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Exposure assessment and risk characterization from trace elements following soil ingestion by children exposed to playgrounds, parks and picnic areas

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ABSTRACT

Soil ingestion is an important pathway for exposure to metals for children. The objectives of this study were to: (1) Assess urban soil contamination by selected metals (As, Cr, Cu, Ni, Pb, and Zn) in 24 sites (127 soil samples) in Istanbul, Turkey, (2) Investigate relationships between soil contamination and site properties (type of site, equipment type, soil properties), (3) Characterize the risk for critically contaminated sites by taking oral metal bioaccessibility and two soil ingestion scenarios into account. Average metal concentrations were similar in the 17 playgrounds, 4 parks and 3 picnic areas sampled. Five out of 24 sites (all equipped with treated wood structures) had systematically higher contamination than background for As, Cu, Cr or Zn, and measured concentrations generally exceeded Turkish regulatory values. High Cu concentrations in these sites were attributed to the leaching from wood treated with Cu-containing preservatives other than chromated copper arsenate (CCA). Risk characterization for these sites showed that hazard index was below one in both involuntary soil ingestion and soil pica behaviour scenarios for all metals. However, probabilistic carcinogenic risk for As uptake exceeded 1×10^{-6} in both scenarios. A sensitivity analysis showed that soil ingestion rate was the most important parameter affecting risk estimation. Risk from As uptake for children from soils of parks, playgrounds and picnic areas may be serious, especially if soil pica behaviour is present.

1. Introduction

Soil metal pollution in parks and playgrounds has received wide interest and recent studies suggest that pollution and children's exposure to metals in these areas are of high concern [1–5]. Soil metal pollution may originate from different sources such as high background soil concentrations [6], industry [7], traffic [8] or treated wood structures [9]. Specifically, CCA is the most commonly used chemical to prevent wood from bacterial, fungal and insect attack and the most widely used type (known as CCA-C) contains 47.5% CrO_3, 18.5% CuO and 34% As_2O_5 [10]. Since CCA is a safer replacement to creosote and pentachlorophenol, there are many wood structures in service treated with CCA. At the end of 2003, CCA-treated wood was phased out from the United States and Canadian residential markets and publicly used facilities [11]. However, CCA-treated wood structures are still found in Turkey. Moreover, other wood preserving agents containing Cu, such as ammoniacal copper arsenate (ACA), alkaline copper quaternary (ACQ), and copper azole [12] are increasingly used. Wood treated with CCA or any of these alternative preservatives has a color from light to dark greenish brown, depending on the exact composition. The leaching rates of alternative copper based preservatives were found higher than CCA which also makes their use a potential environmental concern [13].

When children play outdoors, they either unintentionally ingest soil by putting dirty hands and objects in their mouths, or deliberately eat soil [5]. Moya et al. [14] suggest an average value of 137 mg d^{-1} for soil ingestion, where upper percentile values reach up to 1432 mg d^{-1} and reported children with soil pica behaviour may exhibit even higher amounts of soil ingestion. United States Environmental Protection Agency [15] suggests a recommended mean value of 100 mg d^{-1} for children between 1 and 6 years old and cited a study reporting a soil ingestion rate (SIR) of 10 g d^{-1} for children with pica behaviour. In 2008, the USEPA proposed recommended ingestion values of 50 mg d^{-1} for soil, 100 mg d^{-1} for soil and dust (central tendency values) and 1 g d^{-1} for soil pica behaviour (upper percentile) [16]. When children are exposed to soil, the fraction smaller than 2 mm (mode diameter 39 μm) adheres to their hands [17]. Recently, the carcinogenic risk for children from playing in playgrounds was estimated to be close to the probability level of 1×10^{-5} [3]. Arsenic, Cr and Cu potentially accumulate in the topsoil after leaching from CCA-treated wood [18,19] and Cu concentrations in soil are generally highest, followed by As and Cr [20]. Although it is not considered in this paper, absorption

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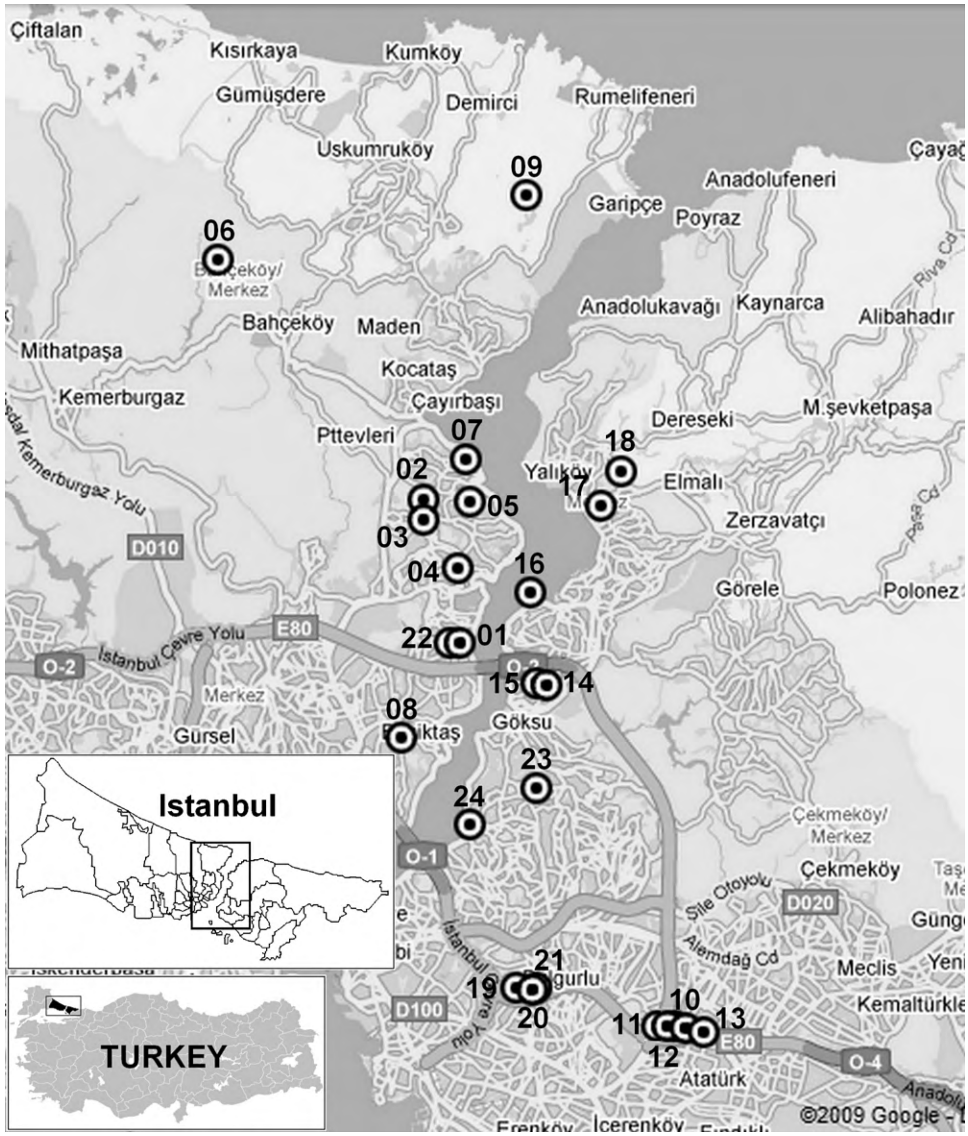

Fig. 1. Study area.

of Cr(VI) by skin is another possible pathway for Cr(VI) uptake [21], in addition to the ingestion of contaminated soil. Thus, in urban sites with potential contamination and children's exposure, it has utmost importance to quantify metal levels and to assess potential metal intake and risk.

Oral bioavailability is the fraction of an ingested contaminant that reaches the systemic circulation from the gastrointestinal tract; and bioaccessibility, in relation to human exposure via ingestion, is defined as the fraction of a toxicant in soil that becomes soluble in the gastrointestinal tract and is then available for absorption [22]. Relative bioavailability (RBA) can be estimated by measuring bioaccessibility via in vitro tests. In vitro bioaccessibility tests are easy to perform and are a good estimator of bioavailability, especially if the test used is validated through comparison to the results of in vivo tests. Recently, a number of bioaccessibility tests for soils have been developed and some of these have been validated for specific metals [22–26]. Bioaccessibility of As, Cr and Cu in CCA-contaminated soils near utility poles has been thoroughly assessed using the IVG (in vitro gastrointestinal) protocol [27–29]. Ljung et al. [5] reported per cent bioaccessibility of different metals in urban playground soils in Uppsala, Sweden in the order of Ni = Cr = Pb $\ll$ As for ingestion of the <50 μm fraction. In their study, average concentrations of As, Cr, Ni and Pb were low (3.4, 39.3, 21.4, and 26.2 mg kg^{-1}, respectively) and whether the sampled playgrounds were equipped with any treated wood structures was not mentioned. In another study by Ljung et al. [30], daily ingestion of soil from playgrounds yielded that tolerable daily intake values can be exceeded under the case of children's pica behaviour, without taking bioaccessibility into consideration. Therefore, there is a need for more extensive studies on risk characterization following soil ingestion by children exposed to playgrounds, parks and picnic areas. Such studies should assess children's metal uptake considering oral bioaccessibility and a variety of play structures including more potentially dangerous treated wood equipment. For that reason, the objectives of this study are:

- To assess the As, Cr, Cu, Ni, Pb and Zn contamination in soil samples collected from parks, playgrounds and picnic areas, where children's exposure is especially of concern.
- To investigate relationships between contamination levels and site properties (type of site, type of recreational structure, main soil properties).
- To assess the exposure and characterize the risk to children who might be exposed to these urban soils.

Table 1
Metal concentrations (<2 mm fraction) and physicochemical properties of soil samples collected in parks, playgrounds and picnic areas.

Site	Type of site	Structure		As	Cr	Cu	Ni	Pb	Zn	pH		TOC	Soil type[a]	Sand	Silt	Clay
			n	(Average, $mg\,kg^{-1}$ dry soil)						(min)	(max)	(average, %)		(% in <2 mm)		
01	Picnic area	Wood	6	6.3	50.6	**93.4**	33.1	11.1	72.6	6.56	7.80	1.40	Sandy clayey loam	52	25	23
02	Playground	Wood	4	6.1	**141**	**302**	31.2	21.2	128	7.21	8.01	3.60	Sandy loam	67	21	12
03	Park	Wood	4	6.7	52.6	26.9	42.5	11.0	88.8	6.73	7.40	3.25	Sandy loam	63	28	9
04	Picnic area	Wood	4	<5.4	71.4	68.9	46.7	44.9	111	5.50	6.93	5.40	Loam	51	31	18
05	Park	Wood	4	**8.9**	32.7	**228**	<5.8	13.9	**206**	7.55	7.75	1.06	Sandy loam	61	21	18
06	Picnic area	Wood	7	<5.9	30.2	31.3	14.7	35.9	33.3	5.93	7.14	2.36	Sandy loam	67	21	12
07	Playground	Wood	4	<5.5	35.7	24.6	17.5	6.6	22.1	7.54	8.03	0.08	Sand	91	5	4
08	Park	Wood	4	<5.7	**91.2**	42.5	25.6	18.7	68.2	7.13	7.80	1.43	Sandy clayey loam	54	26	20
09	Playground	Wood	6	**12.6**	36.3	35.9	<5.2	<5.2	19.1	7.77	8.53	0.02	Sand	97	1	2
10	Playground	Wood	4	6.5	**186**	**230**	20.1	13.9	**182**	6.45	7.34	2.70	Sandy clayey loam	51	27	22
11	Playground	Wood	4	6.8	16.1	13.0	8.9	<5.1	13.6	7.69	7.93	<0.01	Sand	98	1	1
12	Playground	Wood	4	<5.2	15.3	12.4	<5.2	<5.2	13.7	7.02	7.93	0.01	Sand	98	1	1
13	Playground	Wood	4	8.8	**158**	**223**	26.1	22.0	174	6.48	7.37	2.38	Sandy clayey loam	55	23	22
14	Playground	Plastic	4	**8.2**	24.5	9.3	7.3	6.3	26.8	7.20	7.79	0.33	Sand	93	4	3
15	Playground	Metal	4	<5.3	<5.3	<5.3	<5.3	<5.3	43.8	7.43	7.77	0.10	Sand	97	1	2
16	Playground	Plastic	4	**11.4**	33.7	17.8	6.8	7.3	38.9	7.45	8.22	0.42	Sand	91	5	4
17	Playground	Metal	4	5.9	15.8	8.2	14.2	5.6	22.0	7.62	8.03	0.11	Sand	96	3	1
18	Playground	Plastic	4	6.5	13.4	<5.4	8.6	<5.4	16.5	7.44	7.72	0.26	Sand	95	4	1
19	Playground	Metal	5	<5.3	15.1	11.2	<5.3	<5.3	42.7	7.34	7.57	0.49	Loamy sand	85	13	2
20	Playground	Wood	3	<5.5	22.0	25.5	19.1	<5.5	33.4	7.32	7.77	0.15	Sand	94	4	2
21	Playground	Wood	4	<5.4	27.7	**74.3**	7.0	6.5	28.6	7.26	7.75	0.22	Sand	94	3	3
22	Playground	Metal	4	<5.4	24.1	22.1	18.2	10.9	64.4	7.25	7.68	0.94	Sand	92	6	2
23	Park	Wood	4	8.9	68.0	71.9	21.3	38.4	102	7.66	8.03	1.25	Sandy loan	69	18	13
24	Playground	Wood	4	<6.0	24.6	21.7	18.3	18.3	46.8	7.40	7.80	1.90	Sandy clayey loam	50	24	26

Bold values represent at least one of the samples exceeding the limits stated in Turkish Soil Pollution Control Regulation (2005).
[a] According to the United States Department of Agriculture soil classification system.

2. Materials and methods

2.1. Site selection and sampling

Soil samples were collected from 24 sites in the city of Istanbul, Turkey (Fig. 1). The city is one of the biggest metropolitan areas in the world (population exceeds 10 million), and has intense commercial and industrial activities. The sites within the scope of this study were selected according to the following criteria:

- Sampling sites were evenly distributed throughout the city.
- The sites were selected such that children have a high possibility to come in contact with nearby soil. These areas were playgrounds, parks or picnic areas.
- Sampling sites with treated wood structures (determined by visual inspection of wood for greenish brown color), were selected due to the increased possibility of metal contamination from leaching, and playgrounds with metal or plastic structures were also included due to the possibility of contamination via other ways (high background concentrations or traffic-related metal deposition).

Majority of the sampling sites (02, 03, 05, 07, 10, 13–17, and 19–23) were located within 10 m from the main arterials or secondary streets, being under a potentially high impact of traffic activity. Sites 01, 08, 11, 12, and 24 were exposed to moderate traffic activity (roads located more than 10 m away from the sites). The remaining sites 04, 06, 09, 18, and 19 were not impacted by the traffic. Seventeen playgrounds (10 with treated wood, 4 with metal and 3 with plastic structures), 4 parks and 3 picnic areas (all having treated wood structures) were selected for the study. To assess contamination levels and soil properties, soil samples were manually collected with a flat stainless steel sampling instrument and a cutting blade. Sampling instrument and blade were covered each time by a plastic sheet which was disposed of after each operation to avoid possible cross-contamination between samples. Surface soil samples were collected within a distance of 10 cm from the structures and at a depth of 0–2 cm. Each sample consisted in 100–150 g of soil. The number of collected samples ranged between 3 and 7 for each site, depending on the site layout and characteristics (Table 1). An additional representative sample was taken from each site to measure background metal levels in soil, and to determine the particle size distribution. A total of 24 background and 103 potentially contaminated soil samples have been collected.

Soil samples were placed in plastic zip-lock bags after sampling. Collected samples have been air dried at room temperature, sieved (<2 mm), and then refrigerated at 4 °C until analysis. All analyses on samples were completed within a few days after sampling.

2.2. Total metal concentrations

Total metals in soils (<2 mm) were determined on filtered (<0.45 μm) liquid samples with ICP-MS (Perkin Elmer Optima 2100 DV) after chemical solubilization of metals via microwave digestion for 30 min (Berghof Speedwave MWS 3+), based on EPA Method 3052 by adding 9 mL of HNO_3 and 3 mL of HF to 0.5 g of soil sample [31]. Water content of the analyzed samples was determined as per ASTM D 2216-05 [32].

On sites where metal concentrations were found systematically higher than criteria of the Turkish Soil Pollution Control Regulation [33], soil samples were sieved to the <250 μm fraction since it represents the fraction more likely to adhere to children's hands [24,34] and metal digestion has been repeated on the finer fraction.

2.3. Soil physicochemical properties

Soil pH (Hanna HI 221 microprocessor pH meter) was measured according to the ASTM D 4972-01 [35] with a 1:2 soil to water ratio. Total organic carbon (TOC) was measured according to DIN EN 13137 [36] by first soaking the sample in H_3PO_4 to remove inorganic carbon, and then determining the remaining carbon with an elemental carbon analyzer (Costech Instruments ECS 4010). Particle size distribution was determined according to ASTM D 1140-00 [37] and ASTM D 422-63 [38]. The soils were classified using the United States Department of Agriculture (USDA) classification system (gravel [>2 mm], sand [2–0.05 mm], silt [0.05–0.002 mm] and clay [<0.002 mm]).

2.4. Risk assessment and statistical analysis

Since relative oral bioavailability (RBA) data (estimated with bioaccessibility) can be used to provide a more accurate exposure assessment, adjustments to dose (chemical daily intake) values were performed using Eq. (1). The adjusted chemical daily intake for the metals within the scope of the study has been calculated according to the following equation [39]:

$$\mathrm{CDI}_{\mathrm{adjusted}} = \mathrm{CDI}_{\mathrm{metal}} \times \mathrm{B} \tag{1}$$

where $\mathrm{CDI}_{\mathrm{adjusted}}$: adjusted metal daily intake, $\mu g\,kg^{-1}\,d^{-1}$; $\mathrm{CDI}_{\mathrm{metal}}$: metal daily intake, $\mu g\,kg^{-1}\,d^{-1}$, B: bioaccessibility.

Chemical daily intake for each trace element was calculated with the equation provided by USEPA [40]. For the present study, all parameters except exposure point concentrations (EPCs) were taken from the literature or selected in accordance with the exposure scenarios:

$$\mathrm{CDI}_{\mathrm{metal}} = \frac{\mathrm{EPC} \times \mathrm{SIR} \times \mathrm{EF} \times \mathrm{ED}}{\mathrm{BW} \times \mathrm{AT}} \times \mathrm{CF} \tag{2}$$

where EPC: exposure point concentration (determined in the 250 μm fraction), $mg\,kg^{-1}$; SIR: soil ingestion rate, $mg\,d^{-1}$; EF: exposure frequency, $d\,year^{-1}$; ED: exposure duration, year; BW: body weight, kg; CF: unit conversion factor of 10^{-3}; AT: averaging time, d.

Carcinogenic risk was determined according to Eq. (3) and hazard index (also stated as hazard quotient) was calculated using Eq. (4) [39]:

$$\mathrm{Risk} = \mathrm{CDI}_{\mathrm{adjusted}} \times \mathrm{SF} \tag{3}$$

$$\mathrm{HI} = \frac{\mathrm{CDI}_{\mathrm{adjusted}}}{\mathrm{RfD}} \tag{4}$$

where Risk: probability of carcinogenic effect (unitless); HI: hazard index; SF: cancer slope factor $(\mu g\,kg^{-1}\,d^{-1})^{-1}$; RfD: reference dose $(\mu g\,kg^{-1}\,d^{-1})$.

Descriptive statistics and correlations were performed by the computer statistical analysis package S-PLUS 8.0.

2.5. Quality assurance and quality control

All experiments have been performed in duplicates. Additionally, a certified reference material (CRM 025-50) has been analyzed ($n = 4$) to verify the accuracy and precision of the analytical procedure for total metal determination. Certified metal concentrations for CRM 025-050 are 339 $mg\,kg^{-1}$ for As, 441 $mg\,kg^{-1}$ for Cr, 7.8 $mg\,kg^{-1}$ for Cu, 12.2 $mg\,kg^{-1}$ for Ni, 1,447 $mg\,kg^{-1}$ for Pb and 51.8 $mg\,kg^{-1}$ for Zn. All metal concentrations obtained were consistent with the reference values (within the 95% prediction interval). Moreover, 10 procedure blanks were tested during total metal determinations and metal concentrations were below detection limits (50 $\mu g\,l^{-1}$) for all metals.

Table 2
Metal concentrations (mg kg^{-1}) in background soils and near installed structures in selected sites with critical pollution.

	Metal	Fraction	Background	Average	Maximum
Site 01 (n = 6)	As	2 mm	<5.2	6.3 ± 4.3	12.1
		0.25 mm	<5.2	12.6 ± 5.2	20.5
	Cu	2 mm	57.3	93.4 ± 31.3	148
		0.25 mm	195	223 ± 152	503
	Ni	2 mm	<5.2	33.1 ± 19.6	64.2
		0.25 mm	24.2	75.1 ± 48.6	142
Site 02 (n = 4)	Cr	2 mm	77.4	141 ± 9	150
		0.25 mm	38.6	120 ± 27	144
	Cu	2 mm	63.7	302 ± 56	348
		0.25 mm	111	372 ± 59	445
Site 05 (n = 4)	As	2 mm	<6.0	8.9 ± 16.9	34.2
		0.25 mm	<6.0	5.3 ± 0.7	6.0
	Cu	2 mm	63.9	228 ± 102	340
		0.25 mm	153	524 ± 139	616
	Pb	2 mm	76.0	13.9 ± 28.4	40.3
		0.25 mm	179	100 ± 60	161
	Zn	2 mm	555	206 ± 154	357
		0.25 mm	965	497 ± 303	777
Site 10 (n = 4)	Cr	2 mm	89.3	186 ± 24	203
		0.25 mm	52.1	194 ± 44	232
	Cu	2 mm	44.0	230 ± 91	306
		0.25 mm	39.3	255 ± 103	367
	Zn	2 mm	77.1	182 ± 90	182
		0.25 mm	77.2	210 ± 128	392
Site 13 (n = 4)	As	2 mm	<5.5	8.8 ± 3.4	13.5
		0.25 mm	15.7	10.4 ± 6.5	16.1
	Cr	2 mm	54.6	158 ± 63	246
		0.25 mm	56.0	176 ± 73	276
	Cu	2 mm	44.0	22 ± 146	439
		0.25 mm	44.7	252 ± 170	498

3. Results and discussion

3.1. Soil metal content

The trace element concentrations (As, Cr, Cu, Ni, Pb and Zn) in the 103 soil samples (fraction <2 mm) are presented in Table 1. Eleven (sites 01, 02, 05, 08, 09, 10, 11, 13, 14, 16, and 21) out of the 24 sites, had concentrations of As, Cr, Cu or Zn, in at least 1 sample, above the maximum allowable values in Turkish regulations, which are stated as 20 mg kg^{-1} for As, 100 mg kg^{-1} for Cr, 140 mg kg^{-1} for Cu, 75 mg kg^{-1} for Ni, 300 mg kg^{-1} for Pb, and 300 mg kg^{-1} for Zn. Among these sites, 5 sites (01, 02, 05, 10, and 13) had metal concentrations systematically higher than the measured background values (see Table 2 for more detailed results) and were categorized as critically contaminated due to the concentrations close to or exceeding the values stated in the regulation. Pollution levels in the other 6 sites (sites 08, 09, 11, 14, 16, and 21) were not found critical since either measured concentrations did not exceed regulatory values or slightly exceeded them in only 1 sample. In the 103 soil samples, Ni and Pb concentrations were always below the maximum allowable values.

Contamination on the 5 sites having critical metal pollution is discussed below to identify the possible sources of pollution. These sites were also further analyzed for metal content in the <250 μm soil fraction, which is more likely to adhere to children's hands [24]. In site 01 (picnic area), As, Cu and Ni concentrations were higher than the background soil and were close to Turkish regulatory limits (exceeded Cu limit in 1 sample). The average concentrations of these elements were 2–2.5 times higher in the fine fraction compared with the <2 mm fraction. In site 02 (playground) As concentrations were always below regulatory and around background values but Cu and Cr values were higher than both background and regulatory limits in the 4 soil samples. Analysis of finer fraction yielded similar concentrations of Cr and Cu compared with the <2 mm fraction. Site 05 (park) had 1 sample critically contaminated with As and 3 samples with Cu, but Cr concentrations were low. In site 10 (playground) As concentrations were below regulatory values, but Cu and Cr values were higher than background soil and exceeded legal limits in 4 and 3 samples, respectively. Finally, in site 13 (playground), Cu and Cr concentrations exceeded regulatory limits in 3 samples. In the finer fraction, all metal concentrations were higher. In summary, although these 5 critically contaminated sites were equipped with treated wood structures, high concentrations of As, Cr and Cu were not attributed to the possible leaching from CCA-treated wood, because As, Cr and Cu concentrations were not jointly higher in soils. Elevated concentrations of Cu in these 5 sites compared to background soil may be due to the leaching of a Cu-containing wood preservative other than CCA (i.e. ACQ or copper azole). High concentrations of metals other than Cu may be attributed to other sources (i.e. contaminated backfill, atmospheric deposition or heterogeneity of samples). Also, using the total metal content in the <250 μm fraction, instead of the <2 mm fraction, to establish the EPC, is a more appropriate and conservative estimate of the concentration in ingested soil since this fraction tends to have higher metal concentrations in general and is more likely to be ingested by children.

3.2. Site properties and metal pollution

As shown in Table 1, coarse-grained soils were dominant in the selected sampling sites with sand, sandy loam and sandy clayey loam being the most observed soil classes. pH range of soil samples was 5.50–8.53 and most samples had neutral pH values, generally from 7.0 to 7.5. TOC contents were generally lower in sandy soils and values as high as 5.4% have been observed in samples with higher silt and/or clay content.

Classification of soil pollution data (Table 3) according to the type of site (namely playgrounds, parks and picnic areas) and the

Table 3
Average concentrations, standard deviations and maximum values for soil metal concentrations (<2 mm fraction) categorized by type of site and type of installed structure.

		As	Cr	Cu	Ni	Pb	Zn
Playgrounds	Average	5.0	45.8	59.8	11.6	7.1	53.0
(n = 17)	St. Dev.	7.5	56.5	99.8	12.3	9.6	58.4
	Maximum	42.8	246	439	37.1	37.4	305
Parks	Average	7.5	61.1	92.4	21.0	20.5	116
(n = 4)	St. Dev.	8.0	42.5	96.2	19.3	19.1	88
	Maximum	34.2	200	340	47.8	52.3	357
Picnic areas	Average	4.2	47.1	62.0	28.7	29.3	65.6
(n = 3)	St. Dev.	3.3	19.1	34.5	17.6	17.8	33.4
	Maximum	12.1	84.5	148	64.2	56.6	126
Wood structures	Average	5.5	60.3	86.6	19.0	16.2	76.5
(n = 17)	St. Dev.	6.7	54.0	99.7	16.5	17.3	70.9
	Maximum	42.8	246	439	64.2	56.6	357
Metal structures	Average	1.4	13.5	9.9	8.4	4.8	43.2
(n = 4)	St. Dev.	4.1	10.5	10.4	10.0	5.7	30.0
	Maximum	7.1	32.4	37.2	24.7	17.1	116
Plastic structures	Average	8.7	23.8	10.7	7.6	5.7	27.4
(n = 3)	St. Dev.	10.2	10.7	9.4	11.1	3.5	12.7
	Maximum	31.3	40.1	38.6	23.2	12.8	56.9

type of structure (wood, metal and plastic) showed that As, Cu, Cr, or Zn pollution levels exceeded regulatory limits in 9 out of 17 playgrounds in terms of maximum values (n = 70). One park out of 4 (n = 16) and 1 picnic area out of 3 (n = 17) showed As, Cu, Cr, or Zn pollution in some samples. In conclusion, although the majority of the polluted sites were playgrounds, there is not enough data to conclude that the type of site affects pollution characteristics of the soils.

Among the 11 sites with elevated soil concentrations compared to background, 9 of them were equipped with treated wood structures (including 5 critically contaminated sites), and the remaining 2 with plastic equipment. Cu contamination on 6 of these sites (sites 01, 02, 05, 10, 13, and 21) may be attributed to the leaching from wood treated with Cu-containing preservatives other than CCA. In one site (site 08) elevated As, Cr and Cu concentrations in soil may be attributed to leaching from CCA-treated wood. For the remaining sites with treated wood (n = 2) and with plastic structures (n = 2), individual samples with high metal concentrations were thought to occur due to the heterogeneity of the collected samples.

Highest and mean concentrations of As near treated wood structures were comparable to the values reported by Cookson [9] who investigated soil pollution in playgrounds equipped with CCA-treated wood structures near Melbourne, Australia. Ljung et al. [30] also encountered similar As concentrations in playgrounds of Uppsala, Sweden (presence and type of structures in playgrounds were not reported). However, soil As concentrations, reported by Kim et al. [41] near various 1-year-old CCA-treated wood structures in a park located in Seoul, South Korea, were higher than those measured in the present study (average values from different structures were in the range of 15.0–79.8 mg kg^{-1}). Similarly, Townsend et al. [42] reported a higher average As concentration (33.9 mg kg^{-1}) for soils under treated wood structures in a playground in Miami, Florida. Moreover, concentrations of metals in the present study (in the order of Cu > Cr > As) were not in agreement with the values found near CCA-treated utility poles [20,28,29], which stated the order as Cu > As > Cr. In our study, visual inspection was done to determine whether the wood structures were treated with preservatives, however, the exact type of preservative was not investigated. Nonetheless, a relationship was present between increased Cu concentrations in soil and the presence of treated wood structures, which indicates that Cu-containing wood preservatives other than CCA (or a different type of CCA) could have been used to treat the structures in the present study.

Correlation analysis was performed for metal average concentrations (As, Cr, Cu, Ni, Pb and Zn) and soil properties (pH, TOC, sand, silt and clay percentages) with data from the 17 sites equipped with treated wood structures. Pearson correlation values between As–Cr (0.337) and As–Cu (0.339) showed no indication that As, Cr and Cu were originating from a common source, namely CCA-treated wood. In general metal concentrations were negatively correlated with pH and sand content, and were positively correlated with TOC values, silt and clay percentages.

3.3. Total daily intake of metals and risk characterization

An exposure assessment was performed prior to estimating the risk from ingesting contaminated soils by children. The average chemical daily intakes of metals were calculated by considering contaminant source as soil, release mechanism as adherence of contaminated soil to hands and exposure route as ingestion. Risk characterization has been done on chronic basis by calculating carcinogenic risk for As and hazard index values for As, Cr(III), Cu and Zn using respective slope factors and reference dose values from IRIS (Integrated Risk Information System) database [43]. It should be noted that the toxicological profile of Cr(III) has been used instead of Cr(VI), since there is no published slope factor for oral Cr(VI) uptake.

Average chemical daily intakes of metals were calculated based on the equations presented in Section 2.4. These results and background exposure values of metals [44] are presented in Table 4. Exposure point concentrations were taken as the average concentrations of metals in the <250 μm soil fraction (the representative fraction which adheres to children's hands, which is also commonly used to generate the bioaccessibility values) for each critically contaminated site. For the present study, the SIR was taken as 0.1 g d^{-1} for involuntary soil ingestion scenario and 1 g d^{-1} for soil pica behaviour in calculations, as recommended by USEPA [16]. For children 2–6 years old (under the greatest risk due to their common soil eating behaviour) exposure frequency was selected as 180 d $year^{-1}$ for parks and playgrounds, and 50 d $year^{-1}$ (1 d wk^{-1}, 50 wk $year^{-1}$) for picnic areas. Exposure duration was taken as 5 year (from age 2 to 6) and body weight as 18.6 kg for ages 3–6 [16]. Gastrointestinal average bioaccessibility values (determined on the <250 μm fraction) of As, Cr, and Cu for CCA-contaminated sandy soils (n = 4) with moderate to high organic content, were taken from the published work of Zagury and co-workers as 45.8% for As (range: 30.9–51.2%), 19.4% for Cr (range: 9.9–32.9%), and

Table 4
Exposure assessment and risk characterization for As, Cr, Cu, and Zn in critically contaminated sites for children 2–6 years old.

Site	Scenario	Exposure assessment					Risk characterization				
		Chemical daily intake ($\mu g\,kg^{-1}$ bodyweight d^{-1})					Carcinogenic risk	Hazard index			
		Carcinogenic	Non-carcinogenic								
		As	As	Cr	Cu	Zn	As[b]	As	Cr	Cu	Zn
01	Normal	3.03E−04	4.25E−03	6.14E−03	0.133	2.72E−02	4.55E−07	0.014	<0.001	N/A	<0.001
	Soil Pica	3.03E−03	4.25E−02	6.14E−02	1.33	0.272	4.55E−06	0.142	<0.001	N/A	0.001
02	Normal	–	–	6.18E−02	0.800	8.43E−02	–	–	<0.001	N/A	<0.001
	Soil Pica	–	–	6.18E−01	8.00	0.843	–	–	<0.001	N/A	0.003
05	Normal	4.57E−04	6.40E−03	2.42E−02	1.13	0.356	6.86E−07	0.021	<0.001	N/A	0.001
	Soil Pica	4.57E−03	6.40E−02	2.42E−01	11.3	3.56	6.86E−06	0.213	<0.001	N/A	0.012
10	Normal	–	–	9.98E−02	0.548	0.150	–	–	<0.001	N/A	0.001
	Soil Pica	–	–	9.98E−01	5.48	1.50	–	–	0.001	N/A	0.005
13	Normal	8.99E−04	1.26E−02	9.04E−02	0.542	0.132	1.35E−06	0.042	<0.001	N/A	<0.001
	Soil Pica	8.99E−03	1.26E−01	9.04E−01	5.42	1.32	1.35E−05	0.419	0.001	N/A	0.004
			Background exposure ($\mu g\,kg^{-1}\,d^{-1}$)[a]					RfD_{As} = 0.3[c]	RfD_{Cd} = 1500[c]	RfD_{Cu} = N/A[d]	RfD_{Zn} = 300[c]
			0.3	1	30	300					

– Not calculated since average exposure point concentrations were below detection limit. N/A Hazard index is not calculated for Cu since reference dose is not available in the literature.

[a] Taken from Baars et al. [43].

[b] Slope factor for As is taken as 1.5 $(mg\,kg^{-1}\,d^{-1})^{-1}$ from US.EPA IRIS Database [44].

[c] Reference doses (in $\mu g\,kg^{-1}\,d^{-1}$) are taken from US.EPA IRIS Database [44].

[d] Reference dose is not available for Cu, tolerable daily intake is stated as 140 $mg\,kg^{-1}\,d^{-1}$ by Baars et al. [43].

81.2% for Cu (range: 62.2–89.4%) [28,29]. An average bioaccessibility value (<250 μm fraction) of 27% for Zn in residential soils has been reported in the literature [45] and used in the present study. Averaging time was taken as 70 × 365 days (entire lifespan) for assessing the carcinogenic risk of As, and 5 × 365 days (total exposure duration) for assessing non-carcinogenic risk.

For the calculation of carcinogenic risk (Table 4), slope factor for As was taken as 1.5 $(mg\,kg^{-1}\,d^{-1})^{-1}$ for As. For non-carcinogenic risk, reference dose values were taken as 0.3 $\mu g\,kg^{-1}\,d^{-1}$ for As, 1.5 $mg\,kg^{-1}\,d^{-1}$ for Cr(III), and 0.3 $mg\,kg^{-1}\,d^{-1}$ for Zn. Hazard index was not calculated for Cu since there is no reference dose value stated in the literature. Instead, a tolerable daily intake value of 140 $\mu g\,kg^{-1}\,d^{-1}$ [44] was used for the comparison with results.

Calculations for average chemical daily intake values for As, Cr and Cu yielded similar findings for involuntary soil ingestion scenario to the previous studies on CCA-treated wood poles [28,29]. As seen in Table 2, As concentrations in the <250 μm soil fraction were above the detection limit (ranging from 5.3 to 6.1 $mg\,kg^{-1}$) in 3 of the 5 critically contaminated sites (sites 01, 05, 13). In terms of carcinogenic risk, the calculated value for As in soil pica behaviour scenario exceeded 1×10^{-6} in these sites due to the low slope factor value of As, high frequency and long duration of exposure, and relatively low bodyweight although As concentrations were below the Turkish regulatory values (20 $mg\,kg^{-1}$). The risk value stated in the present study was found lower than the cancer risk calculated by USEPA [46] where mean risk was 4.2×10^{-5} for children playing on playsets and decks in warm climate and 2.0×10^{-5} for cold climate (this study also considered direct contact and residue ingestion from CCA-treated wood). However, risk values of the present study were higher than Dube et al. [47] which reported a risk of 4.9×10^{-7} for children between 2 and 6 years old exposed to CCA-treated wood, using a reasonable maximum exposure value for ingestion, dermal and inhalation exposure pathways. For soil pica behaviour, Ljung et al. [30] reported that tolerable daily intake values for As exceeded the acceptable level of 1 $\mu g\,kg\,d^{-1}$ (assuming 10 $g\,d^{-1}$ of ingestion, bioaccessibility was not taken into account) for each of the 25 playgrounds tested in Uppsala, Sweden. Since daily background As exposure from other sources already poses a certain carcinogenic risk for children, additional risk exceeding 1×10^{-6} under the scenario of soil pica behaviour for the children should be considered important.

For the non-carcinogenic risk, hazard index values for As, Cr and Zn under both soil ingestion scenarios were less than 1 in the 5 critically contaminated sites. For Cu, exposure values were well below the daily background exposure and tolerable daily intake. Therefore, toxic risk for these metals appears below dangerous levels even for soil pica behaviour. However, keeping in mind that background exposure for As from other sources is already 0.3 $\mu g\,kg^{-1}\,d^{-1}$, which is equal to reference dose used in HI calculations, additional arsenic uptake from playground soils always leads to a combined HI value larger than 1. Specifically, As uptake under soil pica behaviour scenario combined with background exposure yields HI values of 1.2 and 1.4 in sites 05 and 13. These results state that continuous exposure of children with soil pica behaviour to these soils may result in chronic toxicity of As.

Using the selected parameters stated above for back calculation, As concentrations in soil as low as 7.7 $mg\,kg^{-1}$ yields a carcinogenic probabilistic risk of 1×10^{-6} for parks and playgrounds. A sensitivity analysis has been done with different soil ingestion rates, body weights, exposure frequencies and bioaccessibilities for As exposure for the case of soil pica behaviour. The soil ingestion rate is the factor with the highest uncertainty due to the significant limitations in methodologies used in ingestion studies such as bias in sample selection, poor representation of target populations in terms of race, ethnicity and socioeconomic situation, low reproducibility and limited or absent quality control and assurance [16]. Variations in ingestion rate findings inside a given study and between different studies are very high (in the order of 10^3 in some cases) and individual observations exceeding 50 $g\,d^{-1}$ have been reported in many studies [16]. Taking the soil ingestion rate of 10 $g\,d^{-1}$ for soil pica behaviour in calculations as reported by USEPA gives risk values between 4.55×10^{-5} and 1.35×10^{-4} for carcinogenic risk, and HI values between 1.4 and 4.2. Therefore, more accurate values for soil ingestion rates are needed to better assess the risk. Body weight is another important factor which fluctuates depending on many factors. For example, females tend to weigh less than males and their corresponding average and 10th percentile value is 13.8 and 11.0 kg for ages 2–3 [16]. In this age group, toddlers are more vulnerable and tend to have a more important hand-to-mouth behaviour. Using 10th percentile value gave carcinogenic risk between 7.69×10^{-6} and 2.28×10^{-5}, and a HI in the range of 0.24 and 0.71.

Exposure frequency had a limited effect on risk values. A reasonable maximum estimate of 250 d year^{-1} for parks and playgrounds (assuming access for 5 d wk^{-1}, 50 wk year^{-1}) instead of 180 d year^{-1} increased maximum risk from 1.35×10^{-5} to 1.87×10^{-5} and HI from 0.42 to 0.58. Similarly using an exposure frequency of 100 d year^{-1} for picnic areas (assuming access for 2 d wk^{-1}, 50 wk year^{-1}) instead of 50 d year^{-1} revealed a carcinogenic risk of 9.10×10^{-6} instead of 4.55×10^{-6} and a HI of 0.28 instead of 0.14. Likewise, using a maximum As bioaccessibility of 53.5% instead of an average value of 45.8% had a limited effect on overall results yielding a risk of 1.58×10^{-5} and a HI of 0.49. However, it should be noted that bioaccessibility of As and other trace elements can widely vary with different soil types and is influenced by particle size fraction [5,27–29]. This means that the effect of variations in bioaccessibility in the sensitivity analysis could be higher in other cases.

4. Conclusions

Eleven out of the 24 study sites had at least 1 soil sample with metal content above regulatory limits, and 5 sites, all equipped with treated wood structures, systematically had metal concentrations (As, Cr, Cu, and Zn) higher than background values. Sites with treated wood structures, rather than sites with plastic or metal structures accounted for the majority of the contaminated sites. The elevated Cu concentrations in soil samples from these 5 critically contaminated sites could be linked to leaching from wood treated with copper containing wood preservatives other than CCA. Higher concentrations of As, Cr and Zn may be attributed to other contamination sources. Although average levels of metal concentrations were similar in playgrounds, parks and picnic areas, the latter is of less importance due to the lower possible exposure frequency to the soil.

Probabilistic carcinogenic risk from As-contaminated soil via ingestion has been found high ($>1 \times 10^{-6}$) for soil pica behaviour scenario, even in the soils with low As concentrations. For non-carcinogenic risk, HI values were lower than one for all metals under both involuntary soil ingestion and soil pica exposure scenarios. However, with the presence of high background exposure of children to As, it can be said that even low soil As concentrations in park and playground soils yield a significant additional risk for children.

A sensitivity analysis of the parameters used to calculate the adjusted chemical daily intakes showed that soil ingestion rate was the factor with highest uncertainty and high values may increase the carcinogenic risk and HI values to unacceptable levels. Body weight was another important factor affecting risk due to its large variation in children. Effect of exposure duration and bioaccessibility of As on the risk values were lower compared to the effect of soil ingestion rate and body weight.

Under this conservative approach it can be concluded that, for children 2–6 years old with soil pica behaviour there is a considerable risk in terms of As uptake when playing in soils from playgrounds, parks or picnic areas.

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