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Interactive comment on "Spatial variability of heavy metal concentration in urban pavement joints – A case study" *by* Collin J. Weber et al.

Anonymous Referee #2

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General comments Evaluation of the overall quality of the preprint

This manuscript focuses on the accumulation of various elements, including potentially toxic metals and one metalloid, in the joints of pavements in selected sites spread in Marburg city. The problem has not been in fact dealt with or described in soil science literature, and therefore the related data are - in my opinion - worth publishing. However, the presentation of such data should be accompanied by a thorough analysis and discussion. Unfortunately, the issue has been treated in this paper quite superficially, despite the fact that the text can make the impression of a deep scientific interpretation. This is, however, only the impression. After thorough reading the manuscript, I can see a lot of its shortcomings and errors. Moreover, the overall interpretation is in my opinion improper. The first issue is whether these joints can be called soil, which

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has already been raised by the editor. I myself have also doubts if it is soil, but the presented reasoning and conclusions can be considered a rational voice in the discussion. Of course, one could continue this discussion, mainly because the Technosols should generally show continuity with the rocks / parent rocks occurring underneath, while the pavements, especially the modern ones, are often placed on the completely impermeable layers equipped with drainage. However, if defining the soil as a living layer on the earth's surface, we can consider the joints a discontinuous soil layer. The authors should therefore first raise the issue whether the pavement joints make a soil or not, and then possibly move on to their position in soil classification. Regardless of whether it is soil or a questionable soil, I think the problem of accumulation of pollutants in the urban environment and a related risk assessment is worth tackling. In this manuscript, an added value is the data on concentrations of potentially toxic elements in the joints of pavements. However, the entire interpretation of the results seems to me wrong. Therefore, the results could be published after their re-interpretation. The main concerns are as follows: Firstly: the authors lump together various elements, including those that are the natural macro-components of rocks / soils / building materials: Fe or Al, and those that can be highly toxic, like Cd. The presence of As and Fe in soils is in fact of no importance in terms of pollution or risk. Moreover, their pseudototal concentrations (determined after the digestion in aqua-regia) are difficult to interpret. Secondly: the values of Igeo have been calculated in the manuscript with a purely technical procedure that should not be applied in this case. I am afraid, it would not be possible to calculate the values of Igeo index for the pavement joints. This index, proposed by Muller (1969) for river sediments, should be very carefully applied to soils. This matter was emphasized by Cai et al. in a quoted article (2015). The Igeo index has a geological meaning, and it can be applied to technically undisturbed, untransformed soils. It only makes sense to use it when there is a continuum: soil parent rock - soil. This index illustrates "geoaccumulation", which should be understood as the enrichment of geological material in comparison with its original state. In the case of pavement joints, their original "parent" material is unknown (e.g. cement, concrete, pure sand, gravely loams etc.) and most likely it differs among various sites. The authors proposed a very strange procedure to invent a “common reference material” that deal as a ”local geochemical background”. I do not think it can be justified to average the background values for soils from volcanogenic substrates (n = 94) and external sands (n = 64). Furthermore, the numbers of samples taken to this calculation (n = 94, 64) have not been explained. In my opinion, it also does not make sense to calculate for pavement joints such operationally defined parameters as the pollution load index (PLI) and potential ecological risk (RI), proposed by Hakanson. The authors maintain that "All three indices allow an effective assessment of contamination and the spatial differences (Kowalska et al., 2018; Cai et al., 2015). However, in the cited paper, Cai et al. highlighted various disadvantages of those simplified indices when they are applied to soils. Many authors have applied them indiscriminately to soils, indeed, and such works have been published, but this is not an argument that can justify such a controversial approach. The use of any of those indices should be preceded by their critical analysis and the study of applicability for a given case. While they can be sometimes used for the preliminary assessment of environmental risk, they cannot be used automatically and uncritically. The part of the manuscript that focuses on the PLI and RI indices is written incomprehensibly and it is hard to follow. The authors should try to be more concise. The methods of calculation and related input data have not been clearly presented. The corresponding threshold values have not been clearly shown either. The applicability of the indicators and their substantive sense have not been discussed at all. Based on the calculated values of the above indices, the authors drew conclusions regarding the risk assessment. However, they did not define the types of risk related to the accumulation of PTEs in the joints. Such an analysis should be a real basis for any interpretation of the results. The indices of ecological risk, proposed 40 years ago by Hakanson, were applied originally to the aquatic environment. They can be sometimes used as a simplified tool for very preliminary assessment of the risk associated with pollution of terrestrial environment, though a related kind of risk should be well specified. The risk analysis should take into account the potential human exposure pathways and the elements of possible risk to ecosystems (contaminated particles blown by wind, leaching of contaminants to groundwater, runoff and pollution of natural waters, other pathways of spread in the environment). Additionally, a surface area of pavements joints, exposure time of the target groups, potential infiltration rates (if any?), and the solubility of potentially toxic elements in joints should be taken into account and discussed to make the assessment of risk realistic. In relation to the last point, the importance of pH for the risk assessment should be clearly explained in the paper. The reasoning leads to conflicting conclusions regarding this problem. On Both acidification and alkaline pH have been reported as unfavorable factors. Summing up the overall assessment of the manuscript and my recommendations, I would suggest to narrow the interpretation of analytical data to what is in fact in the title, i.e. the concentrations of selected potentially toxic elements in the pavement joints. I would definitely remove the parts concerning Igeo and risk indices, as they are essentially unfounded and incorrect. Furthermore, I would not emphasize the analysis of spatial variability, and concentrate rather on initial recognition of the issue. Admittedly, the paper contains some elements of spatial analysis, though its statistical correctness and soundness raise my further doubts. I cannot see in which way this manuscript “improves 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” (L. 97-98). In my opinion this statement is too general and the manuscript does not contain clear indications for further usage of presented results. Specific comments Comments to individual scientific questions/issues My particular concerns are those related to the estimation of analytical uncertainty of experimental results (analytical errors). Although there is information that the results were obtained on the basis of triplicates (L. 164), it seems to relate only to the ICP-MS analysis of digests, and not to the differentiation of metal concentrations in “soil” within a given point. The low RSD values (L. 231-2) reported in the paper characterize the precision of instrument and not the heterogeneity of “soils” within a particular joint. Moreover, there is nothing in the manuscript about validation of analytical methods (us-

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age of certified reference materials?) Further, with regard to analytical uncertainty: it is completely unjustified and incorrect to present the concentrations of elements in soils with ridiculously high precision. Some data listed in the table 2 have 6 or 7 digits (Are they considered significant?). The number of digits should be reduced based on the assessment of uncertainty, i.e. either on the basis of minimum 3 separate soil replicates that characterize a given point, or on the basis of other available data on intrinsic heterogeneity of the tested material. Unfortunately, such an assessment may be difficult, because (as declared by the authors themselves) there are no available results of previous research on the pavement joints. On the other hand, however, the aspect of heterogeneity alone could be quite interesting. It would be advisable to list the related legal threshold values, that have been referred to, either in the manuscript itself or in the supplementary materials. I cannot find any comprehensive list of thresholds. Single values were given in the caption to Fig. 3, however they seem to be chaotically collected. I would prefer having the completed reference values in a clearer form (for instance in a separate table). Presumably, these values have been taken from the quoted BbodSchV act (Bundesregierung: 1998). It would be nice if the authors could confirm that these threshold values have not changed since 1998 and that they are currently valid. The article mentions “preventative” values for various forms of land use (L. 236-243), but the related data not been reported. Maybe this information is available in references, but the text cannot be understood without clear defining of terms used. Similarly: a term "absolute concentrations of heavy metals” appears several times in discussion (eg. L. 324). This term is unclear; it should be defined. According to the text (L. 86-88), “pollution retention capability of pavement joints has mostly been determined in laboratory tests and not in the field”. This statement was based on two bibliographic sources. The related data can by no means be generalized. This issue should be discussed in more depth, as the retention capability of pavement joints can undoubtedly be very diverse, and it would govern the environmental effects caused by enrichment in metals (surface runoff vs. infiltration). This aspect seems important and should be taken into account more carefully when interpreting the data. What did the

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authors understand as urban soil stratigraphy (L. 144)? Stratigraphy deals with natural rock layers (strata) and their layering (stratification), mainly in the studies of natural sedimentary and other layered rocks. I do not think this term is applicable to human made materials. The calculations of Igeo do not make sense as it is impossible to apply “the respective background value” (L. 178) The calculation of PLI was poorly explained. PLI was calculated as the square root of all multiplied single pollution indexes (L. 178), but no clear explanation was given what these single pollution indices are. The terms related to various elements should be used precisely. For example, As is not a metal but a metalloid (L. 159) and Al is not a heavy metal (L. 232). In the discussion, the authors introduced (based on their results?) the division into mobile and immobile elements (L. 395-6). This division is inconsistent with the generally known susceptibility of contaminants to mobilization in soils. Pb is known as a metal with very strong adsorption affinity and low mobility in soils. Such a grouping should be confronted with literature and discussed. One of the final conclusions, implying that basaltic rock material should be avoided as the material used for construction of pavements because of its alkaline pH and capacity to accumulate heavy metals, seems completely unjustified, particularly without a comprehensive discussion on risk exposure pathways. A list of references contains 13 items (23%). in German. I would suggest removing some of them and replacing them by English literature. If an aspect of risk assessment is to be included in the work, the literature should be supplemented with related sources

A list of technical corrections, typing errors, etc. L. 42, 83 singular “concentration” should be replaced by plural "concentrations”. L. 66 – Reference: Burghardt 1995 cannot be used as the reference for 2015 WRB-FAO classification L. 80-84 Unnecessary (obvious) references: (Sorme and Lagerkvist, 2002; Sansalone et al., 1998), (Gilbert and Clausen, 2006; Wessolek et al., 2009). L. 86 (Dierkes et al., 1999; Dierkes et al., 2004; Dierkes et al., 2005a) can be simplified: (Dierkes et al., 1999; 2004; 2005). L. 91 a phrase “contain an accumulation of heavy metals: should be reworded L.157 - “Ph value” should be corrected. I would suggest replacement by “The pH value” L. 159 – “pseudo-total concentration .... was” should be replaced by “pseudo-total concentrations .... were” L. 160 A term "extraction” with aqua regia should rather be replaced by “digestion”. Moreover, if a mass of soil sample is recorded (1g), a related volume of aqua regia should also be given. Or both data may be omitted. L. 161 each sample. It should be specified: each soil sample or each digest sample? The heterogeneity of soils is much larger than that of liquid samples. Definitely, solid samples should be analysed in triplicate. L. 192-3 rSP – it can be guessed that this abbreviation stands for Spearman correlation coefficient, but it should be clearly explained. Section 3.1 . (L. 195-210) Repeated discussion on soil classification is unnecessary here and should be removed. This section should focus on concentrations of elements in “soils” as declared in the title of the paper The list of references should be improved. Presently, it is drawn up inconsistently. Journal titles have been sometimes given in full names, sometimes in abbreviations, capital letters should be used. Is there any difference between Dierkes et al. 2005 a and b? The description shows that it is the same source Figure 1 – test sites are shown in Legend but not in Figures (except for the Marburg in the map of Germany) Figure 2 should be, in my opinion, divided into 2 separate graphs as they present different issues. A graph a (Igeo) presents the data concerning various elements, while two other graphs (b, c) – refer to various sites. Red lines placed in the graphs should consistently have the same meaning within one figure. Figure 3 – the graph is unclear and difficult to follow. The comprehensive data of preventive thresholds should rather be described in a table and not in the Figure description.

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