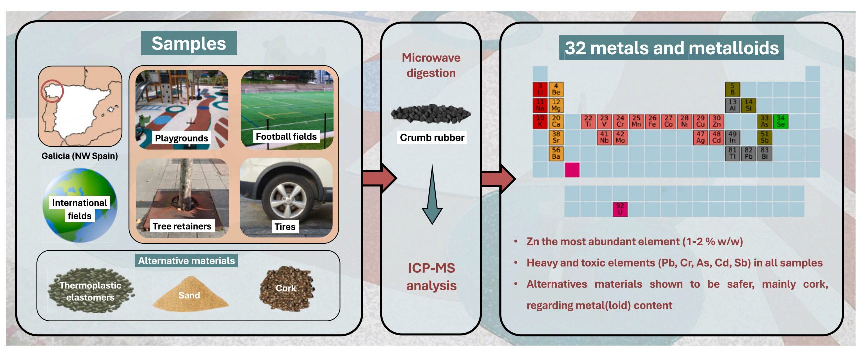
HIGHLIGHTS

- 32 elements in 29 tire crumb rubber facilities (playgrounds and football fields)
- Zn, the most abundant element (1–2 %), above the safety limits for related matrices
- Heavy, toxic elements (Pb, Cr, As, Cd, Sb) in all samples (Pb, Sb up to 100 mg kg^{-1})
- ANOVA shows statistical difference among crumb rubber sample groups for some elements
- Alternative materials shown to be safer, mainly cork, regarding metal(loid) content

GRAPHICAL ABSTRACT



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ABSTRACT

The disposal of end-of-life tires (ELTs) is an important issue in the context of solid waste management. In the last decades, the main recycling route consists of the ELTs transformation in crumb rubber, which is widely used worldwide in playgrounds and sports fields as infill material. Crumb rubber represents the largest source of intentional microplastics in the environment. This microplastic material contains high metal concentration including toxic and heavy metals. Few studies deal with the metal(loid) characterization of real crumb rubber samples taken *in situ* from sports and leisure facilities. Research is especially scarce for playgrounds, despite interest due to the population using these facilities (children).

This study aims at addressing the metal(loid) distribution in a large number of real samples from different urban places, most from Galicia (NW Spain) but also from other countries. 32 elements including metals (Ag, Al, Ba, Be, Bi, Ca, Cd, Co, Cr, Cu, Fe, In, K, Li, Mg, Mn, Mo, Na, Nb, Ni, Pb, Se, Sr, Ti, Tl, U, V, Zn) and 4 metalloids (As, B, Sb, Si) were determined. For comparison purposes, some alternative materials (cork, sand, and thermoplastic elastomers) were collected. The results showed high Zn levels (1–2 %) in crumb rubber, exceeding the safety limits set in the European directives for related matrices. Heavy and toxic elements (Pb, Cr, As, Cd, Sb) were found in all samples, reaching concentrations up to 100 mg kg^{-1}. Co presented concentrations of 200 mg kg^{-1}, well above the safety limits (10 mg kg^{-1} for toys). ANOVA showed statistical differences between

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playgrounds and football fields for some elements. The alternative materials proved safer regarding metal(loid) content. This study is the largest one about metal(loid) characterization in crumb rubber surfaces attending the number of samples, origin, and elements analyzed.

1. Introduction

Crumb rubber infill of artificial turf fields and other facilities made from tire crumb rubber (derived from shredded end-of-life automobile tires) represents one of the largest individual contributors to microplastics at ≈ 38 % of the total estimated release from intentionally added products into the environment (European Chemicals Agency (ECHA), n. d.).

Discarded tires pose an important problem in solid waste management. In the last decades, the main recycling route for end-of-life tires (ELTs) has been their transformation in crumb rubber (CR) which is largely used as flooring material in playground and as infill in synthetic turf sports fields. This microplastic material is a complex chemical mixture that contains hundreds of organic compounds and many elements including toxic and heavy metals (Gomes et al., 2021; Graça et al., 2022; Celeiro et al., 2018). The new EU regulation 2023/2055 bans the use of granular infill in synthetic sports surfaces but these kind of facilities will still survive for at least a transitional period (8 years) (Commission Regulation (EU), 2023).

Previous studies report the occurrence of metals in crumb rubber at relevant concentrations (Bocca et al., 2009; Marsili et al., 2015; Canepari et al., 2018; Han et al., 2008; Menichini et al., 2011; Pavilonis et al., 2014; Pronk et al., 2020; Celeiro et al., 2018). These studies usually refer to a specific site or region, excluding Graça et al. (Graça et al., 2022) which compares metal concentrations in turf pitches from different countries.

In general, the concentration ranges are very different depending on the element. In those studies where Zn was included, it was the element at the highest concentration reaching values at the low percentage, above the safety limits for related matrices such as soils (Council Directive 86/278/EEC, 1986) and toys (Directive 2009/48/EC, 2009). Table S1 contains the values of these safety limits. As regards crumb rubber, the only current regulation is the recent restrictions on PAH content (Commission Regulation (EU), 2021; Celeiro et al., 2021; Zuccaro et al., 2022) and on the uses of this microplastic material (Commission Regulation (EU), 2023).

Bocca et al. (Bocca et al., 2009) evaluated the presence of metals in crumb rubber and water leachates of a synthetic turf field located in Italy and found very high concentrations of Zn in both matrices. Celeiro et al. (Celeiro et al., 2018) investigate two commercial crumb rubber samples underlining the very high concentrations of Zn. Other elements found at high concentrations in the CR were Fe, Mg, Cu and Co. An important metal is Al despite the little information available about its concentration in this type of material. Schneider et al., (Schneider et al., 2020) found values between 1000 and 5400 mg kg^{-1}. Only three other metals were included in this study. Graça et al. (Graça et al., 2022) studied crumb rubber from worldwide football fields, concluding that no remarkable difference could be associated to the geographical origin.

One controversial element that appears at high levels is Si, although it is hardly studied in CR facilities. Its presence is due to the use of silica nanoparticles to reinforce rubber during tire fabrication. The presence of relevant elements, in terms of their known toxicity at trace levels, such as Cd, Cr and Pb, is also described in the literature.

Regarding tire crumb rubber from playgrounds, the number of studies is more limited and there is not much information about the metal content. A recent study (Moreno et al., 2023) including pristine (unused) rubber materials that could be installed in playing surfaces, also confirmed very high concentrations of Zn (2 %) in the 3 tire crumb rubber samples tested (lower levels in the other material, EPDM - ethylene propylene diene monomer). In addition, the authors observed associations among colors and the levels of certain elements such as Fe and Cr. Another recent study (Fawkes et al., 2021) assessed the presence and concentration of fourteen metals from parks in the Bryan-College Station Metropolitan area in Texas ($N = 16$) finding As, Ba, Br, Cd, Cu, Fe, Pb, Ti and Zn. Although the levels of these metals were below the guideline limits set by the United States Consumer Product Safety Commission, they concluded that additional research is required to further assess the safety of parks and playgrounds.

As it was already highlighted, there are only few studies dealing with the analysis of a high number of metals and metalloids in real crumb rubber samples collected from football fields, and even less studies from playgrounds. Not any study includes other minor uses for crumb rubber, such as tree root retainers. In addition, it is necessary to search for alternatives that could replace this material that is already forbidden (Commission Regulation (EU), 2023), which could be safer regarding their toxic element content.

The aim of this study is to evaluate the metal and metalloid content of a wide range of recycle tire crumb rubber samples from playgrounds, football fields and other urban surfaces mainly from Galicia (NW Spain), but also in samples from other countries, to show the widespread use of this material in surfaces and the global dimension of the problem. Some synthetic and natural substitute materials to crumb rubber already used in playgrounds and synthetic turf football fields (cork, sand, and thermoplastic elastomer pellets) were also collected. After being analyzed, the results were compared with CR concentrations. The samples were subjected to microwave digestion followed by ICP-MS for the analysis of 32 elements including 28 metals (Ag, Al, Ba, Be, Bi, Ca, Cd, Co, Cr, Cu, Fe, In, K, Li, Mg, Mn, Mo, Na, Nb, Ni, Pb, Se, Sr, Ti, Tl, U, V, and Zn) and 4 metalloids (As, B, Sb, Si). The results show high element concentrations in several cases as well as the presence of well-known toxic heavy metals at trace levels. As expected, the Zn levels were very high and above the safe limits set in European directives for soils and toy materials and the same happened for Pb, although at lower concentrations, surpassing the toy limit in some samples. The data were subjected to ANOVA analysis showing, in some cases, statistical differences among the different groups of samples such as playgrounds and football fields.

2. Materials and methods

2.1. Reagents and materials

Nitric acid (65 % Hiperpur) was purchased from Panreac (Spain). Dilutions and mixtures were prepared in Nitric acid (6 %). All reagents and solutions were of Suprapur grade.

32 elements including 28 metals (Ag, Al, Ba, Be, Bi, Ca, Cd, Co, Cr, Cu, Fe, In, K, Li, Mg, Mn, Mo, Na, Nb, Ni, Pb, Se, Sr, Ti, Tl, U, V, and Zn) and 4 metalloids (As, B, Sb, Si) were considered. Calibration standards of each element (1000 mg L^{-1}) (Merck, Germany) were employed for instrument calibration after diluting with HNO_3 (6 % *v*/v). Ultrapure water (18 MΩ cm resistivity) used for the dilution of all solutions was prepared by the Milli-Q Element water system. All glassware and materials used in the procedure were soaked in nitric acid (20 % v/v) and then exhaustively washed with ultrapure water.

2.2. Samples

Thirty-four samples were analyzed in this work. Crumb rubber samples were collected from 10 playgrounds (PGs) and 9 synthetic turf football fields (FFs) from Santiago de Compostela and other nearby locations in Galicia (Northwest of Spain). All the football fields are periodically replenished with fresh crumb rubber since part of the material is daily lost because of the use and the meteorological conditions. Some urban surfaces were also collected: tree root retainers (TRs) ($n = 3$), one commercial crumb rubber tile (TILE), and a discarded tire (TIRE). In addition, some alternative surface and infill materials were collected from playgrounds and football fields: sand ($n = 1$), cork ($n = 2$) and thermoplastic elastomer pellets (TE) ($n = 2$). Regarding global samples, crumb rubber from 5 international synthetic turf sports fields (IFFs) were acquired (Sweden, Poland, Chile, Thailand and The Netherlands). The total number of samples was 34, including 29 samples from tire rubber. The samples were manually collected, always avoiding any damage to the surface. In football fields, since the crumb rubber material is quite homogeneous, sampling was conducted in a practical manner selecting accessible areas. After manual sampling, all samples were washed with ultrapure water to remove any contamination of soil and dust, air-dried and placed into glass vials, sealed with an aluminum cap, and stored at room temperature and protected from light until analysis. The shape and size of the particles of the rubber crumb samples are heterogeneous, including particles of <1 mm to about 5 mm. The average density of the samples is 0.6 g mL^{-1}. A detailed description of the samples is given in Table S2 and Figure S1.

2.3. Microwave digestion and ICP-MS analysis

The samples were subjected to an acid microwave digestion (Ultrawave from Milestone, Sorisole BG, Italy). 0.25 g of sample were weighed in a Teflon tube and 3 mL of nitric acid were added to the sample. Then, the digestion was carried out at 1500 W, 260 °C as the final temperature and 40 bar during 40 min. Finally, the extracts were adjusted to 50 mL with deionized water and analyzed by ICP-MS. Some samples were digested in duplicate or triplicate, and a procedural blank was conducted for each digestion batch to assess possible external contamination.

The metal contents of the samples were determined by ICP-MS analysis (Agilent 7900). The sample introduction system was equipped with a Micromist glass low-flow nebulizer, a double pass spray chamber with Peltier system (2 °C) and a quartz torch. The working conditions are summarized in Table S3.

For the quantitative determinations, calibration standards were prepared in nitric acid (6 % *v*/v) and external calibrations (weighted linear regressions) were used. A further description of the analytical method is included in the supplementary material, Table S4.

To avoid possible external contamination, all glassware was immersed in a 10 % (*v*/v) nitric acid bath for 24 h and rinsed with ultrapure water before use. In addition, a standard cleaning digestion protocol with a 30 % (v/v) nitric acid and ultrapure water mixture was carried out prior to every sample microwave digestion.

2.4. Statistical analysis

Basic and descriptive statistical analysis was performed using Microsoft Excel and Statgraphics Centurion XVIII (Manugistics, Rockville, MD, USA). One-way analysis of variance (ANOVA) was carried out to find out significant differences among the different groups of crumb rubber samples (PGs, FFs, TRs, IFFs). In addition, differences between each two groups of samples were evaluated by *t*-test. If the *p*-value given by the software is below the significance level ($p = 0.05$), the H_0 is rejected and H_1 is accepted indicating significant differences among/between groups at the 95 % significance level.

Box-and-whisker plots are graphics that display the variation in a set of data. The box represents the middle 50 % of the data, with a line inside indicating the median. Whiskers extend from the box to the smallest and largest data values excluding outliers. The mean is represented by a cross inside the box.

3. Results

34 samples (see description in section 2.2 and in Table S2) were analyzed by ICP-MS after microwave digestion for the determination of 32 elements (28 metals and 4 metalloids). The concentrations measured are included in Table 1 showing very different levels for the different elements. The results were compared by means of basic statistical data analysis including ANOVA. The statistical parameters for each element (detection frequency and minimum, maximum, mean and median concentrations) are included in Table S5. Fig. 1 depicts the mean element concentration in the different groups of CR. To facilitate visualization, values are presented on a logarithmic scale and in order of abundance. The data subjected to ANOVA analysis included 4 groups of samples namely playgrounds (PGs), football fields (FFs), tree root retainers (TRs) and international football fields (IFFs), showing in some cases statistical differences among metal content in the different sample groups. ANOVA results are included in Table 2 with the information obtained in the multiple range test comparing each two groups of samples (see a description in section 2.4.). In the supplementary material, the mean concentrations for the 4 groups (PGs, FFs, TRs and IFFs) are included in Table S6.

3.1. Most abundant elements

The BW plots for the most abundant elements are displayed in Fig. 2.

The first most abundant element is Zn. This element is found at quite homogeneous concentrations between 1 and 2 % in most samples from FFs, PGs, IFFs and TRs (see Table 1, Fig. 1 and Fig. 2). The high levels of Zn may be attributed to zinc oxide, which is used as a vulcanization additive during the rubber production (Celeiro et al., 2018). The CR tile and the tire samples also showed concentrations in this range. Only three samples, a FF and 2 PGs, showed lower values. Differences were observed among FFs and PGs (see ANOVA in Table 2). These high levels are in consonance with those observed in recent studies in CR surfaces (Celeiro et al., 2018; Moreno et al., 2023), excluding the previous study including world fields with lower concentrations (0.3–0.5 %) (Graça et al., 2022). The alternative materials gave much lower concentrations, especially cork and sand (84 mg kg^{-1} and 8 mg kg^{-1}, respectively).

The second most abundant element was Al presenting concentrations between 500 and 5700 mg kg^{-1} (see Table 1 and Fig. 2). Two samples gave concentrations above the high limit, a tree protector material with 6920 mg kg^{-1} and an indoor playground (airport) with a very high value of 20,000 mg kg^{-1} (2 %). These two samples are above the EU limit for toys (5625 mg kg^{-1}) (Directive 2009/48/EC, 2009). No differences were observed in the ANOVA study (see Table 2). This element was one of the four studied by Schneider et al. (Schneider et al., 2020) with average values between 1000 and 5400 mg kg^{-1}, in consonance with the results obtained in this study. The same author indicates that there is little information about Al level in this type of material.

Fe concentrations were also high especially in the playgrounds with a mean concentration of 2800 mg kg^{-1}, four times above those measured in FFs and IFFs (700 mg kg^{-1} in the fields) (see mean values in Table S6 and see Fig. 2). The concentrations in two of the three tree retainers were especially high, with values of 11,000 and 30,000 mg kg^{-1} (1 and 3 %). In this case, statistical differences among the different groups of samples (FFs, PGs, IFFs) and TRs were observed (see Table 2 and BW plot in Fig. 2). In addition, the CR tile showed a value close to the levels observed in the PGs, and the tire close to the FFs. These higher values for playgrounds and tree retainers could not be compared with literature due to the lack of studies. Regarding FFs, the results were similar to those found by other authors (Bocca et al., 2009; U.S. Department of Health and Human Services, 2019). As regards the alternative materials, the results were quite different, being very low for cork but very high for the TE material (16,000 mg kg^{-1}) (see Table 1 and Fig. 2).

Si was another element also detected at high concentration in the CR. 2.4 million tons of silica are produced every year, most of which are used

Table 1

Concentrations (mg kg^{-1}) of the target metals and metalloids in the analyzed samples. FF: football field, PG: playground, TR: tree root retainer, IFF: international football field, TILE: commercial rubber tile, TIRE: car tire, TE: thermoplastic elastomers, CORK: cork infill, SAND: sand infill.

mg kg^{-1}	Zn	Al	Fe	Si	Ca	Mg	K	Na	Co	Cu	Mn	Ti	Ba	Pb	Cr	As	Cd	Sr	B	Ni	Li	V	Sb	Se	Mo	Bi	Nb	In	U	Be	Tl	Ag
FF-1	19,892	659	407	2971	730	389	727	464	218	33	4.6	23	3.5	12	1.8	0.57	0.47	2.0	1.4	2.6	5.8	1.3	1.8	0.13	0.14	0.16	0.057	0.027	0.030	0.014	0.058	n.d.
FF-2	22,724	466	307	1917	1170	563	757	470	171	24	4.9	22	2.4	14	1.4	0.84	1.0	1.7	2.5	3.0	2.5	1.8	0.71	0.16	0.15	0.16	0.043	0.052	0.041	0.021	0.068	n.d.
FF-3	19,208	2055	695	1954	1398	488	782	655	212	41	7.3	38	3.6	11.	2.6	0.65	0.60	3.0	2.3	5.0	5.7	3.0	0.28	0.11	0.17	0.19	0.060	0.022	0.071	0.048	0.052	n.d.
FF-4	18,976	3714	1694	1879	3648	929	1296	645	255	60	23.4	57	5.6	16	4.0	1.1	1.1	5.0	2.7	5.4	6.5	4.9	0.26	0.18	0.20	0.38	0.030	0.035	0.36	0.11	0.10	n.d.
FF-5	20,224	2468	855	2092	1586	539	786	530	235	62	8.4	25	6.2	13.6	2.5	0.88	0.84	3.7	3.2	3.6	3.8	3.9	0.22	0.14	0.20	0.19	0.015	0.029	0.077	0.052	0.065	n.d.
FF-6	19,032	901	565	2043	1168	510	833	606	159	25	11	33	11	9.6	1.6	0.77	0.40	3.0	1.7	6.1	4.3	3.0	0.36	0.14	0.13	0.28	0.059	0.033	0.10	0.050	0.056	0.0014
FF-7	21,619	1782	1024	2034	2516	635	905	458	341	44	17	43	9.6	18	2.3	1.2	1.5	4.0	7.3	3.3	2.4	2.8	0.39	0.090	0.14	0.16	0.061	0.061	0.19	0.039	0.048	0.0025
FF-8	17,699	648	401	4649	2831	523	508	495	159	15	8.4	23	7.7	12	1.2	0.46	1.5	3.5	1.2	3.0	2.7	1.9	0.53	0.10	0.28	0.071	0.052	0.062	0.059	0.025	0.037	0.0025
FF-9	1664	204	29	2131	102	35	35	n.d.	14	2.5	0.37	3.2	0.61	3.1	0.20	0.048	0.14	0.33	0.22	0.32	0.19	0.23	0.081	n.d.	0.018	0.12	0.064	0.0066	0.010	0.0030	0.0092	0.016
PG-1	14,777	3154	6550	1687	1810	1240	1418	671	128	53	31	71	18	7.2	12	1.6	0.35	4.4	1.9	7.3	3.5	3.4	0.093	0.10	1.1	0.31	0.13	0.021	0.15	0.12	0.053	0.011
PG-2	22,143	2358	6491	1993	2289	599	922	595	251	119	30	25	15	18	8.2	0.45	0.63	4.6	4.3	7.3	1.9	1.7	0.032	0.0087	0.023	0.091	0.028	0.031	0.11	0.089	0.077	0.0015
PG-3	11,501	3657	1522	1651	1262	696	1020	2705	14	21	16	65	13	16	6.1	1.3	0.42	4.8	5.8	6.7	4.4	3.2	0.027	0.36	0.17	0.61	0.030	0.069	0.16	0.049	0.079	0.0067
PG-4	21,081	1074	1213	2169	3001	546	728	682	352	134	25	3.5	9.4	13	2.1	0.25	0.55	3.8	14	7.2	2.1	0.27	0.0064	0.0064	0.0036	0.0083	0.0030	0.044	0.060	0.031	0.061	n.d.
PG-5	11,917	19,696	7657	1519	78,214	946	1169	347	236	56	52	72	21	11	20	1.2	1.3	107	12	7.3	7.5	12	0.029	0.090	0.54	0.33	0.034	0.015	0.21	0.38	0.039	0.011
PG-6	11	599	1034	690	185,048	836	123	128	0.86	1.9	179	21	5.9	1.1	2.6	0.23	0.08	361	1.1	1.8	0.70	0.98	0.038	n.d.	0.048	0.079	0.30	0.0026	0.056	0.034	0.0075	0.28
PG-7	38	1022	735	943	200,731	1047	214	88	0.99	245	183	36	10	2.2	3.8	0.39	0.10	350	0.93	2.3	1.0	1.6	0.094	0.032	0.17	0.062	0.25	0.0041	0.11	0.053	0.014	0.30
PG-8	16	882	1414	893	179,868	938	142	69	0.94	2.5	177	27	6.6	1.2	3.0	0.25	0.074	347	0.93	2.0	0.95	1.5	0.041	n.d.	0.058	0.022	0.26	0.0024	0.065	0.038	0.0080	0.22
PG-9	4515	1049	664	223	232,659	1695	113	195	1.3	5.7	35	48	8.6	7.7	3.8	0.58	0.28	141	5.3	1.3	1.2	1.9	0.059	0.18	0.27	0.044	0.096	0.0059	0.29	0.11	0.015	0.17
PG-10	3784	567	379	857	185,946	1104	115	5.7	0.42	113	18	21	5.2	4.4	3.8	1.1	0.12	178	1.1	1.4	1.0	4.6	0.14	0.27	0.091	0.12	0.11	0.0018	2.7	0.064	0.0097	0.095
TR-1	14,799	6920	11,238	1099	102,679	1439	1342	422	210	73	119	78	37	8.8	23	1.4	0.46	211	6.6	8.5	2.9	15	0.081	0.10	1.5	11	0.13	0.026	0.092	0.11	0.049	0.011
TR-2	3426	1162	29,358	1147	180,002	5229	264	48	4.1	27	38	171	22	8.2	16	3.3	0.14	80	1.3	4.1	2.1	3.2	0.026	0.015	1.0	0.13	0.093	0.0052	0.52	0.099	0.019	0.17
TR-3	13,265	837	4841	378	1111	408	466	250	202	23	435	22	14	52	7.2	2.7	2.1	3.5	3.8	5.0	4.5	3.1	0.72	0.28	1.5	0.27	0.20	0.084	0.11	0.021	0.062	0.11
IFF-1	20,023	953	572	2758	4476	343	643	515	119	54	7.7	23	3.6	16	1.9	0.92	1.26	5.1	3.0	3.3	1.3	2.5	0.95	0.13	0.12	0.20	0.065	0.096	0.043	0.042	0.06	0.0023
IFF-2	16,604	2024	539	3342	9990	880	630	517	46	28	7.7	13	8.8	21	1.7	1.5	2.6	5.6	2.0	3.5	1.6	2.2	1.0	0.11	0.20	0.22	0.060	0.062	0.042	0.078	0.11	0.010
IFF-3	11,558	5731	1869	1814	181,960	35,763	1160	443	9.1	22	67	46	2149	106	8.1	4.8	1.7	158	4.4	4.9	3.0	7.7	76	0.33	0.099	0.56	0.095	0.20	0.22	0.12	0.056	0.053
IFF-4	10,363	4765	545	9061	47,528	1123	559	118	2.9	3.9	14	187	107	12	3.1	0.85	0.44	38	8.9	3.3	0.90	2.8	166	0.38	0.45	0.74	1.2	0.068	0.34	0.11	0.087	0.036
IFF-5	13,811	1220	461	22,418	3166	300	424	320	81	60	4.0	43	4.7	22	1.6	0.66	0.64	4.5	21	2.6	1.8	1.9	1.5	0.25	0.18	1.1	1.2	0.076	0.12	0.052	0.16	0.041
TILE	17,480	2153	5532	1685	11,922	1516	814	535	45	133	12	15	36	38	6.2	3.4	2.4	10	20	5.6	3.8	4.3	1.6	1.2	0.50	1.7	0.14	0.41	0.056	0.062	0.14	0.047
TIRE	8246	906	806	3778	508	446	466	1602	68	5.3	5.6	38	12	0.40	4.6	0.30	0.019	2.0	17	0.78	6.4	0.59	0.34	0.038	0.13	0.024	0.041	0.0012	0.050	0.012	0.051	0.0035
TE-1	1884	718	15,882	1676	225,208	7347	378	217	0.33	87	22	128	6.6	1.2	6.1	0.98	0.080	170	1.2	1.2	4.3	1.2	0.032	0.018	0.23	0.058	0.16	0.0010	0.16	0.046	0.014	0.019
TE-2	1006	520	8944	1494	133,806	3147	213	112	0.21	48	16	30	7.0	2.1	3.1	0.81	0.053	221	0.83	0.90	7.5	1.2	0.016	n.d.	0.21	0.14	0.073	0.0020	0.50	0.026	0.032	0.11
CORK-1	84	53	42	149	4479	97	58	48	0.11	3.9	23	2.6	11	0.56	0.15	0.040	0.017	9.1	12	0.30	0.10	0.21	0.030	0.015	0.042	0.010	0.010	n.d.	0.025	0.0074	0.0049	0.0082
CORK-2	587	77	50	583	6422	192	48	8.6	0.13	4.2	11	3.2	20	0.68	0.17	0.13	0.045	14	1.6	1.3	n.d.	0.34	0.15	0.024	0.077	0.060	0.015	n.d.	0.17	0.015	0.0051	0.13
SAND	8.1	3623	1878	688	50	316	680	3.9	0.56	1.4	20	54	8.9	2.1	1.5	1.8	0.0034	1.6	0.61	1.1	9.2	2.0	0.0082	n.d.	0.023	0.16	0.20	0.0078	1.8	2.1	0.073	0.21

n.d.: not detected.

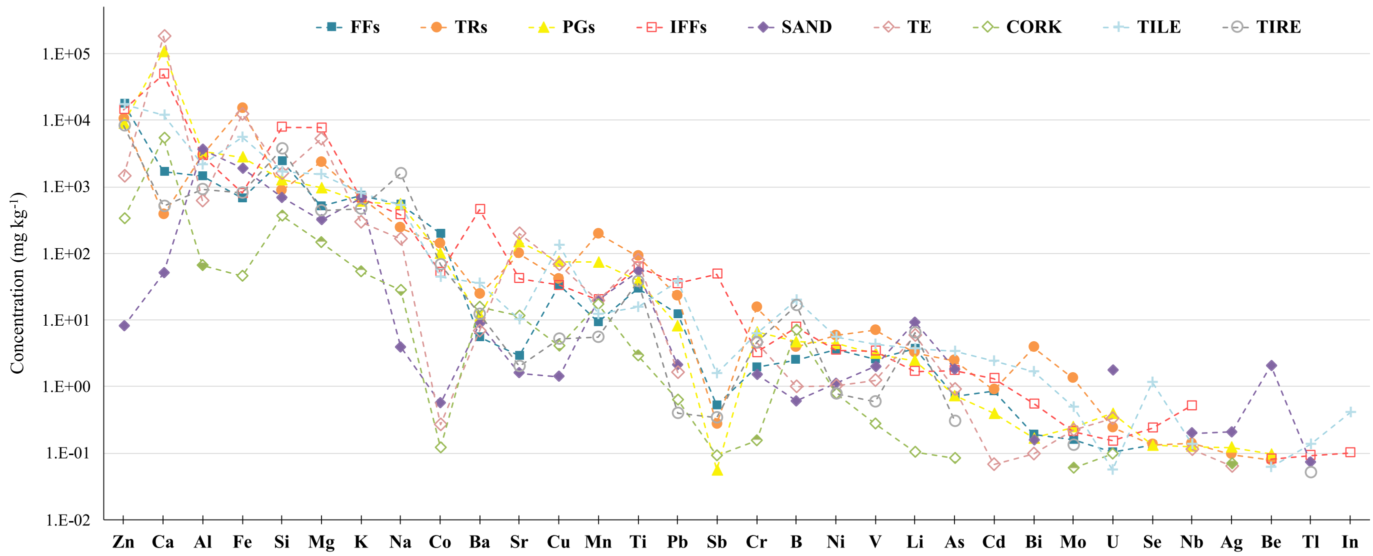

Fig. 1. Mean element concentration in the different groups of samples: football fields (FFs), tree root retainers (TRs), playgrounds (PGs), international football fields (IFFs), thermoplastic elastomers (TE), cork infill (CORK), sand infill (SAND), crumb rubber tile (TILE) and a tire (TIRE) (Y-axis with a logarithmic scale).

Table 2
Results of the ANOVA analysis showing the F-ratios and p-values (4 groups FFs, PGs, TRs, IFFs). Values in bold indicate statistical significance (p-value < 0.05). FFs: football fields, PG: playgrounds, TR: tree root retainers, IFF: international football fields.

	F	P	Pairs showing statistical differences
Zn	1.82	0.1750	FFs-PGs
Al	1.33	0.2900	–
Fe	**9.85**	**0.0002**	FFs-TRs, IFFs-TRs, PGs-TRs
Si	**4.13**	**0.0177**	FFs-IFFs, IFFs-PGs, IFFs-TRs
Ca	2.41	0.0954	FFs-PGs
Mg	1.47	0.2501	–
K	0.17	0.9124	–
Na	0.36	0.7807	–
Co	2.04	0.1395	FFs-IFFs
Cu	1.27	0.3089	–
Mn	**5.23**	**0.0074**	FFs-TRs, IFFs-TRs, PGs-TRs
Ti	2.02	0.1393	FFs-TRs
Ba	1.71	0.1922	–
Pb	2.13	0.1272	FFs-IFFs, IFFs-PGs
Cr	**7.40**	**0.0012**	FFs-PGs, FFs-TRs, PGs-TRs, IFFs-TRs
As	**4.58**	**0.0118**	FFs-IFFs, FFs-TRs, IFFs-PGs, PGs-TRs
Cd	1.94	0.1542	IFFs-PGs
Sr	**3.37**	**0.0357**	FFs-PGs
B	1.44	0.2566	–
Ni	1.00	0.4090	–
Li	1.53	0.2332	–
V	1.53	0.2338	FFs-TRs
Sb	**3.50**	**0.0317**	FFs-IFFs, IFFs-PGs, IFFs-TRs
Se	0.73	0.5461	–
Mo	**19.95**	**0.0000**	FFs-TRs, IFFs-TRs, PGs-TRs
Bi	**3.31**	**0.0382**	–
Nb	**3.94**	**0.0210**	FFs-IFFs, IFFs-PGs, IFFs-TRs
In	**5.98**	**0.0036**	FFs-IFFs, IFFs-PGs, IFFs-TRs
U	0.58	0.6351	–
Be	0.77	0.5252	–
Tl	**3.10**	**0.0468**	IFFs-PGs, IFFs-TRs
Ag	1.68	0.2097	–

to make tires. The rubber reinforced by silica nanoparticles have better wet skid resistance and lower rolling resistance than carbon black, improving the fuel economy and reducing CO_2 emissions (Zhai et al., 2021). The use of silica can be controversial, particularly regarding playgrounds and sports fields. This element was at quite homogeneous levels between 1000 and 3000 mg kg^{-1} in most samples. Nevertheless, two IFFs presented higher values of 9000 and 22,000 mg kg^{-1}, which originated a statistical difference in the ANOVA results (see Table 2). We have not compared these results with literature since this element is not analyzed in other studies despite its importance regarding health implications (Gomes et al., 2021). The alternative materials showed similar levels for the TE pellets, while lower concentrations in sand and even lower in cork (see Table 1 and Fig. 2).

Ca appeared at concentrations between 1000 and 4500 mg kg^{-1} in close to half of the samples, including all the FFs. Nevertheless, some of the samples presented much higher levels including many PGs and TRs, as well as some IFFs with much higher concentrations reaching 10–20 %. The *t*-test revealed statistical differences between FFs (lower values) and PGs. These high values were also found in the TE pellets. In contrast, sand presented very low Ca concentration (50 mg kg^{-1}). These results were not compared to data from literature since this element is hardly studied (Gomes et al., 2021).

Mg showed values between 300 and 1700 mg kg^{-1} in most samples and not any significant difference was observed among playgrounds, FFs, and IFFs. These values were similar to those reported by Graça et al. (Graça et al., 2022) (188–1795 mg kg^{-1}). Only two samples presented higher values, a tree retainer and an IFF (5200 mg kg^{-1} and 36,000 mg kg^{-1}, respectively). The commercial tile and the tire presented values within the range indicated.

In general, the most abundant metals, Zn, Fe, Mg and Al, were also the predominant elements in the study that encompasses worldwide infill football field samples (Graça et al., 2022), demonstrating a good agreement between our study and the worldwide study as regards the CR material. The results of the other two predominant elements, Ca and Si, could not be compared since these elements were not included in other studies. In addition, we also observed a good agreement with our previous study conducted in commercial crumb rubber (Celeiro et al., 2018) in which these six elements were analyzed.

3.2. Other abundant elements

The following most abundant elements were K and Na. The results are visualized in Fig. 3, which also includes the sample distribution for Cu, Co, Mn. K was found between 100 and 1400 mg kg^{-1}, with non-significant differences between fields and parks. Na appeared between 400 mg kg^{-1} and 700 mg kg^{-1} in most samples, excluding a PG with a concentration four times higher close to 3000 mg kg^{-1} (0.3 %); much lower values were found for both elements in cork. Co was at levels about 200 mg kg^{-1} in many samples, well above the safety limit set in

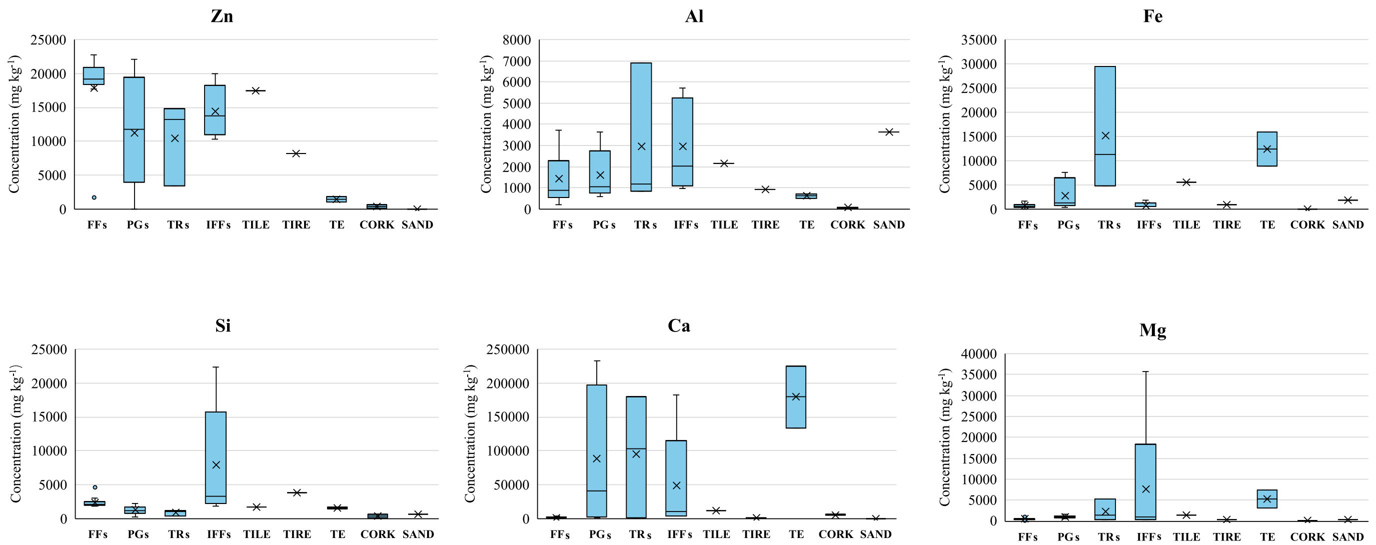

Fig. 2. Box-and-whisker (BW) plots of the major elements, Zn, Mg, Si, Fe, Al, and Ca, in the different groups of crumb rubber samples and alternative materials (for tile, tire and sand, the single value is represented).

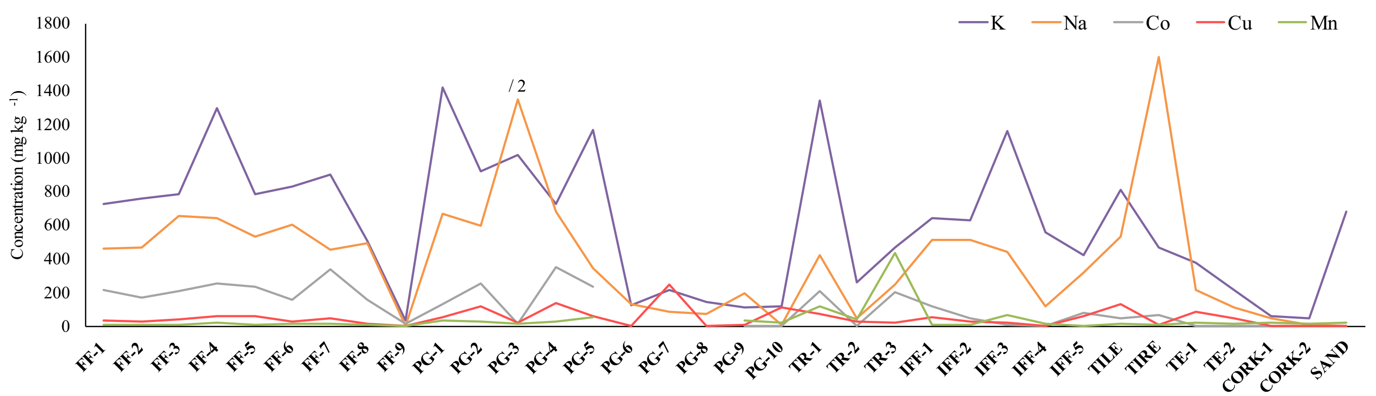

Fig. 3. Line plot showing the individual concentrations of K, Na, Co, Cu, Mn and Ti in the 34 samples. Na concentration in sample PG-3 has been divided by 2.

toys (10 mg kg^{-1}), as already observed in previous studies (Celeiro et al., 2018; Moreno et al., 2023; Schneider et al., 2020), but about one order of magnitude higher than in other ones (Marsili et al., 2015; Han et al., 2008; Menichini et al., 2011). Few samples presented low values <10 mg kg^{-1} and much lower levels were found in the alternative materials ($<$ 0.6 mg kg^{-1}). Cu was measured between 15 and 73 mg kg^{-1} in most samples, as reported by other authors (Gomes et al., 2021) excluding three playgrounds and the commercial tile that gave concentrations close to 100 mg kg^{-1}, and few samples of different origin giving lower concentrations (1–4 mg kg^{-1}). Once more, the alternative materials cork and sand showed very low concentrations. Mn concentrations ranged from 4 to 67 mg kg^{-1}, in consonance with the values found in worldwide FFs (Graça et al., 2022). Higher values were registered in one PG and two TR (100–400 mg kg^{-1}), which made statistical differences in the ANOVA. The alternative materials showed close concentrations among them (11–23 mg kg^{-1}).

Ti content was generally between 13 and 72 mg kg^{-1}, as well as the tire and the commercial tile, as shown in Fig. 4. Two samples, a TR and a field, showed high values close to 200 mg kg^{-1}. In general, few data are available in the literature for this element, but our values were similar to those reported in other study (Graça et al., 2022). The alternative samples presented similar values excluding cork with very low concentrations.

Ba presented concentrations between 2 and 36 mg kg^{-1} and no differences were observed in the ANOVA (Table 2). Nevertheless, an anomalous value was obtained for a foreigner field 2148 mg kg^{-1} and 107 mg kg^{-1} for other IFF (see Table 1). For this element, the alternative materials showed values in the same concentration range as CR samples (7–20 mg kg^{-1}). The distribution of this element is included in Fig. 4, showing the highest values for IFF due to the very high concentrations observed in two samples. Graça et al., 2022 observed a wider distribution values 0.5–161 mg kg^{-1} (Graça et al., 2022).

3.3. Heavy and toxic elements: Pb, Cr, As and Cd

Heavy and toxic elements such as Pb, Cr, As and Cd were found in most samples reaching high values above 20 mg kg^{-1} for Pb and Cr. Some high values were found in tree root retainers, this could facilitate the plant intake of these toxic metals which could be relevant if they are part of the trophic chain (Lopes et al., 2012). The results are shown in Table 1 and Fig. 5.

Pb was detected between 7 and 21 mg kg^{-1} (excluding three lower values), in consonance with the literature (Celeiro et al., 2018; Schneider et al., 2020; Moreno et al., 2023; Plesser and Lund, 2004). Most values were above the safety limit set in toys (13.5 mg kg^{-1}). No significant statistical differences were observed between fields and playgrounds, but in contrast, they were significant between IFFs and PGs. It should be mentioned that one IFF sample presented a very high Pb concentration, above 100 mg kg^{-1}; Values for a TR and the commercial tile were also high (52 and 38 mg kg^{-1}, respectively). Some high

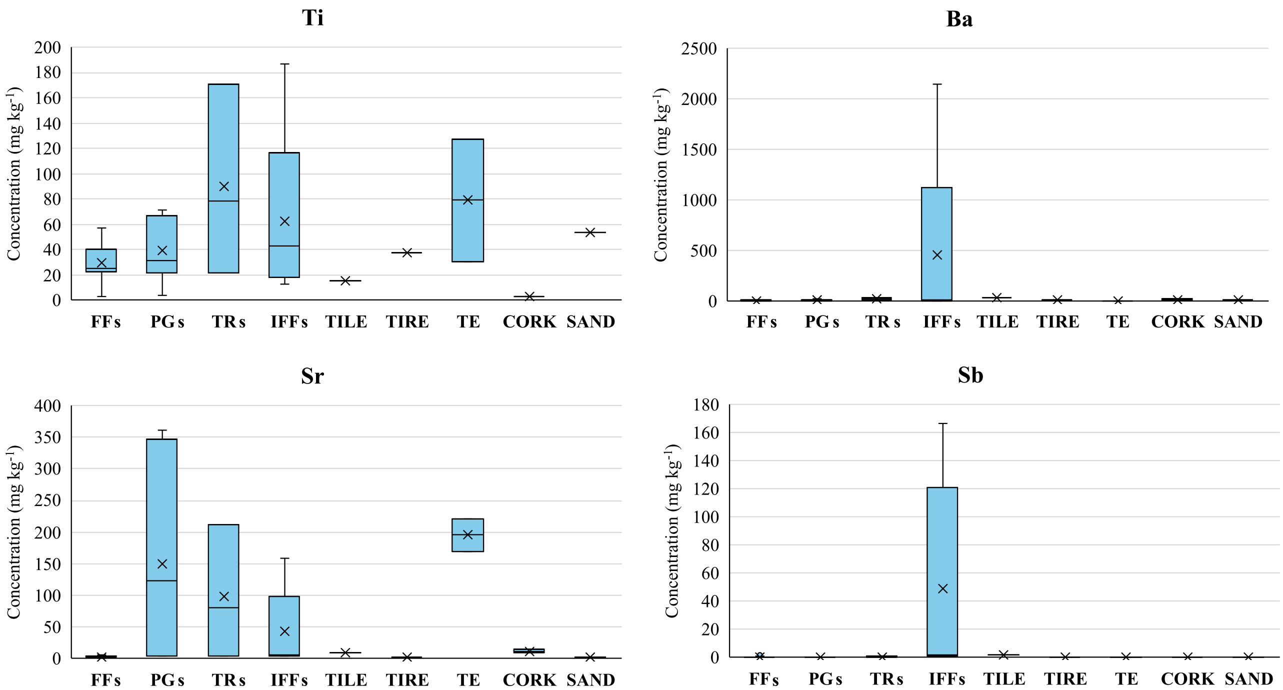

Fig. 4. Box-and-whisker (BW) plots for Ba, Sr and Sb, in the different groups of crumb rubber samples and alternative materials (for tile, tire and sand, the single value is represented).

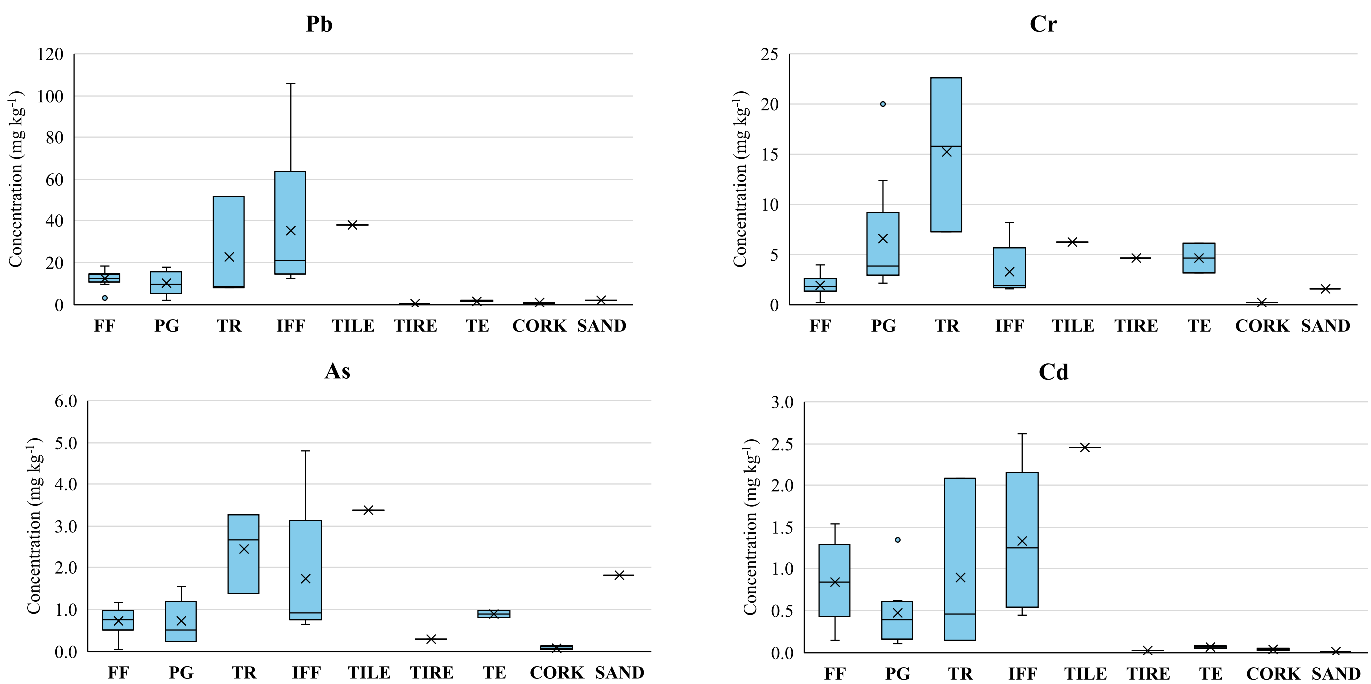

Fig. 5. Box-and-whisker (BW) plots for the heavy elements Pb, Cr, As and Cd, in the different groups of crumb rubber samples and alternative materials (for tile, tire and sand, the single value is represented).

Pb concentrations, reaching 200 mg kg^{-1}, were measured in the international study of Graça et al. (Graça et al., 2022). The alternative materials presented lower values, particularly the cork with concentrations below 1 mg kg^{-1}, being therefore the safest material regarding Pb levels (see Fig. 5).

Cr concentration values were in most cases between 1 and 8 mg kg^{-1}, in consonance with the literature data which does not include playground samples (Gomes et al., 2021). Higher values were found for a playground with 20 mg kg^{-1} (airport), a kindergarten playground (12 mg kg^{-1}) and the two tree retainers (16 and 23 mg kg^{-1}), which made statistical differences among the sample groups (see Table 2, and Fig. 5). In other study, Cr showed high values in colored EPDM material used in playgrounds, as well as in green turf infill (up to 157 mg kg^{-1}) (Moreno et al., 2023).

As contents were in the range of 0.25–1.5 mg kg^{-1} in most CR samples, and not any significant differences were observed between FFs and PGs. These values agreed with those reported in other studies (Gomes et al., 2021; Graça et al., 2022; Celeiro et al., 2018; Moreno et al., 2023).

Three samples presented higher concentrations, two TRs and one IFF (4.8 mg kg^{-1}, above the safety limit of 3.8 mg kg^{-1}), and the commercial tile with 3.4 mg kg^{-1}. The cork material was safer regarding this element with values close or below 0.1 mg kg^{-1}.

Cd concentrations ranged from 0.4 to 2.6 mg kg^{-1} for most CR samples with the highest values for one field sample and the commercial tile. Once again, some samples gave values above 1.9 mg kg^{-1} (safety limit for toys). Similar values were found in the literature (Gomes et al., 2021; Graça et al., 2022; Celeiro et al., 2018). The alternative materials showed very low levels, mainly cork and sand with concentrations lower than 0.05 mg kg^{-1}.

3.4. Other elements at trace levels

Sr was measured at concentrations of 2–6 mg kg^{-1} in the fields and in some PGs but several samples, mainly playground samples (PGs, TRs and an IFF), presented much higher concentrations at about 100 mg kg^{-1} or above. In other studies, the concentrations in CR fields were between 5 and 16 mg kg^{-1} (Bocca et al., 2009; Menichini et al., 2011). The alternative materials showed concentrations between 2 and 14 mg kg^{-1} excluding the TE pellets (about 200 mg kg^{-1}).

Fig. 6 displays the concentration distribution for B, Ni, Li and V. B presented concentrations between 1 and 20 mg kg^{-1} very close to the concentrations found by Graça et al. (Graça et al., 2022) (0.35–34 mg kg^{-1}). In other studies, this element is not included. Ni and Li concentrations were in the range of 1 to 9 mg kg^{-1}, including the alternative samples and no significant differences were observed (except cork with very low Li level of 0.1 mg kg^{-1}. V content was between 1 and 5 mg kg^{-1} excluding one sample of TR, of PG and of IFF with higher values of 15, 12 and 8 mg kg^{-1}, respectively.

Sb appeared with values between 0.01 and 2 mg kg^{-1}, excluding two IFFs with very high values of 76 mg kg^{-1} and 166 mg kg^{-1}, well above the safety limit set in toys (45 mg kg^{-1}) (see Fig. 4). This element is hardly studied in literature.

Lastly, Se, Mo and Bi, Nb, In and U were found at concentrations of 0.6 mg kg^{-1} and below, excluding Bi in a TR (11 mg kg^{-1}) and U in an IFF sample (2.7 mg kg^{-1}). The lowest levels were for Be, Tl and Ag ($\leq$ 0.1 mg kg^{-1}).

3.5. Alternatives materials

The results obtained with the alternative materials can be summarized as follows: the element levels in the thermoplastic elastomers (TE) pellets were in general lower than those found in crumb rubber, although some elements such as Ca, Mg, Sr, Ti, and particularly Fe, were found at very high concentrations (16,000 mg kg^{-1}). In addition, the TE pellets (as well as crumb rubber) are not a biodegradable material thereby contributing to pollution by microplastics. The use of this material as infill in leisure and sports facilities has also been banned since 2023, with a transitional period of 8 years to replace it (Commission Regulation (EU), 2023). Therefore, TE does not appear to be a good choice for substituting CR. On the other hand, cork and sand showed much lower element concentrations. The biodegradable nature of cork, and its use as an industrial byproduct (circular economy), would stand out this material as a safe, green and sustainable alternative to CR. In addition, cork is also free of compounds such as PAHs and other hazardous organic chemicals (Celeiro et al., 2021; Duque-Villaverde et al., 2024).

4. Conclusions

In this study, the presence of heavy and toxic metals and metalloids such as As, Cd, Pb, Cr, Co, Cu, Zn or Fe, among others, in a high number of recycled crumb rubber facilities (29 samples) built for children's play and sports practice surfaces was demonstrated. Other elements of potential concern such as Si were also found at high levels. Furthermore, it should be noted that crumb rubber is the first source of microplastics intentionally added that reaches the environment (European Chemicals Agency (ECHA), n.d.). This study constitutes the first one that includes the two principal surfaces made of tire crumb rubber as the main applications of this material, as well as other minor uses like tree root retainers. In addition, the study includes other alternative natural and/or biodegradable materials that could replace crumb rubber, which could be safer regarding their toxic element content.

Several elements reach concentrations up to thousands of mg kg^{-1}, as is the case of Zn, the most abundant one, with quite homogeneous values between 1 and 2 %. Al, Fe, Si, Ca and Mg also showed high concentrations in the parts per thousand, even reaching the percentage in some samples as Al in an indoor playground, Fe in a TR, or Si in 2 IFFs. The high levels of Si are linked to the use of high amounts of silica nanoparticles to improve tire properties. Nevertheless, the use of silica can be controversial, more particularly here having regard to playgrounds and sports fields. In spite of being abundant metals in CR, Al, Si and Ca are hardly studied in literature. In general, Zn, Fe, Mg and Al concentrations obtained for CR are consistent with the worldwide study previously mentioned (Graça et al., 2022).

Other abundant elements (K, Na and Co) reached concentrations up to the hundreds of mg kg^{-1}. Cu, Mn and Ti contents were in most samples between 4 and 73 mg kg^{-1}, except for some playgrounds, the commercial tile and some TRs for which values higher than 100 mg kg^{-1} were found.

Hazardous elements such as Pb, Cr, As, Cd and Sb were found in most samples reaching high values above 20 mg kg^{-1} for Pb and Cr. In the

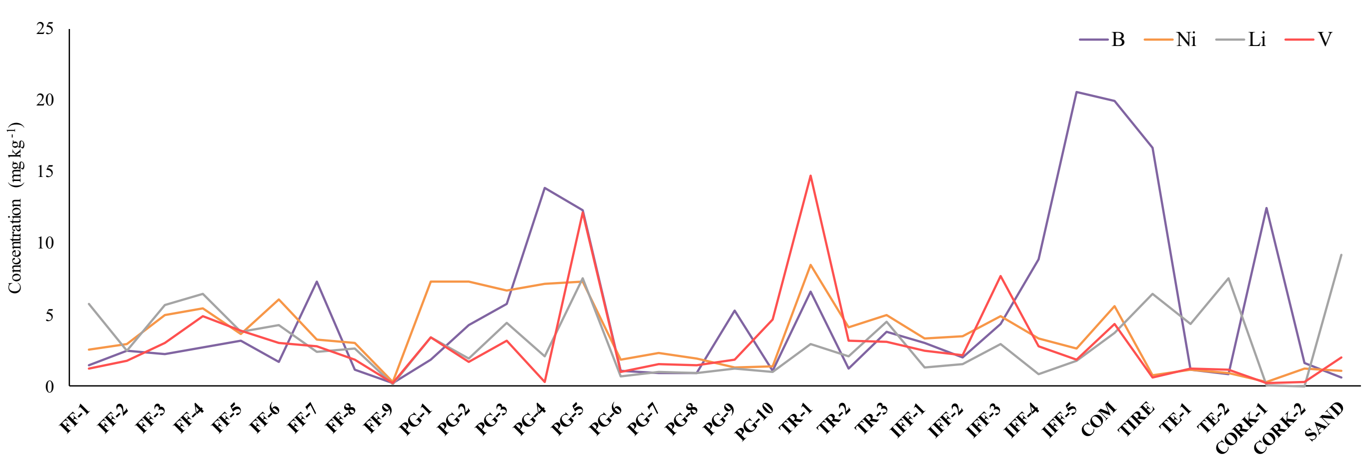

Fig. 6. Line plot showing the individual concentrations of B, Ni, Li and V in the 34 samples.

case of Pb, many samples were very close or above the EU limit set in toys (13.5 mg kg^{-1}), and two samples presented high values, especially a IFF (100 mg kg^{-1}). Cd showed lower concentrations, but some samples such as one FF and the commercial tile presented concentrations above 1.9 mg kg^{-1} (safety limit). Sb appeared at low concentrations ($\leq$ 2 mg kg^{-1}), excluding 2 IFFs with very high values above the safety limit in toys (45 mg kg^{-1}).

Se, Mo, Bi, Nb, In and U were found at concentrations of 0.6 mg kg^{-1} and below. The lowest levels detected among all the analyzed elements were for Be, Tl and Ag.

Concerning regulations, there is a lack of data related to the element content in crumb rubber even though some of the target elements are included in the EU Directive regarding the safety of soils and toys (Gomes et al., 2021; Council Directive 86/278/EEC, 1986; Directive 2009/48/EC, 2009). Zn contents are well above the safety limits, and those limits are also surpassed for other elements, such as Al in an indoor playground (airport), Pb in several samples and Co in most of the CR samples.

ANOVA analysis revealed no statistical differences among groups of CR samples (PGs, FFs, TRs, IFFs) in many cases. However, the differences among PGs and football fields were significant for some elements such as Zn, and Si, the latter with higher values for IFFs. IFF presented higher values for some other elements such as Ba, Sb, Nb and Tl due to the high concentrations measured in some samples and a wider distribution range than FFs. On the other hand, major metals such as Ca, Fe, Sb and Cr, showed higher values for PGs and TRs, which, in some cases, might be attributed to the colored materials.

As regards the alternative materials, the element levels in the TE pellets were in general lower than those found in CR, although some elements such as Ca, Mg, Sr, Ti, and particularly Fe, were found at very high concentrations. Besides, the TE pellets contribute to microplastic pollution. On the other hand, cork and sand showed much lower element concentrations. Since cork is biodegradable and an industrial byproduct, it could serve as a safe and sustainable alternative to CR.

CRediT authorship contribution statement

Andres Duque-Villaverde: Writing – review & editing, Writing – original draft, Visualization, Validation, Investigation, Formal analysis, Data curation, Conceptualization. **Sergio Sóñora:** Writing – review & editing, Visualization, Validation, Investigation, Formal analysis, Data curation. **Thierry Dagnac:** Writing – review & editing, Funding acquisition. **Enrique Roca:** Writing – review & editing. **Maria Llompart:** Writing – review & editing, Writing – original draft, Supervision, Resources, Project administration, Investigation, Funding acquisition, Formal analysis, Data curation, Conceptualization.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.scitotenv.2025.179267.

Data availability

Data will be made available on request.

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