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Spatial variability of heavy metal concentration in urban pavement joints – A case study

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Abstract

Heavy metals are known to be among the one of the major environmental pollutants especially in urban areas and, as is generally known, can pose environmental risks as well as direct risks to humans. This study deals with the spatial distribution of heavy metals in different pavement joints in the inner-city area of Marburg (Hesse, Germany). Pavement joints, defined as the joint between paving stones and filled with different materials, have so far hardly been considered as anthropogenic urban soils. Nevertheless, they have an important role as possible sites of infiltration for surface runoff accumulation areas, and are therefore a key feature of urban water regimes. In order to investigate the spatial variability of heavy metals in pavement joints, a geospatial sampling approach was carried out on six inner-city sampling sites, followed by heavy metals analyses via ICP-MS, and additional pH and organic matter analyses. To obtain a risk assessment of heavy metal pollution, different pollution indices were calculated based on regional geochemical background values.

Pavement joints examined consist mainly of basaltic gravel, sands, organic material and anthropogenic artefacts (e.g., glass, plastics) with an average joint size of 0.89 cm and a vertical depth of 2 – 10 cm. In general, the pavement joint material shows high organic matter loads (average 11.0% by mass) and neutral to alkaline pH values. Besides high Al and Fe content, the heavy metals Cr, Ni, Cd and Pb are mainly responsible for the contamination of pavement joints. From the Geo-accumulation Index, the pollution in pavement joints regarding those metals, can be considered as moderate to high. Deterioration of soil quality was reported according to the Pollution Load Index (PLI) for 82.8 % of all sampling points, as well as a very strong potential Ecological Risk (RI) for 27.6 % of the points. The identified spatial pattern of maximum heavy metal loads in pavement joints, could not be attributed solely to traffic emissions, as commonly reported for urban areas. Higher concentrations were detected at runoff accumulation areas (e.g., drainage gutters), and at the lowest sampling points with high drainage accumulation tendencies. Additional Spearman correlation analyses show clear positive correlation between runoff accumulation value and PLI or RI index ($r_{sp} = 0.83$; $p < 0.01$). Further correlation analyses revealed different accumulation and mobility tendencies of heavy metals in pavement joints, based on sorption processes with humic substances, and an overall alkaline pH milieu, especially Cu, Cd and Pb, showed a higher mobility due to low sorption tendencies and pose a specific risk if recaptured by surface runoff. As the presence of heavy metals in pavement joints poses a direct risk for urban environments, and may also affect environments out of urban areas, if drainage transports accumulated heavy metals, we encourage further research to give more attention to this special field of urban soils. Overall urban geochemical background values, and the consideration of runoff related transport processes on pavements, are needed to develop effective management strategies of urban pavement soil pollutions.

Keywords.
Heavy metals, Pavement joints, Urban soils, Urban stormwater, Technosols, Pollution

1. Introduction

The study of heavy metals as environmental pollutants, and their effect on different ecosystems as well as organisms, forms a major research field in environmental science (Alloway, 2013; Blume et al., 2016; Blume et al., 2011; Craul, 1999). In contrast to other pollutants (e.g., organic pollutants) heavy metals are far more widespread, as they are natural components of the earth’s crust (Alloway, 2013). Anthropogenic activity, especially mining, industrial processes, traffic and transport, have led to a global increase of heavy metal concentration in different environmental media like water, air and soil (Cai et al., 2015; Hakanson, 1980; Kowalska et al., 2018; Strode et al., 2009). Along with several emerging threats to the environment, the occurrence and behaviour of heavy metal contamination in soils poses significant challenges for soil ecosystems. Next to the extreme consequences of heavily contaminated soils (e.g. “dead” soil in former mining areas or industrial sites), the presence of heavy metals poses a risk for environmental security, food production, soil organisms and human health (Gałuszka et al., 2014; Strode et al., 2009). It is in this context, and the long-term research on heavy metals in soils, that many of today’s management practices have become established. Taking Germany as an example, various regulations and laws deal with the topic of heavy metals, and provide recommendations or legislated limits regarding concentrations for soils (e.g., Federal soil protection ordinance) (Bundesregierung, 1998; Blume et al., 2011).

In addition to the natural occurrence of heavy metals in soils, urban areas and their soils are particularly exposed to anthropogenic heavy metal sources. Emissions from industrial or home incinerators, traffic, garbage and construction materials could all be seen as the major sources of heavy metals in urban areas (Gunawardena et al., 2015; Craul, 1999; Manta et al., 2002; Sansalone et al., 1998; Defo et al., 2017; Lu and Bai, 2010; Mahanta and Bhattacharyya, 2011). Although it is possible to distinguish between point sources (e.g., industrial exhaust fumes) and diffuse sources (e.g., brake abrasion, corrosion), urban areas are often very densely built up and heavily exploited, resulting in extensive contamination throughout the area (Manta et al., 2002). Therefore, it is not surprising that the contamination status has become an important key feature of urban soils (Lehmann and Stahr, 2007). In contrast to the extensive research of heavy metal contamination in soils generally, the number of studies specifically investigating urban soils is still small (Burghardt et al., 2015; Schad, 2018).

Pavement joints, defined as the joint space between two or several pavement pieces and filled often by gravel, sand and organic material, could be considered to be part of the urban soils. Basically, the question arises whether this material should be defined as "soil" or as anthropogenic material similar to soils. Since urban soils are often very complex in structure, a possible classification for these soils has only existed for a short time (FAO, 2006; Burghardt et al., 2015). Burghardt (1995) has attempted to classify soils in pavement joints as “Interruptosol” and, thus, the soil could be considered as Anthrosol. In terms of the World Reference Base for Soil Resources (WRB) update in 2015, these urban soils are part of the Technosol reference soil group (Burghardt, 1995; Burghardt et al., 2015; Schad, 2018; IUSS Working Group, 2015). One possibility would be to regard it as the uppermost section of an underlying urban soil (urban topsoil or urban surface) or to consider it as its own small-scale urban soil. As a possible part of the urban soils, pavement joints are a common feature of paving on sidewalks, parking lots and access roads, and are used as a design element in public places. Compared to full sealing, they offer the advantage that infiltration is ensured, which plays an important role in the management of urban runoff (especially stormwater runoff) (Sorme and Lagerkvist, 2002; Sansalone et al., 1998; Dierkes et al., 2005a). In this context, they could also be seen as the only soils or anthropogenic material which assumes soil functions in extremely sealed inner-urban areas, that can still perform natural functions, such as interaction with the atmosphere. For these reasons, and because the material in pavement joints can also reach humans directly (e.g., raising dust, playing children), a pollution assessment of pavement joints becomes very important.

If one considers the pollution of urban soils, however, then urban water management must also be included, because with surface and subsurface runoff, sealed or partially-sealed surfaces and urban soils become a source of pollutants and may pollute urban waters. Therefore, a number of studies have focused on the water quality of surface runoff from urban areas that have been sealed or partially-sealed (Drake and Bradford, 2013; Drake et al., 2014). Sörme & Lagerkvist (2002) examined wastewater, and Sansalone et al. (1998) researched urban roadway stormwater, with a focus on heavy metals (Sorme and Lagerkvist, 2002; Sansalone et al., 1998). With a focus on urban water and effluent flow, Gilbert & Clausen (2006) studied drainage from different road surfaces, and Wessolek et al. (2009) examined drainage and pollution in sealed areas (Gilbert and Clausen, 2006; Wessolek et al., 2009). In summary their findings show, that surface runoff generally has a high concentration of heavy metals, hydrocarbons and further trace elements. However, the studies so far focused on parking areas or roads, and few studies mention the pavement joints of sealed or partially-sealed surfaces as an urban interface, which can act as a source or sink of heavy metals (Dierkes et al., 1999; Dierkes et al., 2004; Dierkes et al., 2005a). Furthermore, the pollution retention capability of pavement joints has mostly been determined in laboratory tests and not in the field (Fach and Geiger, 2005). Thus, the understanding of their long-term capacity to retain heavy metals is still limited (Zhang et al., 2018). This may also be due to the fact that the material in pavement joints was not always clearly defined as soil. Therefore, these sites have not been considered as the subject of scientific research on urban soils. Apart from this, the question of whether already installed and used pavement joints, not only in car parks but also, for example on pavements, contain an accumulation of heavy metals, and whether these could possibly act as sources, still remains unclear (Dierkes et al., 2005b).

In this paper, we report on a case study in the inner-city area of Marburg (Hesse, Germany). The goals of our study were (1) the implementation of a heavy metal pollution assessment of pavement joints distributed in an inner-city area, which to our best knowledge represents the first assessment considering diverse installed pavements and different sampling sites, and (2) to empirically depict possible sources and mobilities of heavy metals in pavement joints with a geospatial approach. The results of this study will improve the understanding of the spatial variability of heavy metal contaminations in pavement joints, which is necessary for the development of targeted urban land management strategies.

2. Material and Methods

2.1 Study area

Our case study was conducted in the inner-city area of Marburg (Hesse, Germany) located 75 km North of Frankfurt. The city of Marburg covers an area of 123.91 km² including suburban areas (Hessisches Statistisches Landesamt, 2019) (Figure 1). With 67,851 inhabitants (620 persons/km²; Hessian average: 297 persons/km²) the city of Marburg is the eighth-largest city in Hesse. The city is a central town in a rural region. Land use is divided into settlement area (14.9 %), traffic areas (7.5 %) and vegetation containing green spaces, forests and agricultural area (76.5 %) (Hessisches Statistisches Landesamt, 2019). In contrast to the greater city area, the inner city consists of a medieval town centre with dense urban structures, surrounded by residential, university and commercial districts. Traffic in the inner city is composed of local public transport, delivery traffic and individual traffic. Various main streets with high traffic volumes, especially during rush hours, alternate with traffic-calmed zones and squares. Unfortunately, the data base on traffic counts is very limited. A traffic census conducted by the "Hessen Mobil - Road and traffic management" agency in 2015, counted 44,195 vehicles/24h (national highway B3, which passes the city from north to south), 11 728 vehicles/24h (main road L3125, located in the south of Marburg) and 6 145 vehicles/24h (main road L3092, located in the northwest of Marburg) (Figure 1) (Hessen Mobil, 2015). All census points are located at city area entry roads. A second census conducted by the city administration in 2019 counted 13 039 vehicles/24h for an inner-city main road (Ketzerbach, along with sampling site KB) (Bürgerversammlung Marbach, 2019). Out of this limited dataset the traffic volume in the city area of Marburg could be regarded as moderate compared to similar sized cities. However, by concentrating traffic on certain main routes (resulting from urban development and the locations of the main employers) the traffic volumes reach levels above the limits at rush hours.

For the investigation of the spatial variability in the presence of heavy metals in pavement joints, we based our selection of possible study sites firstly on the traffic volume, and secondly on the location of other possible sources of heavy metals. As traffic emissions, and especially brake abrasion or exhaust fumes, are regarded as the main sources of heavy metals on road sites, traffic volume and shielding from roads are important factors for limitation of heavy metal emission (Duong and Lee, 2011; Gunawardena et al., 2015). However, other sources like house emissions, runoff and weathering of urban installed materials (e.g., pavement itself), should also be considered as possible sources (Gunawardena et al., 2015). Based on this, we aimed to investigate sites that exhibited: 1) differences in traffic volume, 2) difference in the distance to roads and 3) are representative of the different types of paving and construction used within the inner-city zone.

Following these criteria, six sampling sites were chosen across the inner-city zone (Figure 1; Figure S2): "Ketzerbach" road (KB) and "Zwischenhausen" road (ZH) as parallel streets, while KB is an arterial road, and ZH is a side road with very low traffic volume, in a traffic calmed area. "Elisabethkirche" (EK), a square around a medieval church with no direct traffic, but exposed on two sides to streets which carry a heavy volume of traffic. The “Audimax” (AM) square and the parking area at the “Schwanhof” (FV), are private places without traffic, but are also exposed to streets. Finally, we chose the site “Marktplatz” (MP), which is the centre of the medieval district of Marburg, in a traffic calmed area with a high volume of pedestrian traffic. The site MP is located on a hillside clearly above the main traffic routes and completely shielded by buildings.

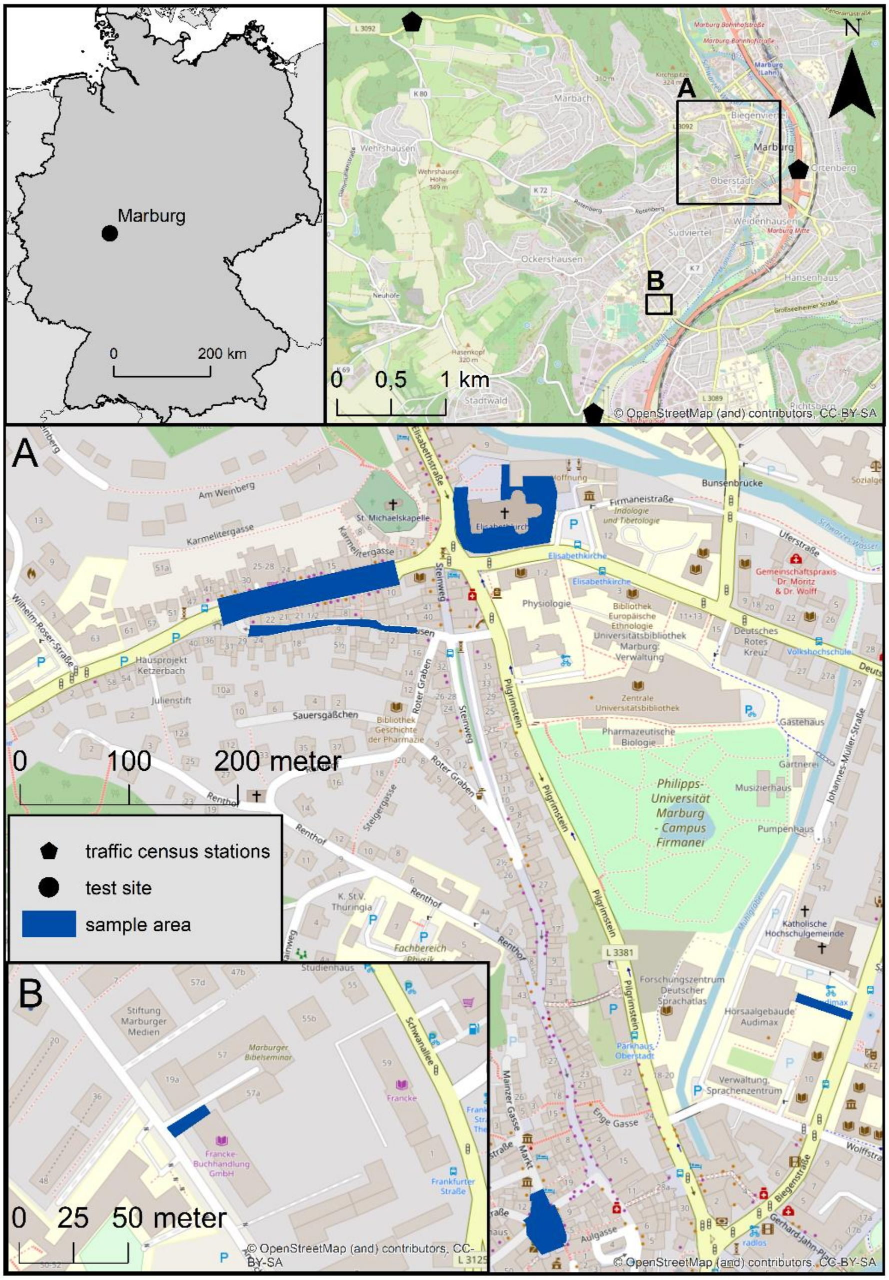


Figure 1: Study area map composition. A: Inner city area of Marburg. B: Sampling site FV in the south-west area of inner city. Data source: © OpenStreetMap contributors 2020. Distributed under a Creative Commons BY-SA License.

2.2 Soil sampling

This study aims to investigate the spatial variability of heavy metals in urban pavement joints, on six study sites located in the inner city of Marburg, and selected according to the above-mentioned criteria (Table 1). At each site, five composite soil samples from a 1 m² pavement area were taken out of the pavement joints (Figure S1). The sampling was carried out with a metal spatula (stainless steel) and plastic spoon. Material was collected from the joints to a depth of 2 to 7cm in 5 places on each 1m^2 site. Each composite sample was stored in airtight plastic (PE) bags until further analyses. Soil sampling points were selected randomly, with the aim of covering the respective sampling area. At site FV and AM the sampling points follow a straight line between street and the next main building. Distance to the nearest road was measured during field work. Urban soil stratigraphy in pavement joints was documented according to German soil classification standards (Ad-hoc AG Boden, 2005), and international soil classification standards (FAO, 2006; IUSS Working Group, 2015). In addition, the joint size and the size and type of installed stones were determined. Slope, potential runoff accumulation and absolute heights (metres above mean sea level) were determined by field measurement and additional height data from LiDAR measurements (Hessian administration for land management and geoinformation, 2019). Potential runoff accumulation was classified according to: 0 = no accumulation (slopes > 2°, highest point at site), 2.5 = moderate accumulation (slope < 2°, no specific feature) and 5 = high accumulation (no slope, lowest point at site, drainage gutter or discharge way). Furthermore, the vegetation coverage of pavement joints by mosses, lichens and small vascular plants, was classified (classes: no vegetation = 0; very low coverage < 1% = 1; low coverage < 2% = 2; medium coverage < 10% = 3; strong coverage < 50% = 4; very strong coverage > 50% = 5). Substructure was finally determined by the removal of individual paving stones.

2.3 Laboratory analyses

All soil samples were oven-dried at 105 °C for 24 hours. Afterwards each sample was ground by mortar and sieved through a 2 mm stainless steel mesh. The content of organic matter (OM) was measured by loss of ignition (DIN ISO 19684-3:2000-08) (Deutsches Institut für Normung e.V., 2000). Ph value was determined in potassium chloride (KCl) with a pH 91 electrode (WTW, Weilheim, Germany) in accordance with DIN ISO 10390:1997-05 (Deutsches Institut für Normung e.V., 2000).

Pseudo-total concentration of the following metals (Al, V, Cr, Fe, Co, Ni, Cu, Sn, As, Cd, Hg, Pb) was performed after extraction of 1 g prepared subsample with aqua regia (12.1 M HCl and 14.4 M $HNO_{3,}$ ratio 1:3) (DIN ISO 11466:2006-12). Metal content was quantified with an ICP-MS (X Series 2; Thermo Fisher Scientific; Bremen, Germany). Each sample was measured three times, and averaged. The resulting mean metal concentrations were converted into the unit mg/kg. Relative standard deviation (RSD) was quantified for all single measurements, after threefold measurement to account for data reproduction and effects of heterogeneous matrixes (Weihrauch, 2018; Voica et al., 2012). Data with an RSD ≥20% were excluded from further evaluation (Thomas, 2001).

Table 1 Sampling site and pavement joint features.

Sampling Station	Site Features			Sampling points	Distance to street (m)	Pavement features				
	Usage	Traffic	Slope			Pavement material	Average joint size (cm)	plant coverage	pH (KCl)	OM[a]
EK	Pedestrian, square around church	not direct frequented, but surrounded from strong frequented streets	medium (2° - 5°)	1	57.02	sandstone paving stones (50x37 cm) on basalt gravel	0.87	0	6.54	10.30
				2	55.42		1.40	4	6.31	9.84
				3	21.48		1.26	2	5.64	14.66
				4	15.49		0.91	3	7.18	9.85
				5	13.82		0.86	2	6.95	-
AM	Pedestrian, square around university building	not direct frequented, single strong frequented road besides	medium (2° - 5°)	1	4.17	concrete paving stones (40x40 cm) on basalt gravel and sand	0.90	0	-	4.74
				2	6.14		0.45	1	-	-
				3	8.60		0.55	1	6.91	11.12
				4	30.56		0.55	1	-	19.44
				5	56.00		0.30	0	7.52	-
ZH	Pedestrian and traffic, side road	direct frequented, traffic calmed area	low (0° - 2°)	1	points direct on street	sandstone paving stones (25x20 or 35x20 cm) on basalt gravel and sand	1.06	1	7.35	3.55
				2			0.83	1	7.81	-
				3			0.93	0	7.60	-
				4			0.94	0	6.99	2.00
				5			0.93	1	-	3.29
KB	Sidewalks at main road	Strong frequented road besides, 13,039 vehicles (24h. 2019)	low (0° - 2°)	1	6.84	sandstone paving stones (25x20 or 35x20 cm) on basalt gravel and sand	1.20	3	6.44	5.94
				2	6.89		1.27	4	6.94	10.55
				3	4.66		1.47	3	6.80	3.02
				4	11.00		1.41	4	7.16	5.41
				5	8.90		1.43	3	-	5.66
MP	Pedestrian, historic marked square	direct frequented, traffic calmed area	low (0° - 2°)	1	21.46	sandstone and basalt paving stones (heterogenous)	0.57	1	7.53	7.03
				2	19.83		1.28	1	7.23	8.41
				3	9.50		0.86	1	7.97	5.19
				4	9.70		1.23	2	7.32	6.01
				5	7.35		1.47	1	6.97	9.49
FV	Delivery traffic, carriage entrance company	Individual traffic only, side road ahead	medium (2° - 5°)	1	3.41	concrete on basalt gravel	0.34	1	6.20	32.69
				2	8.36		0.41	1	6.48	28.14
				3	14.04		0.35	0	9.44	12.09
				4	18.55		0.31	1	4.01	38.08

[a] OM = organic matter (mass %)

2.4 Statistics and data evaluation

Basic statistical operations were carried out using Microsoft Excel 2016 (Microsoft, Redmond, USA), R (R Core Team, 2019) and RStudio (Version 3.5.3; RStudio Inc.; Boston, MA, USA). Additional analyses of height data from LiDAR measurements were carried out with ArcGIS (Esri, Redlands, USA) and QGIS (QGIS Development Team).

In order to give an effective risk assessment, and the pollution characteristics of heavy metals in pavement joints, the "Geoaccumulation Index" (Igeo) for A and O horizons, "Pollution Load Index" (PLI) and the 'Potential Ecological Risk Index' (RI) according to Hakanson (1980) were calculated. All three indices allow an effective assessment of contamination and the spatial differences (Kowalska et al., 2018; Cai et al., 2015). Igeo was determined via the second logarithm of individual HM concentrations divided by 1.5 times the respective background value (Kowalska et al., 2018; Cai et al., 2015). PLI was calculated as the square root of all multiplied single pollution indexes for each individual HM and RI, and multiplying the toxicity response coefficient (given by Hakanson (1980) of the individual metal by the sum of element specific indices of ecological risk (Hakanson, 1980; Kowalska et al., 2018). Because no background values are available for urban soils, we decided to apply the regional background values of natural soil material used during the construction of pavements. These materials are firstly the substructure of basalt gravel (origin: Vogelsberg, Westerwald mountains; alkaline basalt) and sand for the first filling of the pavement joints (origin: sand and gravel pits, Lahn valley). From this, we have calculated a geogenic background value, by averaging the background values for soils from volcanogenic substrates (n = 94) and external sand substrates (n = 64) for topsoil and subsoil in Hesse (Friedrich and Lügger, 2011). In accordance with legal requirements, national comparison and limit values were used to evaluate the heavy metal concentration (Bundesregierung, 1998; Bund-/Länderarbeitsgemeinschaft Bodenschutz, 2003).

To investigate the possible accumulation of heavy metal and their spatial variability in pavement joints, we tested for different correlations between our dataset. Spearman correlation analyses, and tests for normal distribution (Shapiro-Wilk test) were performed with the R-packages "graphics", "stats" and "corrplot" (R Core Team, 2018; Wei and Simko, 2017). Interpretation of significant ($p \leq 0.05$) correlation coefficients was carried out according to the following criteria: weak (rSP 0.4 – <0.6), clear (rSP 0.6 – <0.8), and strong (rSP >0.8) (Zimmermann-Janschitz, 2014).

3. Results and Discussion

3.1 Pavement joint properties – the underestimated urban soil

Pavement joints are common in urban as well as suburban areas; they are purely anthropogenic in origin, and therefore linked to human settlement. The build-up material of pavement joints consists of mineral components (gravel, sand), organic components (organic matter) and artefacts (glass, waste, plastics). Based on their properties and their important substitute function for natural soils in urban areas, pavement joints should be regarded as anthropogenic urban soils. Considering pavement joints as urban soils, makes it necessary to distinguish between the urban subsoils in the substructure of pavements and the soil material in pavement joints, which can be considered as urban pavement topsoils.

The pavement joints in the inner city of Marburg are mainly built up sands with basaltic gravel. From the surface, starting with a thin layer (< 0.5 cm) of organics (often mosses, lichens, single plants) and organic material (comparable to O or A horizon), the major part is built up of sands or sandy loams, with organic material and artefacts (waste fragments, glass fragments) comparable to a C horizon. A layer of sand or basaltic gravel (partly mixed) follows further down, with concrete or mortar in a few places (comparable to the R horizon). This structure results in a classification of pavement joint soil as "Interruptosol" from anthropogenic, deposited, natural and anthropogenic substrates (Burghardt et al., 2015). According to WRB (2015), each pavement joint soil could be classified as: Ekranic Urbic Isolatic Technosol (Arenic, Humic) or Linic Urbic Isolatic Hyperskeletic Technosol (Arenic, Humic) (IUSS Working Group, 2015). Overall, Technosols built up in pavement joints are very young and have not been undisturbed by conversion measures for long periods.

The average lateral joint size is 0.89 cm (± 0.15) by a vertical depth of 2 – 10 cm. The most common pavement material installed is sandstone (natural pavestones), followed by concrete and basalt paving in different size ranges (Table 1). Plant coverage in joints is heterogenous and occurs in small or medium coverage classes (Table 1). Thus, organic matter varies widely, with a total average of 11.10% by mass; with a standard deviation of 9.25%. Maximum values above 30.0% by mass, occur at single points at sampling station FV. The extent of variability is explained by the variety of growth upon pavement joints, the building age of pavement areas and the joint size. For example, wide pavement joints with a massive growth of moss and other vegetation have a high OM content. However younger, smaller joints, filled only with sand or brash, show overall less OM content. As pavement joints are affected by dust and partly even light plant growth, higher organic matter content is typical for urban soils (Lehmann and Stahr, 2007).

The pH in pavement joints ranges between 4.01 and 9.44 (average: 6.97). These overall neutral, to slight or medium alkaline properties, can be traced back to the general surrounding of alkaline materials (e.g., plaster itself, concrete) (Räsänen and Penttala, 2004; Björk and Eriksson, 2002). Additionally, the basaltic underbed could be another factor, as regional basaltic rocks are highly alkaline (Jung and Masberg, 1998). Like OM enrichment, this pH range is specific to urban soils (Lehmann and Stahr, 2007).

In general, these young urban soils, which fulfil the common characteristics of urban Technosols appear in a wide variety, resulting from different materials and construction conditions (e.g., size, substructure). Overall, pavement joints individually are small in size, but through their widespread occurrence they account for a large proportion of urban soils, especially as the only "topsoils" in heavily sealed areas.

3.2 Heavy metal pollution of pavement joints

All metals studied were detected in each of the samples taken, and analysed via ICP-MS. In general, relative standard deviation (% RSD) ranging between 0.71 % and 2.84 % for 11 metals, indicates that our data is clustered around mean values with a small overall variation (Table 2). Only Al (35853.00 mg/kg) and Fe (65968.50 mg/kg) content, reached absolute maximum values. Both elements are ubiquitous in each of the soil and rock materials. In addition, the high values of Al and Fe are a result of the fact, that the under bedding of each pavement is basaltic grit, which when weathered, releases Al and Fe at an increasing rate through decay of the rock (Bain et al., 1980).

For heavy metals, in contrast to Al and Fe, the concentrations of Cr, Ni, Cu, Sn, Cd and Pb exceed the preventative-values for natural soils, specified by the Federal Soil Protection and Contamination Ordinance in Germany (Bundesregierung, 1998). Apart from this, the legal requirement values, which are the legal basis for the need for action in case of soil contamination, are exceeded by Cr, Ni, Cd and Pb (Figure 2a). Depending on the specific requirement value depending on land use, all four metal concentrations exceed the preventative threshold for sandy substrates. Furthermore, the range of Cr and Ni concentrations were above the preventative values for playgrounds (200 mg/kg Cr; 70 mg/kg Ni) at 5 sampling points for Cr (27 sites for Ni respectively). The high level of different heavy metals in our samples is not surprising, as each sampling site is exposed to different anthropogenic heavy metal sources. Besides the release of heavy metals, either by dust or gas in urban areas through incineration, there are other traffic associated sources (Bryan Ellis and Revitt, 1982; Duong and Lee, 2011). For example, Cd, Cr and Ni as a product of combustion of fossil fuels, can reach pavement joints through emissions (Duong and Lee, 2011). Pb, Cu and Sn are strongly associated with traffic emissions (fossil fuel), or the abrasion from brake pads and tyres (Duong and Lee, 2011; Yan et al., 2013). For the spatially narrow inner-city area, both larger point sources and a large number of diffuse sources, must be considered with this in mind, the highest concentrations of heavy metals coincided with typical urban source patterns.

Although increased contamination is typical for urban soils (Schad, 2018; Lehmann and Stahr, 2007), partial concentrations can also be attributed to the materials used in pavement construction. Jung & Masberg (1998) noticed high concentrations of Ni, Cr and Co for mafic volcanic rocks from the Vogelsberg mountains located next to the city of Marburg, and with important quarries for regional construction activities (Fach and Geiger, 2005). However, a challenge lies in the fact that there are no background values for anthropogenic soils or Technosols. Therefore, we calculated the Geoaccumulation index (Igeo) with regional background values of natural soils from the substrate which is used in pavement construction (as described above). Igeo values in total range between -4 and 4, with metal specific differences. Whereas Al, V, Co and Ni remain below the limit of 2, as the threshold between moderately polluted and moderately to highly polluted, outliers of Cr and single values of Pb, exceed this value (Figure 3a). Highest Igeo values are calculated for Cu (mean: 1.26) and Cd (median: 1.98). From these results pollution of pavement joints could be assumed for Cr, Ni and Cu with even clearer values of Cd and Pb.

Table 2 Overview of metal and heavy metal concentrations (given in mg/kg).

Sampling station		Al	Cr	Fe	Co	Ni	Cu	Sn	As	Cd	Pb
						mg/kg					
All stations	min	9090.75	43.35	16977.90	7.65	48.45	35.70	104.55	1.30	0.33	2.92
n = 29	median	17926.50	109.65	40060.50	25.50	140.25	94.35	387.60	3.98	1.11	50.90
	mean	20144.91	157.09	40703.28	25.94	160.21	101.65	560.21	3.95	1.20	61.35
	max	35853.00	1290.30	65968.50	48.45	474.30	252.45	3728.10	7.61	4.08	187.73
	%RSD	2.76	0.71	2.84	2.62	1.65	1.85	0.87	2.74	1.60	1.41
AM	min	9090.75	43.35	16977.90	7.65	48.45	45.90	104.55	2.76	0.54	2.92
n = 5	mean	21462.33	121.89	38028.15	25.50	144.33	77.52	569.67	4.01	1.23	52.39
	max	35853.00	221.85	65968.50	48.45	351.90	107.10	971.55	4.88	2.31	96.62
EK	min	11888.10	56.10	25035.90	15.30	73.95	96.90	252.45	2.94	0.58	22.90
n = 5	mean	15578.46	77.52	30597.45	19.89	99.96	147.90	458.49	4.51	0.88	106.90
	max	20961.00	109.65	40060.50	25.50	124.95	229.50	749.70	5.78	1.35	187.73
FV	min	14269.80	107.10	32079.00	20.40	140.25	79.05	433.50	3.13	1.64	30.70
n = 4	mean	15514.84	422.66	46920.00	24.23	232.05	131.33	1410.15	4.94	2.43	62.23
	max	17064.60	1290.30	58395.00	30.60	474.30	181.05	3728.10	6.45	4.08	119.77
KB	min	14392.20	48.45	30549.00	22.95	76.50	48.45	211.65	1.30	0.33	18.47
n = 5	mean	20088.39	102.00	38652.90	27.54	161.67	87.21	446.76	3.15	0.93	64.12
	max	27642.00	155.55	46894.50	35.70	252.45	165.75	640.05	5.64	1.52	107.41
MP	min	19821.15	124.95	40723.50	25.50	124.95	38.25	221.85	1.59	0.40	17.25
n = 5	mean	26236.95	168.30	50260.50	33.66	222.87	106.08	452.88	3.09	0.82	57.82
	max	34782.00	216.75	65739.00	45.90	344.25	252.45	867.00	4.19	1.51	93.84
ZH	min	10439.70	66.30	20655.00	15.30	99.45	35.70	104.55	2.78	0.81	18.62
n = 5	mean	21062.49	103.28	41004.00	24.48	114.75	65.79	193.29	4.22	1.15	24.78
	max	30982.50	161.93	62041.50	33.15	147.90	94.35	260.10	7.61	1.86	33.81

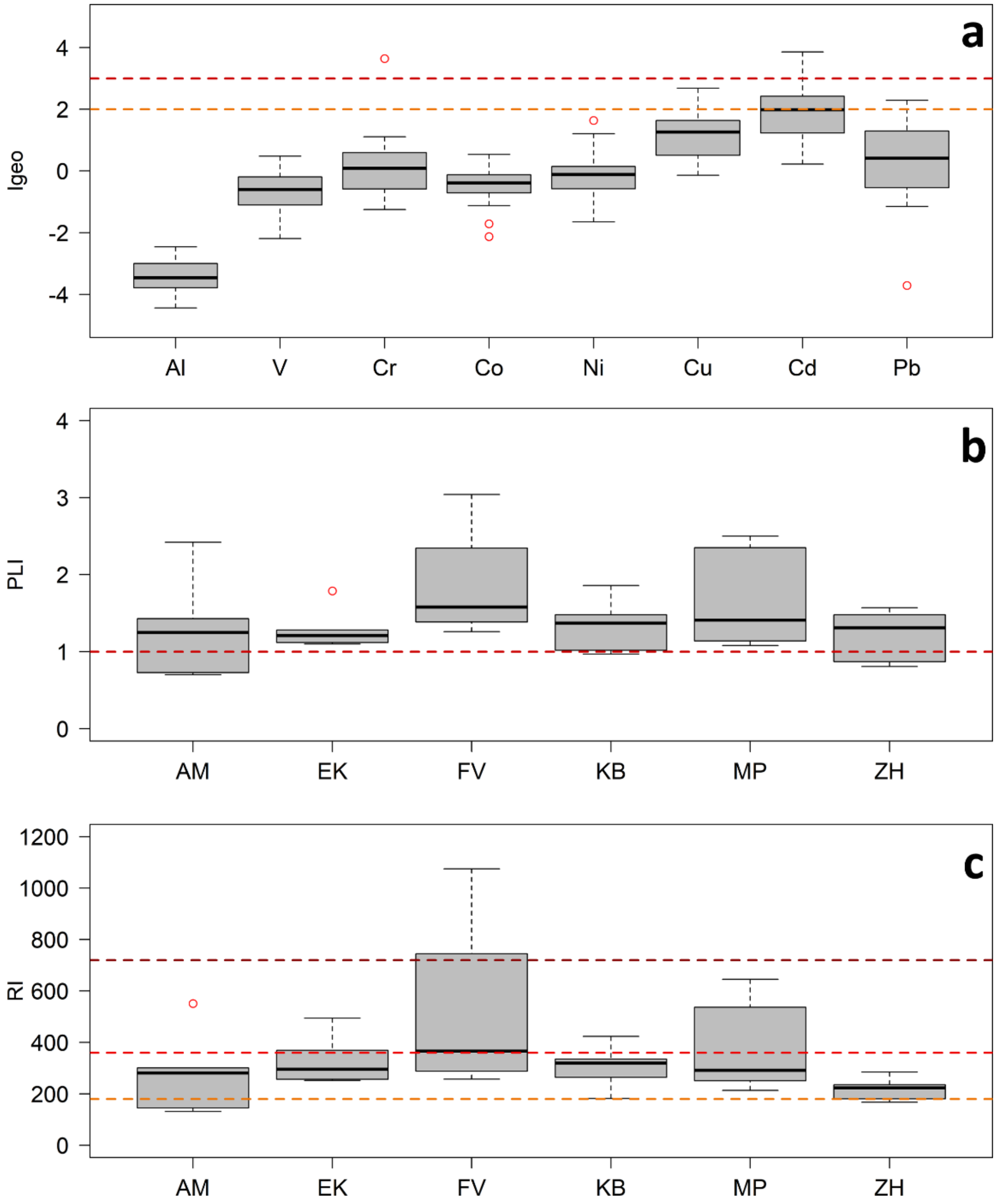


Figure 2: Heavy metal pollution indices. a) I_{geo} Index according to single elements (n = 29) with threshold between moderately and moderately to highly polluted soil (orange line, 2) and threshold to highly polluted soil (red line, > 3). b) PLI Index according to sampling sites (n = 5, FV n = 4) with threshold between only baseline levels of pollution (1) and deterioration of soil quality (red line, > 1). c) RI Index according to sampling sites (n = 5, FV n = 4) with threshold values between moderate and strong pollution (orange line), strong and very strong pollution (red line), very strong and highly strong pollution (dark red line).

As the Igeo index reduces possible variations of lithogenic effects, but strongly depends on the right choice of geochemical background value, these results must always be considered in comparison to natural soils and not directly to urban environments (Kowalska et al., 2018). However, the pollution in pavement joints can be considered as moderate to high for Cr, Ni, Cu, Cd and Pb.

This estimation is also supported by the values of PLI and RI. The results of PLI calculation reflect a deterioration of soil quality for 24 of 29 sampling points, with values between 0.70 (minimum, site AM-4) and 3.04 (maximum, site FV-3) (Supplementary Data). Although the potential of pavement joints as an ecologically important habitat seems to be negligible, they are often the only urban sites with substrate for plant growth in fully sealed spaces. Furthermore, people, especially children, may have direct contact with contaminated soil material. The RI was calculated in order to evaluate these ecological risks. The values show a very high ecological risk at 8 sampling points (point FV-3 with very high risk). At all other sampling points, with the exception of three points (AM-3, AM-4 and ZH-4), a high ecological risk exists (Supplementary Data).

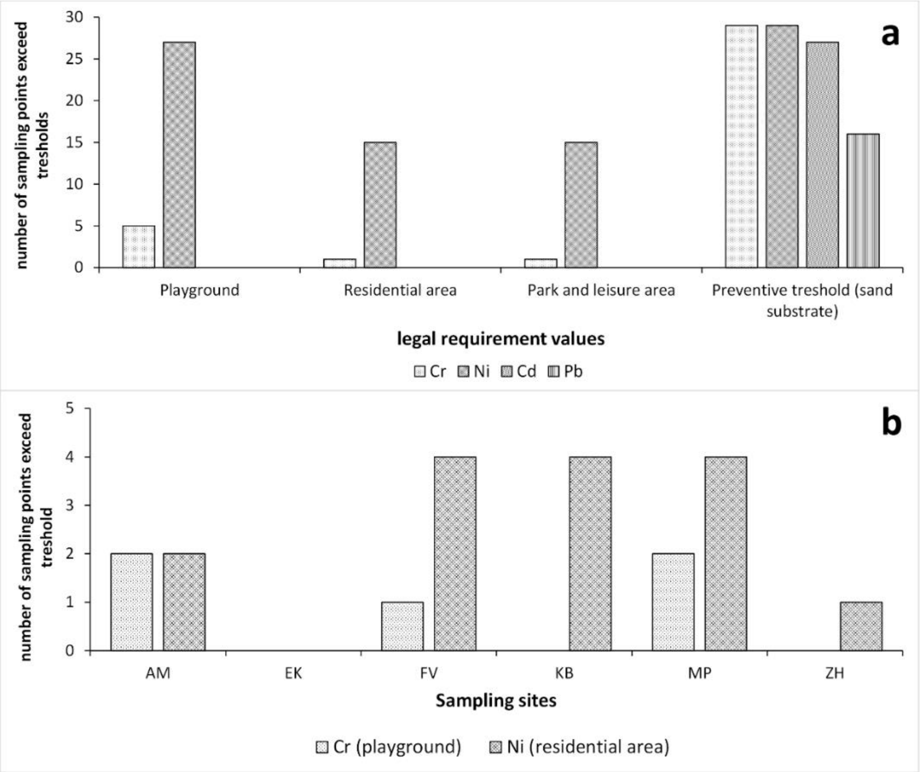


Figure 3: Exceeding legal limits in accordance to sampling points. a) Number of sampling points which exceed the legal requirement values of Cr, Ni, Cd or Pb (given by BBodSchV) for different land use or human activities. Requirement values are: Playground (Cr: 200 mg/kg; Ni: 70 mg/kg; Cd: 10 mg/kg; Pb: 200 mg/kg), Residential area (Cr: 400 mg/kg; Ni: 140 mg/kg; Cd: 20 mg/kg; Pb: 400 mg/kg), Park and leisure area (Cr: 1000 mg/kg; Ni: 350 mg/kg; Cd: 50 mg/kg; Pb: 1000 mg/kg), Preventative threshold (sand substrate) (Cr: 30 mg/kg; Ni: 15 mg/kg; Cd: 0.4 mg/kg; Pb: 40 mg/kg). b) Number of sampling points which exceed the legal requirement values of Cr (for playgrounds) and Ni (for residential areas) according to sampling sites.

3.3 Spatial variability of heavy metal pollution

Different studies dealing with the topic of heavy metal contamination of pavement or urban soils, have noted significant influence of: a) spatial traffic differences (e.g., traffic volume, braking points at crossings) and or b) land use (e.g., industrial sites versus parks) and or c) if considering urban runoff (especially stormwater runoff), the distance to inlets or other emission sources as the major drivers for spatial patterns of urban heavy metal pollution (Bryan Ellis and Revitt, 1982; Duong and Lee, 2011; Herngren et al., 2006; Tedoldi et al., 2017; Logiewa et al., 2020). Through the selection and distribution of our sampling sites and points, we can give results for spatial distribution of heavy metals in pavement joints on two spatial levels: Level one with a differentiation between sampling sites located in the inner-city area of Marburg and, level two including the comparison of single sampling points at each site with its neighbourhood and distance to different heavy metal sources.

Comparing the individual heavy metal loads on the spatial level one, with PLI and RI values (Figure 2b and 2c), it becomes clear that median values for each site exceed threshold values significantly for deterioration of soil quality (PLI, > 1), or moderate to high ecological risk (RI, > 180). As a function of the geochemical background value applied, a general pollution of pavement joints in the inner-city area could be concluded. However, within this general pollution, differences between sampling sites and points occur. Maximal levels of both indices are reached at site FV, followed by MP and AM for PLI Index, and MP and EK for RI Index. Although the median values are quite narrowly spaced, the differences become clear in the maximum values. This trend remains unchanged, even when considering absolute heavy metal loads instead of indices, with element specific deviations. Considering traffic emissions (a) or a specific land use (b) as main sources of urban heavy metal pollution and accumulation in pavement, it is interesting that the maximum PLI and RI values are reached at FV (sidewalk and private driveway at secondary road) and MP (historic market place, traffic-calmed area). In both cases no particular exposure to traffic or contamination-promoting land use (e.g. industry) can be identified. However, exposure to traffic could play an important role for sites AM and EK, as both sites, though not used directly by vehicles, are exposed to major roads. In comparing the locations KB and ZH, the heavy metal concentration at KB is 2.5 times the values from ZH. This finding could be traced back to the higher traffic frequency at site KB (major road), in contrast to side road ZH (Manta et al., 2002). Individual differences between the five sampling points at site KB are explainable by stop and go traffic, as levels of Cu, Cd, and Pb are often higher in areas around traffic lights, and Ni as well as Cu are attributed to braking (Duong and Lee, 2011). The locations EK and MP are traffic-calmed areas, and the highest concentration at EK is reached at EK-3, which is the nearest point to both heavily used streets (Figure S2). The lowest concentration is reached at EK-5, a sample point which is shielded from the streets by the church building, and where traffic emission sources cannot fully reach as Hagler et al. (2011) noted in their study. At site MP the highest level is clearly reached at MP-4 and MP-5. They are the only sample points with higher concentrations. MP-4 is located directly in a rainwater drainage channel and MP-5 is subject to a lot of surface runoff. At site AM, maximal values are reached for Cr, Ni, Cu and Pb next to the major street (AM-1, AM-2) (Figure S2). As sampling site AM was a linear one, AM-5 (greatest distance to street) also shows higher values, especially for Cd, and the second highest PLI is recorded at this site, whereas the other linear sampling site FV, reflects an alternative trend. At site FV the highest concentration of each heavy metal and pollution index is FV-3, followed by FV-4. Both sites are located furthermost from the road.

Considering the pollution indices on spatial level two, they are close to the absolute concentration of heavy metals. Absolute maximum of PLI and RI is reached at point FV-3 (3.03 PLI; 1074.93 RI), followed by MP-5 (2.50 PLI; 645.60 RI) and AM-2 (2.42 PLI; 551.06 RI). Critical legal maximum values for Ni (residential area) are reached by 100.0 % of sampling points at FV and 80.0 % at KB and MP (Figure 3b). The legal maximum value for Cr (playground) is reached at a maximum of 40.0% of all points at site AM and MP. It could therefore be concluded that single pollution hotspots occur in the inner-city area of Marburg. The origin of these hotspots cannot be attributed exclusively to traffic, as stated in the majority of other studies dealing with heavy metals in urban soils (Yan et al., 2013; Herngren et al., 2006; Duong and Lee, 2011; Bryan Ellis and Revitt, 1982). Spearman correlation between the distance from each sampling point to the next traffic frequented street, shows weak positive correlations with single element concentrations for subordinate data (all sampling sites) (Table 3). For sites AM and EK, single strong positive (AM) and negative (EK) correlations occur. However, of the metals mostly relevant for pollution of pavement joints, these concern only Cd at AM and Ni at EK. From these findings, traffic emissions do not seem to be the main reason for the spatial distribution of heavy metals.

Another possible factor in understanding the distribution patterns, could be urban drainage and surface runoff with stormwater runoff. In general, drainage has been shown to be a possible transport medium for heavy metals on paved areas (Gilbert and Clausen, 2006; Tedoldi et al., 2017). The sites AM and FV are examples where the influence of slope and drainage for distribution of heavy metals in pavement joints can be monitored. The highest individual metal concentration and pollution indices are reached at the lowest section in these areas, and nearby gutters. FV has a consistent slope from FV-1 to FV-3. Only FV-4 is located beyond the gutter on a flat section. Site AM presents a similar case: sample point AM-2 with higher concentrations, is also the lowest point and nearby the gutter. The samples were taken in a straight line from AM-1 near a road, over a gutter to AM2 and across a permanent incline to a second gutter (AM-5), in front of a building.

Table 3 Spearman correlation between distance to next road and pollution indices and metal concentrations.

Variable A	Variable B	All sampling sites		AM[a]		EK[a]		KB[a]		MP[a]	
		spearman ρ	p	spearman ρ	p	spearman ρ	p	spearman ρ	p	spearman ρ	p
Distance to next road (m)	OM[b]	**0.50**	**0.01**	**1.00**	**0.00**	0.00	1.00	0.10	0.87	-0.10	0.87
	pH	-0.15	0.50	**1.00**	**0.00**	-0.20	0.80	0.80	0.20	0.30	0.62
	PLI[c]	-0.01	0.96	-0.20	0.75	-0.20	0.75	-0.10	0.87	-0.40	0.50
	RI_av[d]	0.17	0.37	-0.50	0.39	0.60	0.28	-0.30	0.62	-0.70	0.19
	Cu_Igeo[e]	0.32	0.09	-0.50	0.39	0.60	0.28	0.20	0.75	-0.40	0.50
	Cd_Igeo[e]	0.07	0.73	**0.90**	**0.04**	0.60	0.28	0.80	0.10	-0.70	0.19
	Pb_Igeo[e]	**0.58**	**0.00**	0.40	0.50	0.60	0.28	0.50	0.39	-0.40	0.50
	Al	-0.31	0.10	-0.60	0.28	**-0.90**	**0.04**	-0.30	0.62	-0.10	0.87
	V	-0.36	0.05	-0.70	0.19	**-0.90**	**0.04**	-0.60	0.28	-0.10	0.87
	Cr	-0.21	0.27	-0.60	0.28	-0.70	0.19	-0.60	0.28	-0.60	0.28
	Fe	-0.28	0.14	-0.60	0.28	**-0.90**	**0.04**	-0.70	0.19	-0.10	0.87
	Co	-0.36	0.05	-0.60	0.28	-0.87	0.05	-0.67	0.22	-0.46	0.43
	Ni	-0.27	0.16	-0.60	0.28	**-0.90**	**0.04**	-0.60	0.28	-0.70	0.19
	Cu	0.32	0.09	-0.50	0.39	0.60	0.28	0.20	0.75	-0.40	0.50
	Sn	**0.52**	**0.00**	0.70	0.19	0.60	0.28	**0.90**	**0.04**	-0.50	0.39
	As	0.20	0.29	0.20	0.75	-0.50	0.39	-0.20	0.75	-0.20	0.75
	Cd	0.07	0.73	**0.90**	**0.04**	0.60	0.28	0.80	0.10	-0.70	0.19
	Hg	**0.49**	**0.01**	0.50	0.39	0.70	0.19	-0.10	0.87	-0.70	0.19
	Pb	**0.58**	**0.00**	0.40	0.50	0.60	0.28	0.50	0.39	-0.40	0.50

[a] = Sampling sites; [b] = Organic matter; [c] = Pollution load index; [d] = average Ecological risk index; [e] = Geoaccumulation index (element specific)

Table 4 Spearman correlation between runoff accumulation value and pollution indices and metal concentrations for all samples.

Variable A	Variable B	Spearman's rho	p-value
runoff accumulation value	PLI[a]	0.83	0.0002
	RI[b]	0.83	0.0002
	Cu-I_{geo}[c]	0.76	0.0017
	Cd-I_{geo}[c]	0.35	0.2181
	Pb-I_{geo}[c]	0.68	0.0070
	Al	0.31	0.2737
	V	0.28	0.3348
	Cr	0.80	0.0007
	Fe	0.72	0.0036
	Co	0.59	0.0254
	Ni	0.76	0.0016
	Cu	0.76	0.0017
	Sn	0.65	0.0124
	As	0.61	0.0205
	Cd	0.35	0.2181
	Pb	0.68	0.0070

[a] PLI = Pollution Load Index; [b] RI = Ecological Risk Index;
[c] Igeo= Geoaccumulation Index (element specific)

Spearman correlation analysis was carried out to test the relationships between potential runoff accumulation and absolute height (above mean sea level) of each sampling point, with metal concentrations and pollution indices (Table 4). Overall, clear to strong positive correlations ($p < 0.05$) were found between runoff accumulation value, PLI, RI and individual heavy metal concentrations. However, the correlation with absolute heights (metres above mean sea level) shows no clear subordinate correlations. For the sites AM, EK and FV a clear trend is apparent when plotting absolute heights against PLI data (Figure 4). Highest pollution loads are reached at the sites clearly at the lowest point. In the case of site MP, the trend is opposite. As concluded from field work, sampling points MP-4 and MP-5 are higher, but have the highest potential runoff accumulation, as they are reached by a larger surface runoff area above, and located next to discharge points in the pavement or drainage gutters. This result is contrary to other findings, as Tedoldi et al. (2016) reported highest accumulation of heavy metals at inflow points, as a consequence of filtration capacity.

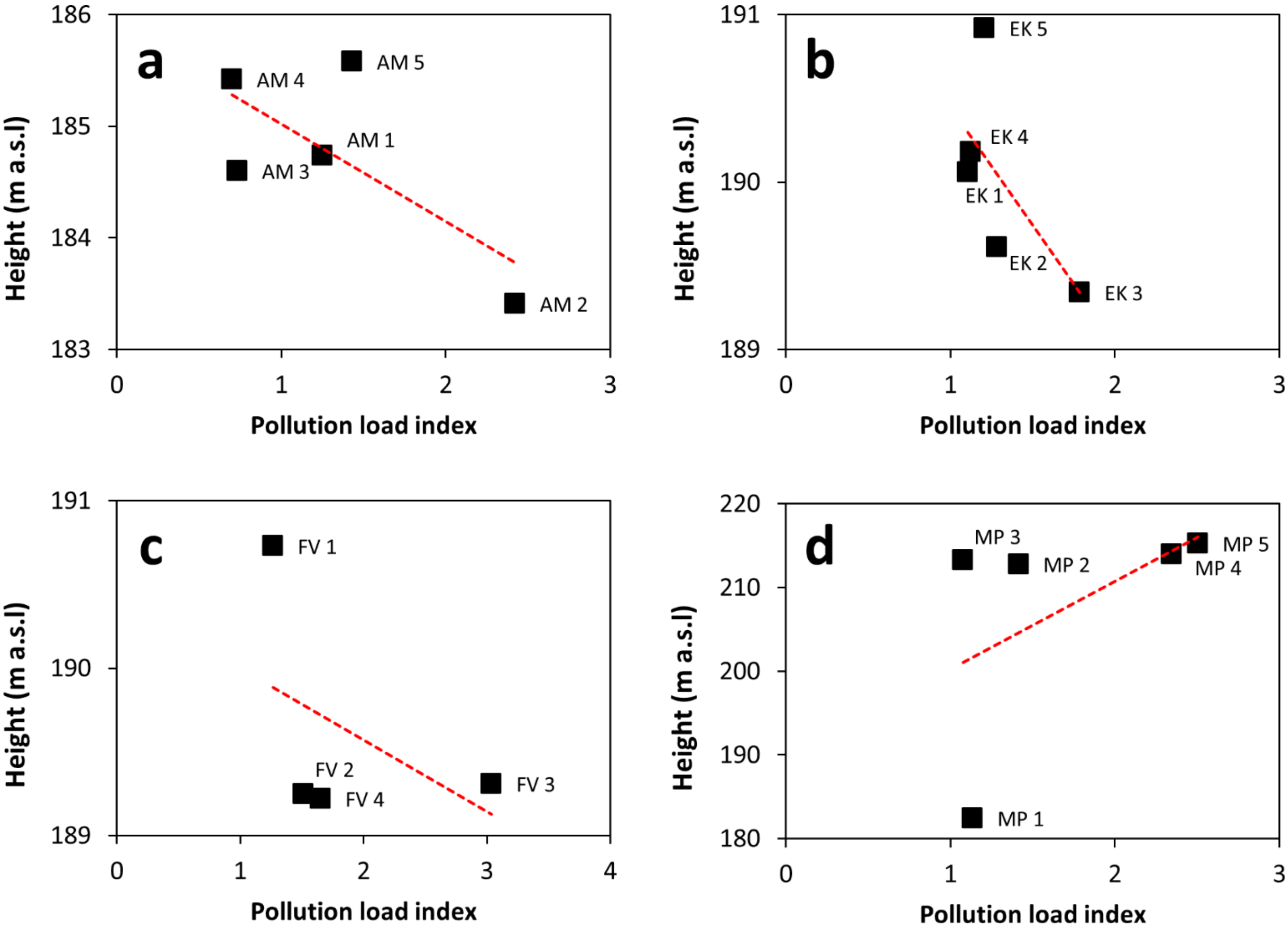
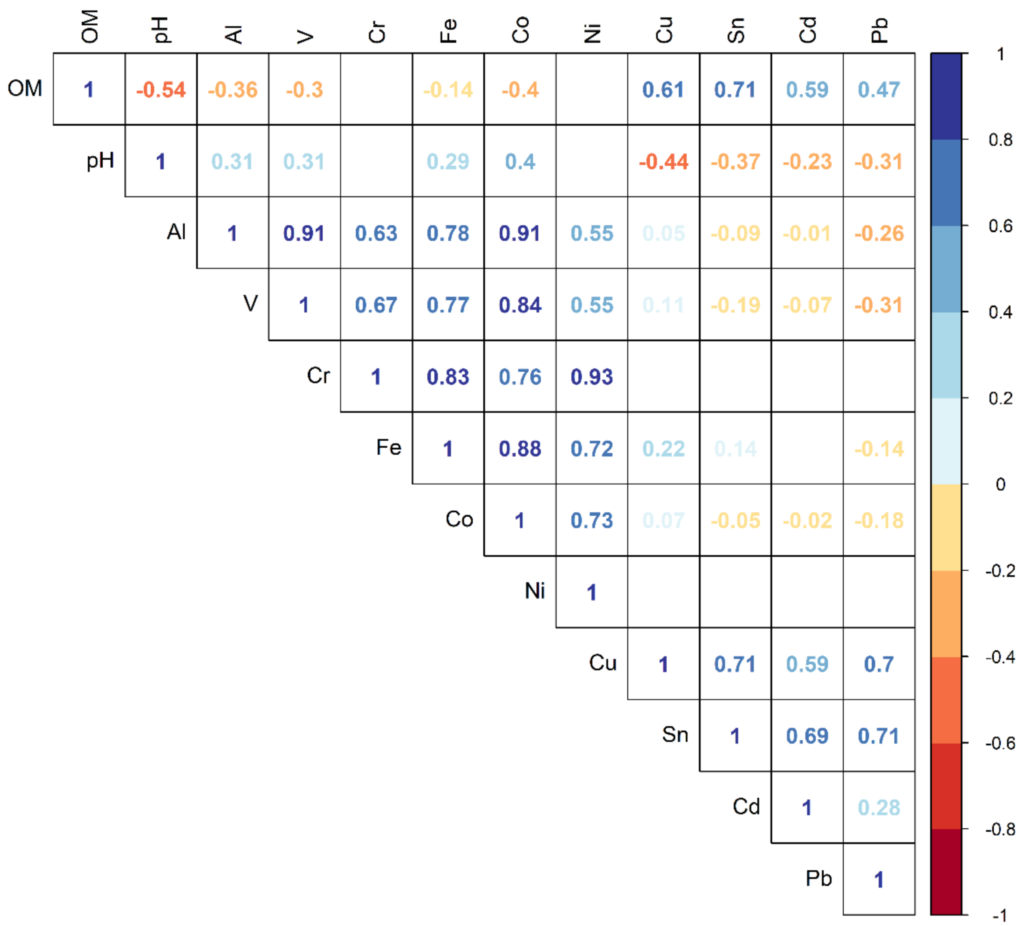


Figure 4: Pollution Load Index (PLI) according to sampling point height (metres above mean sea level) for all sampling sites with significant height differences. a) Sampling site AM; b) Sampling site EK; c) Sampling site FV; d) Sampling site MP. Linear trend showed in red dotted line.

3.4 Accumulation and mobility tendencies of heavy metals in pavement joints

Each sampled pavement joint in the inner city of Marburg shows an alkaline soil milieu and high organic matter (OM) content. Additional correlation analyses of OM content and pH with metal concentration, reveals two groups with significant relationships ($p = < 0.05$), but opposite and reversed conditions (Figure 5). Group 1 including Al, V and Co shows slightly weak negative correlations with OM and weak positive correlations with pH. Accordingly group 2 including Cu, Sn, Cd and Pb shows weak positive correlation with OM and very weak negative correlation with pH. Additional clear and strong positive correlations appear between Al, group 1 metals (Al, V, Cr) as well as the values of Fe, Co and Ni. In contrast, group 2 metals

(Cu, Sn, Cd, Pb) and Pb show no significant or clear correlation with other metals or one another, in contrast to group 1. Those strong inter-element relationships suggest a combined multi heavy metal pollution from similar long-term sources (urban anthropogenic activities) (Manta et al., 2002; Lu and Bai, 2010). As group 2 shows no significant inter-element relationships, this group could be related to different sources from possible short-term processes (e.g., traffic emissions).



	OM	pH	Al	V	Cr	Fe	Co	Ni	Cu	Sn	Cd	Pb
OM	1	-0.54	-0.36	-0.3		-0.14	-0.4		0.61	0.71	0.59	0.47
pH		1	0.31	0.31		0.29	0.4		-0.44	-0.37	-0.23	-0.31
Al			1	0.91	0.63	0.78	0.91	0.55	0.05	-0.09	-0.01	-0.26
V				1	0.67	0.77	0.84	0.55	0.11	-0.19	-0.07	-0.31
Cr					1	0.83	0.76	0.93				
Fe						1	0.88	0.72	0.22	0.14		-0.14
Co							1	0.73	0.07	-0.05	-0.02	-0.18
Ni								1				
Cu									1	0.71	0.59	0.7
Sn										1	0.69	0.71
Cd											1	0.28
Pb												1

Figure 5: Spearman correlation matrix for organic matter (OM), pH and elemental concentrations in pavement joints. Spearman's rho displayed in grid if correlation is significant (p-value < 0.05). Positive correlation in blue colours, negative correlations in red colours.

Next to those inter-element relationships, different correlation clusters with OM and pH could allow suggestions about heavy metal retention in pavement joints. First of all, the pavement joint substrate, built up from sands and coarse gravel with artefacts, allows only poor adsorption of heavy metals on clay minerals, silt or pedogenic oxide surfaces (Blume et al., 2016; Alloway, 2013). In contrast, the high content of OM could lead to the sorption on humic substances, and the formation of metal-humus-complexes (Herms and Brümmer, 1984; Alloway, 2013). In this context, we interpret the clear to strong positive correlation between group 2 metals (Cu, Sn, Cd, Pb) and OM, as evidence of those processes. The adsorption was also found in urban soils for Cu and Pb, but not for Cd (Mahanta and Bhattacharyya, 2011). In contrast, Defo et al. (2017) found a major influence of OM on the retention of Pb as well as Cd, in urban soils (Defo et al., 2017). The overall neutral to alkaline pH milieu (6.97 total average), supports a fixation of metals by specific bindings, since unspecific bonds and dissolutions only occur at more acidic pH values (Herms and Brümmer, 1984; Blume et al., 2016). Comparing the opposite relationships of group 1 and group 2 metals, with additional inter-element relationships, we conclude a strongly bound group of metal-complexes including Al, V, Cr, Co, Fe and Ni (with low potential mobility and a strong adsorption tendency), and a more mobile group of Cu, Sn, Cd and Pb (with high mobility and low adsorption tendency). Regardless of this finding, the widespread pollution with comparatively high concentrations in pavement joints, also indicates an accumulation of metals. Available sorbents, the alkaline environment and a constant supply of heavy metal emissions, provide suitable conditions for pollution accumulation. This point deserves special attention, as other authors have noted heavy metal enrichment by organic material, especially in gutters, which would demonstrate a link between transport by water and accumulation at runoff gathering points (Bryan Ellis and Revitt, 1982). The existing ability for the mobile group of metals to become mobilised, becomes especially problematic in the case of infiltration of surface stormwater runoff and precipitation. If urban surface or stormwater runoff is regarded as a major driver for heavy metal transport and accumulation at the lowest points, a further transport after the infiltration in pavement joints is possible. Particle uptake and transport as suspended load, as well as the transport as dissolved metals in surface runoff are possible (Gilbert and Clausen, 2006). Applying this on a larger scale, and considering urban pavements as pollution sources for the environment out of urban areas, polluted urban runoff could provide a link between both systems, as stormwater runoff especially is discharged directly into receiving waters from urban areas, less polluted environments like rivers, wetlands and floodplains in downstream areas may also be affected.

4. Conclusion

In our study, pavement joints, mentioned as an important part of the urban soils, were found to be polluted by Cr, Ni, Cu, Cd and Pb at each sampling site, as shown by absolute concentrations and Geoaccumulation index. Spatial comparison of PLI and RI with different features, showed that traffic emissions are not the main cause of the spatial distribution of heavy metals in pavement joints. Instead, we found strong correlations between runoff accumulation and heavy metal pollution, mainly at runoff gathering points. The accumulation of heavy metals at gathering points is supported by an alkaline pH milieu and adsorption processes on organic material, which makes up a substantial part of pavement joints. Therefore, the material used during construction of pavements should be carefully considered, so as to avoid anthropogenic soil environments that foster heavy metal accumulation (basaltic rock material with highly alkaline milieus). As pavement joints are mainly constructed with the function of water infiltration in sealed areas in mind, solution and transport of accumulated heavy metals poses a possible risk for the environment outside of urban areas. In addition to the direct risks of accumulated heavy metals (e.g., direct human contact, dust emissions in dry seasons) current research needs to pay more attention to this special field of urban soils. We encourage the following topics to be regarded as relevant for further research:

- More attention should be paid to pollution of pavement joints and urban soils in general, as these soils play a major role in urban environments, can react as pollutant accumulative soils and may pose a direct risk to humans.
- More research on urban soil pollution could enable the development of urban geochemical background values for different pollutants, which promote more effective risk assessments and spatial comparisons, even with pollution indices.
- Different sources of heavy metals besides traffic and transport in urban areas (e.g., surface runoff), need to be considered to develop effective management strategies of urban soil pollution.
- The role of the runoff must be examined more closely. Further studies about pollution concentration in drainage from pavement, and infiltration of drainage, are necessary. Not only on single areas, but with spatial (e.g., geospatial sampling approaches) and temporal (e.g., long-time studies, event-based sampling) resolution.

Data availability

Our research data is available in the following data repository: Weber, Collin, J.; Santowski, Alexander; Chifflard, Peter (2020), “Spatial variability of heavy metal concentration in urban pavement joints – A case study”, Mendeley Data, v1 http://dx.doi.org/10.17632/b3d66r56k8.1

Author contribution

Collin J. Weber has carried out the conceptualisation and selection of methodology. Collin J. Weber and Alexander Santowski performed the data curation, investigation and formal analyses. Peter Chifflard carried out the project administration and supervision and provided the resources. Visualisation and writing of the original draft were performed by Collin J. Weber with contributions of all co-authors. Writing during review & editing was performed by Alexander Santowski and Peter Chifflard.

Competing interest

The authors declare that they have no conflict of interest.

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