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Metals in soils of children's urban environments in the small northern European city of Uppsala

Karin Ljung [a,*], Olle Selinus [b], Erasmus Otabbong [a]

[a] Department of Soil Science, Swedish University of Agricultural Sciences, PO Box 7014, SE-750 07 Uppsala, Sweden
[b] Geological Survey of Sweden, BOX 670, SE-751 28 Uppsala, Sweden

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Abstract

Metals occur naturally in soil, but contents are generally increased in the urban environment due to anthropogenic activities. The presence of elevated metals in soils of the urban environment has been recognized as an important source of metal intake in children and is linked to elevated metal levels in children's blood. Several metals have undesirable health effects, especially on children due to their still developing nervous system and small body volumes. Playgrounds are where urban children spend most of their time outdoors and are also where children most frequently come in contact with soil. Elevated contents of metals in playgrounds are therefore of great concern for children's wellbeing. This study investigates the soil metal content of 25 playgrounds located in different land use areas in urban Uppsala, Sweden's fourth largest city. Uppsala covers an area of approximately 100 km^2 and has a population of 136,000. The soil samples were analysed for 12 metals (Al, As, Cd, Cr, Cu, Fe, Hg, Mn, Ni, Pb, W, Zn) using aqua regia. Median metal contents were found to be 1.8, 3.4, 0.21, 32, 25, 2.5, 0.14, 494, 19, 26, 0.35 and 84 mg kg^{-1} soil for each of the above metals, respectively. The median clay content was around 20% while the organic matter content was measured by loss on ignition at a median of 8%. The land use areas included industrial land, the city center, road verges, natural land and former industrial land. The results showed that land use did not have the expected large influence on the total metal contents of the soils tested. The clay content together with the age of the site proved to be a more important factor. Sites with elevated clay contents had in general elevated metal contents, which were explained by the relatively high adsorption capacity of clay particles. The soils at sites where land use had not been altered since the 1800s had increased metal contents compared to playgrounds constructed in the late 1900s. The immobility of metals once they had entered the soil system was the reason for increased metal content in soils of old playgrounds. It was concluded that in cities with few internal pollution sources, the soil characteristics of the site and the time the soil has been on-site to accumulate metal residues become important factors in determining the soil metal content.

Keywords: Urban soil; Metals; Children; Land use

1. Introduction

Soil parent material imparts a natural range of concentration of metals in soil. In the urban environment, soils receive a higher load of metals than their rural counterparts from traffic and industries and, since metals are rather immobile once they reach the soil

* Corresponding author. Tel.: +46 18 67 12 45; fax: +46 18 67 27 95.

E-mail address: Karin.Ljung@mv.slu.se (K. Ljung).

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system, accumulation occurs resulting in levels being reached that can be harmful to humans, especially children, upon repeated exposure (Thornton, 1991). As urban areas are densely populated, good quality of urban soil is essential to the health of the urban inhabitants (Li et al., 2001). Children in particular are more susceptible to the adverse health effects of soil metal pollution due to their small body size, developing nervous system and high absorption rate. For example, the portion of ingested Pb that is absorbed in an adult's body is typically less than 5%, whereas it is as high as 50% for children due to their less developed gastrointestinal tract (Ziegler et al., 1978; Maddaloni et al., 1998). The Center for Disease Control and Prevention has established a blood Pb level of concern for children at 10 μg dL^{-1} or above (ATSDR, 1999). This value is also used as an international action level. In New Orleans, US, 29% of children aged 0.5–5 years had elevated blood Pb levels and in Johannesburg, South Africa, 78% of children aged 6–9 had blood Pb levels above the international action level (Rabito et al., 2003; Mathee et al., 2002).

Children also ingest more soil than adults via the hand-to-mouth pathway, where soil is ingested intentionally, or through unintentional ingestion when children put dirty hands and objects into their mouths (Thornton et al., 1994; Sheppard and Evenden, 1994; Lanphear and Roughmann, 1997; Schütz et al., 1997). Soil ingestion has been recognized as an equally important exposure route of contaminants to humans as water and food ingestion (McKone and Daniels, 1991), especially for children up to the age of six due to their hand-to-mouth behaviour (Mielke and Reagan, 1998). For Pb, inhalation is a minor exposure route, and since the elimination of Pb in water pipes and tanks, Pb from soil has become a major source of exposure (Filippelli et al., 2005; Laidlaw et al., 2005). Contaminated urban soils have been identified by several authors as a significant source of Pb exposure in children (Berglund et al., 1994; Lanphear and Roughmann, 1997). In a study on 2-year-old children carried out in the UK by Thornton et al. (1994), it was found that ingestion of dust as a result of hand-to-mouth activity accounted for 50% of a child's Pb intake. Likewise, in a study conducted on urban topsoil from New Orleans, a significant association was found between the blood Pb levels of children <6 years and the soil lead levels (Mielke et al., 1999).

Soil ingestion and soil metal exposure are also dependent on the time spent outside and the location of the outdoor playing facilities. While some children in urban areas have private gardens for outdoor activities, others are confined to public playgrounds. In Uppsala, where the current study was carried out, 85% of children aged 1–5 attend some type of day-care, while nearly all 6-year olds attend preparatory classes at either private or municipal day-care homes (Uppsala City Council, 2003). Children in Uppsala subsequently spend most of their outdoor time in the playground of their respective day-care center. In Sweden during the 1960s, an urgent need for day-care centers in urban areas arose as families moved from the countryside to the cities. In the 1980s it was discovered that the sites as well as the construction of the day-care centers from the 1960s were unsuitable for the health of children and employees. Some playgrounds had been built directly below power lines, where there was available space, and the materials used for the construction were causing allergies. This resulted in demolition of some day-care centers while at the same time an urgent need for day-care centers arose for another reason: People born in the 1940s in Sweden were the first generation to be given the opportunity to study at university level without regard to their families' financial situation. This resulted in many women working full-time in the 1970–1980s so that the need for more day-care centers became urgent. The location of day-care centers was often decided upon with regard to available space and accessibility for parents, and such centers were thus commonly situated either close to homes or close to the parents' place of work. Consideration was given to the traffic intensity and power lines in the area, but seldom to the degree of soil pollution, or the effect past and present surrounding land use, apart from traffic and industries, may have had on the degree of pollution in the ground cover (A. Ljung, pers. comm.). This was most likely due to lack of knowledge at that time of the effects that elevated pollution levels have on children, or of the degree to which past activities may have affected the soil quality.

The playgrounds sampled in the current study were chosen from different land use areas, in order to investigate whether land use affects the soil metal content. In a study of the Swedish capital of Stockholm and its surroundings performed by Linde et al. (2001), the present land use was found to be a more significant factor affecting soil metal contents than geographic distance to the city center. Soils on wasteland and city center soils showed elevated metal contents in their study, while Li et al. (2001) found elevated metal contents in Hong Kong's commercial districts with high traffic emissions, as well as in industrial areas. The areas of land use selected for this study were (1) natural land use, with no visual impact of human activity, (2) playgrounds located close to major roads, (3) playgrounds located in or close to the city center, (4) playgrounds located in industrial areas and (5) playgrounds located on plots where there has been polluting industrial activity in the past. Twenty-five playgrounds were sampled and analysed for the metals As, Al, Fe, Cr, Cd, Cu, Hg, Mn, Ni, Pb, W and Zn using aqua regia.

2. Materials and methods

2.1. Area description

Uppsala is Sweden's fourth largest city with a population of approximately 136,000, excluding the non-urban population, and covers an area of 100 km^2. Uppsala is situated 51 m above sea level and has a mean annual precipitation of 548 mm and a mean annual temperature of 5.9 °C, with a range of −30 °C to +34 °C. The city sits on mainly granite bedrock which in large areas is covered by postglacial clay of 0.5 to 3 m depth. In the city center along the River Fyris the clay is found at depths down to 25 m (Gretener, 1994). Uppsala is mainly an academic city with a large student population and its two universities constituting large employers. Uppsala does not have any large polluting industries and the main metal point sources located in Uppsala are the sewage treatment plant, the heat and energy plant, car washes and the air force base. Important diffuse sources include traffic and long-range atmospheric deposition of metals (Stock, 1996).

2.2. Selection of sampling sites

There are approximately 190 day-care centers in urban Uppsala (2005). In this study, 19 playgrounds attached to such day-care centers were selected for sampling, while 6 sampled playgrounds were located in public parks. The playgrounds were located in different land use areas including industrial land (I), natural land (N), city center (C), traffic (T) and land with discontinued industrial activity (D) and are presented in Fig. 1. The figure also shows the location of the main pollution sources within the city boundaries as well as the location of the Swedish University of Agricultural Sciences. The playgrounds located on discontinued industrial land were selected after review of a report released by Uppsala City Council, investigating discontinued polluting industries (Lönnberg, 2001). Day-care centers that were located on the same plot as former polluting industries were selected to represent this land use category and were given the abbreviation D. The previous activities on these sites include metal works, dry cleaners and chemical industry.

Few of the day-care centers located in the city center of Uppsala have their own playgrounds, but instead use the playground facilities of public parks. Soil from the playground areas of parks Vasaparken, Engelska Parken, Stadsträdgården and St Göransplan were sampled to represent the city center and were given the abbreviation C. One playground attached to a day-care center was also included in the city center category. Since Uppsala is mainly an academic city rather than an industrial one, the industrial areas in the city are limited and naturally do not contain many day-care centers or playgrounds. Only four playgrounds were found within this category and were selected for sampling. Two day-care centers with accompanying playgrounds were found in two of the main industrial areas, Boländerna and Fyrislund, while one site selected was on the same plot as the Pfizer AB pharmaceutical factory, on the outskirts of the city. A fourth playground was situated in a park bordering an industrial area with several mechanical workshops (Börjetull). The selection of sampling sites in the category of traffic (T) was based on information from a study on traffic intensities in Uppsala (Nayeri, 2002). Two such day-care centers were adjacent to the most intensely trafficked road in Uppsala, the E4 (28,000 vehicles per day). The other three playgrounds were adjacent to roads with traffic intensities of 14,000, 13,500 and 9000 vehicles per day. One of these was located in a public park. The fifth category was natural land and included playgrounds located in areas with little human impact on the ground cover and not near any point sources, such as in the outskirts of forests. Six playgrounds were sampled in this category and were selected through visits to all playgrounds that were located in representative areas.

2.3. Sampling and analysis

The sampling points at each playground were selected on-site by finding spots where the grass had been eroded due to repeated trampling, since that was where the soil was directly exposed to playing children. This was commonly in the vicinity of designated playing equipment; by swings and at the end of slides. No spatial pattern design for sampling was thereby used and no samples were collected from sandboxes. The sand in sandboxes is commonly replaced every other year, when it has become hard and no longer fulfils its purpose (B. Mäkinen, pers. comm.) and sandbox sand is therefore not likely to have elevated metal contents. The soil surrounding the play equipment in playgrounds is not commonly replaced, although this might occur when a new playground is being built.

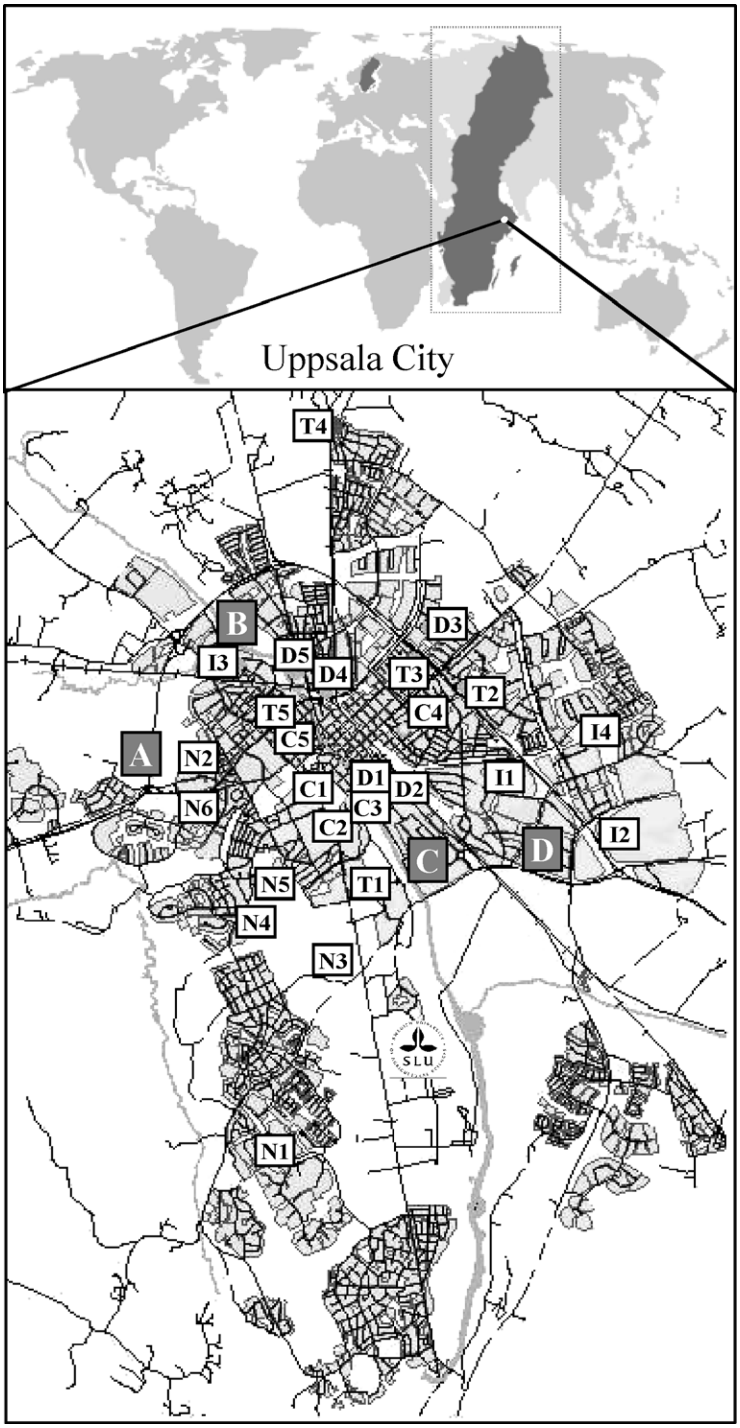

Fig. 1. Selected sampling sites throughout Uppsala. The letters represent land use categories (C=city center, I=industrial area, T=traffic, D=discontinued industrial areas, N=natural land). The location of the Swedish University of Agricultural Sciences as well as pollution sources is shown. The pollution sources are A—the crematorium, B—Börjetulls industrial area, C—the sewage treatment plant and the heat- and energy plants, and D—Boländerna and Fyrislund industrial area.

The year of construction for each playground was found through various sources: the City Planning Office, the Recreation and Nature Office of Uppsala City Council, through contacts with the day-care centers and through former employees of Uppsala City Council.

Soil sampling was carried out in October and November of 2002. Five playgrounds from each of the land-use categories traffic, city center and discontinued industry, four playgrounds from the industrial area category and six playgrounds from the category natural land were sampled. At each site, three sampling points were selected, giving a total of 75 sampling points. At each sampling point, five sub-samples from 1 m^2 were collected using a stainless steel auger and the soil cores were divided into depths of 0–5 cm, 5–10 cm and 10–20 cm. The five sub-samples from each layer were mixed giving three composite samples from each sampling point, in total nine samples from each of the 25 playgrounds. The samples were put in plastic bags and kept at 4 °C until preparation for analysis. The samples were ground and sieved to pass through a 2 mm sieve, after which dry matter was determined by oven-drying the samples at 105 °C overnight. Loss on ignition was determined at 550 °C and pH in H_2O (1:5) according to ISO (2002a). All samples were digested in aqua regia according to ISO (2002b). One blank and two reference samples were included in each batch of the digestion. The digestates were analysed for heavy metal concentration (Al, As, Cd, Cr, Cu, Fe, Hg, Mn, Ni, Pb, W, Zn) using ICP-AES and/or ICP/MD-DRC. The 0–5 and 5–10 cm depths from all three sampling points from each playground were combined for the loss on ignition and the mechanical analysis, which was performed using the pipette method (ISO, 2001), giving a total of 25 samples.

2.4. Statistical treatment

The data were tested for normality prior to further statistical treatment and it was found that non-parametric statistical analyses were best suited to the data set. The data were therefore displayed by medians, and comparisons made with ranked data. Data from the mechanical analysis and organic matter content were displayed by means, because of the lesser number of observations. For correlation analysis, Spearman's rank correlation was used, which presents a measure of the degree of linear relationship between variables, like the Pearson product moment correlation, but uses ranked data. Data from the 0–5 cm layer were used to investigate the correlations between metal contents and pH, while data from the 0–10 cm layer were used for correlations between metals, organic matter (LOI), clay, silt and sand contents. Mielke et al. (2005) used multiple metal accumulation (MMA) in order to compare the effect of the sum of metal accumulation on learning achievement in New Orleans elementary schools. In the current study, the contents of each metal from the top soil layer (0–5 cm) of the different playgrounds were ranked in order to standardize the values and ranked multiple metal accumulation (rMMA) was then used to identify the total metal distribution in the urban playgrounds. A cluster analysis was performed in order to identify sites affected by anthropogenic metal inputs. For comparison with Swedish guideline values, an average value of the three sampled layers was used to represent the metal content of the 0–20 cm layer. Minitab™ Statistical Software (2000) release 14 for Windows® was used for the statistical analysis.

3. Results

Table 1 presents the results from the mechanical analysis, pH and loss on ignition, together with the results from the metal analysis. Results are displayed for the whole data set as well as by land use categories and presented by means and medians. The soils of Uppsala's playgrounds were in general sandy clayey loams with low organic matter content and neutral to alkaline pH, in agreement with the calcareous soils of the Uppsala region.

When the general characteristics of the different land use categories sampled were compared, it was found that the pH was similar between the different land uses, while a higher organic matter content was found in the natural land use category. The organic matter content of the traffic category was lowest, while the remaining categories had similar organic matter contents. The discontinued industrial areas displayed markedly lower clay and silt contents than the remaining categories and a corresponding higher sand content. The remaining categories showed similar distributions of particle sizes.

3.1. Metal distribution

The distributions between the different land use categories were similar for most metals. The city center category had higher median contents of all metals except As and Hg than the remaining categories. The largest increases in this category were found for Mn, Pb, W and Zn, which were found at contents of 20%, 24%, 52% and 30% above the median of the entire data

Table 1
Mean clay (%), silt (%), sand (%) and organic matter content (LOI, %) for the 0–10 cm layer, and median pH and soil metal contents (mg kg^{-1}) for the 0–5 cm layer of the playgrounds in each land use category sampled

mg kg^{-1}	C	N	T	I	D	Median	Guideline values[a]
	$n=15$	$n=18$	$n=16$	$n=11$	$n=15$	$n=75$	
Al	2.07	1.98	1.78	1.90	1.36	1.84	–
As	3.40	3.27	3.72	3.90	3.33	3.40	15
Cd	0.249	0.225	0.214	0.174	0.167	0.214	0.4
Cr	37.5	33.0	29.9	31.6	25.4	31.6	120/5[b]
Cu	28.9	23.9	25.5	21.1	22.4	24.9	100
Fe	2.78	2.54	2.41	2.46	1.97	2.49	–
Hg	0.139	0.162	0.159	0.110	0.098	0.139	1
Mn	591	518	506	459	403	494	–
Ni	21.4	19.7	16.8	17.8	14.0	18.5	35
Pb	31.6	23.7	25.6	19.0	17.3	25.5	80
W	0.526	0.222	0.236	0.318	0.371	0.346	–
Zn	109	79.6	82.9	78.7	81.7	84.0	350
pH	7.4	7.2	7.1	7.1	7.5	7.4	
%	$n=5$	$n=6$	$n=5$	$n=4$	$n=5$	$n=25$	
Clay	24.4	23.9	23.2	24.2	14.8	21.6	
Silt	15.8	14.4	13.8	14.6	10.0	13.9	
Sand	52.3	52.3	56.5	53.6	67.8	57.2	
LOI	7.5	9.3	6.6	7.7	7.3	7.6	

C = city center, N = natural land use, T = traffic intense area, I = industrial area, D = discontinued industrial area.
[a] Swedish Environmental Protection Agency (Elert et al., 1997).
[b] The different values present soil contents with and without Cr(VI).

set, respectively. The highest median content for Hg was found in the natural land use category, which exceeded the median by 17%. This category also had elevated contents of Cd, Cr, Fe, Mn and Ni, although the increase was small (2–6% above the data set median). The industrial area playgrounds had the highest median content of As, which exceeded the median by 15%. The Al content of this category was found to be slightly above the median, while the remaining elements were found at contents below the median. The contents of As and Hg for the traffic category were elevated by 9% and 14% respectively, while only slight increases were found for Cu and Mn. In the discontinued industrial areas category, only contents of W were found to be elevated, by 7% compared to the median.

Strong correlations were found between soil texture classes (clay, silt and sand) and soil contents of As, Al, Cu, Fe, Ni, Pb and Zn, while none of the metals investigated correlated strongly with either pH or the organic matter content. The correlation with the above metals was positive for clay and silt, but negative for sand. All of the remaining elements except W also showed positive, although not strong, correlations to the soil texture classes. Tungsten differed from the other elements by also being positively correlated to pH, although the correlation was not strong. The most pH dependent element was Cd, with a Spearman's ρ of -0.599.

Since there was no clear distinction in metal content distribution between the land use categories investigated apart from the city center category, it was hypothesized that the surrounding land use was not the most important factor for the soil's metal content. To identify other factors determining the metal contents, multiple metal accumulations for each playground were calculated. In order to give equal weight to all metals, the data were ranked and the sum of all ranked values (ranked multiple metal accumulation, rMMA) was calculated for each playground. The results are depicted in the bar chart of Fig. 2, together with pH values and clay content at each site. Year of construction of each playground is displayed at the top of the chart and the site ID at the bottom on the x-axis. Three out of the four sampling sites of category I and four out of the five sampling sites of category D were found on the right-hand side of the chart, i.e. with an rMMA of less than 160. Likewise, four out of the five C sampling sites were found to the left in the chart with high rMMAs (>200). The sampling sites with low rMMAs generally had high W contents, whereas sampling sites with high contents of the remaining metals displayed low W contents.

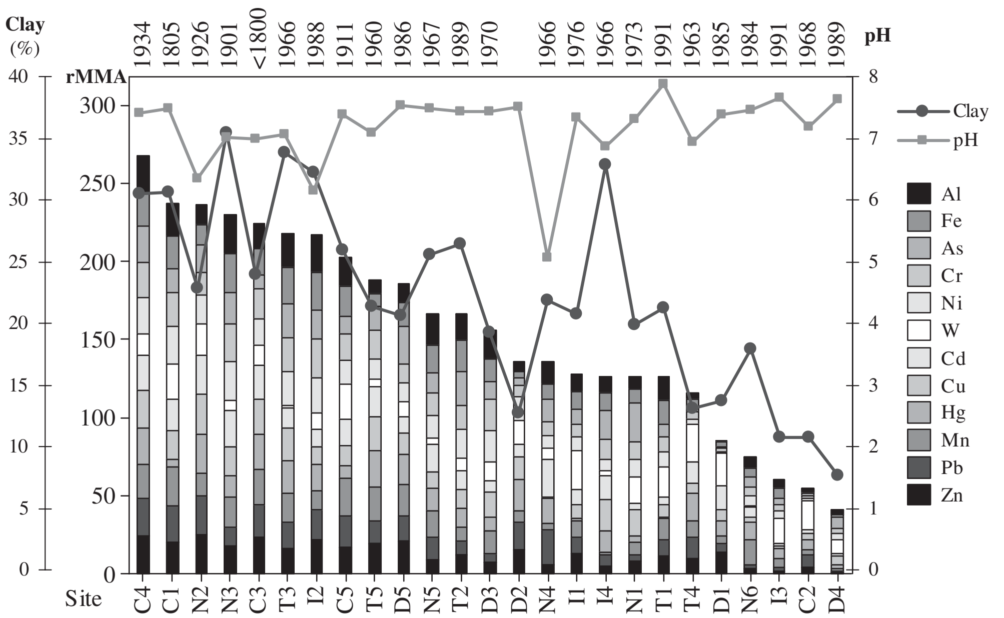

Fig. 2. Bar chart of the ranked (1–25) multiple metal accumulation (0–5 cm depth) for each site together with pH, clay content (%) and year of construction. The sites with the highest accumulated metal content appear to the left of the diagram (C = city center, I = industrial area, T = traffic, D = discontinued industrial areas, N = natural land).

No pattern was found in Fig. 2 that arranged playgrounds of the same land use category in clusters. Although most city center playgrounds were found to have an rMMA of above 200, one was found to have the second lowest rMMA of all playgrounds investigated. Likewise, the playgrounds located on natural land, expected to have low metal contents, were found to have both high and low rMMAs. There was a slight increase in pH with decreased rMMA, but the correlation was not strong. The clay content showed a strong correlation with the rMMA, increasing with increased rMMA, although a great variation was found.

Considering the multiple metal accumulations with regard to year of construction, it was found that the sites with increased rMMA were in general also the oldest. This was most apparent in the city center parks investigated. Four out of the five sampling sites from the city center category showed increased metal contents compared to the remaining sampling sites in Uppsala. All of these sites were situated in public parks that were founded 1800–1934. However, no metal contents were found at extreme values or at levels above the guideline values. The only city center site that was built during the latter part of the 1900s and that is not located in a public park (site C2) had the lowest rMMA (55) of the city center sites, with only contents of W exceeding the median. While the other city center sites had clay contents between 24% and 31%, site C2 had a lower clay content of 11%. The site with the lowest rMMA was located in an area that has accommodated a number of metal industries such as automobile workshops and metal works since the late 1800s. The area is today residential and the day-care center was opened in 1989 (City Planning Office, pers. comm). This site also had the lowest clay content of the sites investigated.

Only one site within the traffic category had an rMMA above 200 (site T3). This site is situated next to the most intensely trafficked stretch of E4 in Uppsala, with an average of 28,000 vehicles passing per day, and had contents of all metals except W and Cd above the median. However, site T2 was situated along the same stretch of road but showed lower soil metal contents (rMMA 166), with only Al, As, Fe, Cr and Ni above the median, none of these metals specifically related to traffic emissions. Both sites had similar clay contents. The former was constructed in 1966 and the latter in 1989. In the industrial area category, only one site (I2), constructed in 1988, had an rMMA above 200. This site had contents of all metals except Cd and Cu above median values. The neighbouring area is currently the site of a large pharmaceutical manufacturing company, Pfizer AB (formerly Pharmacia Upjohn). The site has housed pharmaceutical manufacturing companies since 1942 (R. Agius, pers. comm.).

The natural land use category was believed to have low metal contents since there was no visually apparent influence of anthropogenic pollution for these sites. Most sites were located in forest areas adjacent to residential areas. However, two of the sampled sites, N2 and N3, were found to have rMMA's above 200 (236 and 230, respectively), with N2 having contents of all metals above the median and extreme values of Cu, Cd, Hg and Pb. These four elements were above the guideline values at this site. However, extreme values of Cd, Cu and Hg were found at only one of the three sampling points at this site, while Pb contents showed extreme values at two sampling points. The second natural site with a high rMMA had contents of all metals except W and Pb above the median, but none exceeding guideline values. These two sites are both attached to old schools, built in 1901 (N2) and 1926 (N3). Two other sampling sites were located in the vicinity of the same forest as N3, but held markedly lower metal contents. These playgrounds were built in 1966 and 1967 and had clay contents of 22% and 26%, respectively, while N3 had a clay content of 35%.

Extreme values of As were found at sampling site N1. This site also had elevated contents of Cr, Cu and W. Out of the three sampling points at this site, only one had elevated metal contents. The playground was built in 1973 and is situated in a residential area in the vicinity of a forest. The clay content at this site was 20%.

3.2. Anthropogenic metals

In a previous study on the origins of metals in the playgrounds investigated here (Ljung et al., in press) it was concluded that the main origin of the elements Cd, Cu, Hg, Pb and Zn was anthropogenic. In order to distinguish between sampling sites that were affected by anthropogenic metals and those that were less affected a cluster analysis was performed (see Fig. 3). Two clusters were distinguished in the dendrogram. Cluster A included sites with low contents of anthropogenic metals, while the sites in cluster B held elevated contents of these metals. Site N2 was excluded from the analysis due to its extreme values but belongs in the group affected by anthropogenic sources. The sites in cluster A were all found to have rMMAs between 41 and 130 in Fig. 2 above, with the exception of sites T2 and D3. These two sites had rMMAs above 150 due to high contents of the natural elements Fe, As and Cr, Ni, respectively. In contrast, site T4 was found in cluster B but had an rMMA of 116, due to the site having high contents of anthropogenic metals in comparison to its contents of natural elements.

3.3. Guideline values

The Swedish Environmental Protection Agency has set up guideline values for soil contents of As, Cd, Cr, Cu, Hg, Ni and Pb to be used for risk classification of polluted soils (Elert et al., 1997). The values are presented in Table 1. The guideline values are set for soil sampled at 0–20 cm depth and digested with 7 M HNO_3. Therefore, an average metal content of the 0–20 layer of the sites investigated was calculated for comparison. Digestion with aqua regia is comparable to HNO_3 digestion for As, Cd, Cu and Pb, while HNO_3 digestion dissolves 17% less Ni (K. Lax, pers. comm.).

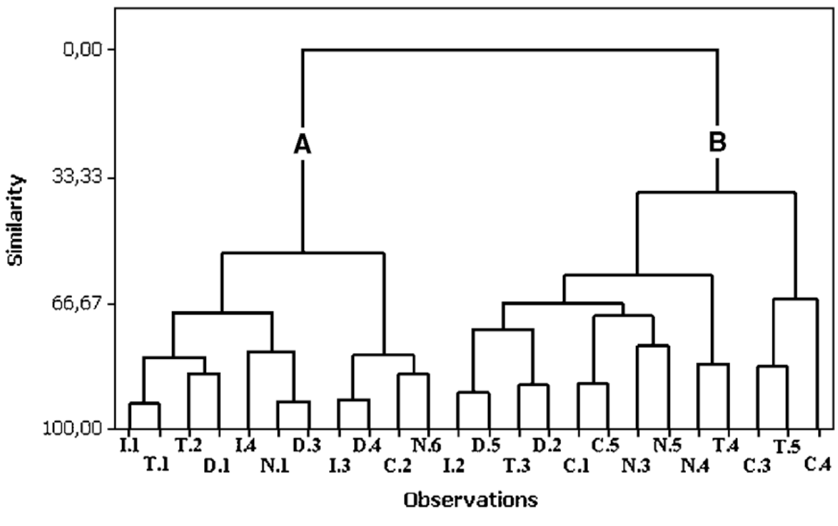

Fig. 3. Dendrogram of cluster analysis of observations; Cd, Cu, Hg, Pb and Zn (Complete linkage, Pearson's distance), showing (A) sites less affected by anthropogenic metals (Cd, Cu, Hg, Pb and Zn) and (B) sites with elevated contents of anthropogenic metals (C = city center, I = industrial area, T = traffic, D = discontinued industrial areas, N = natural land).

Five playgrounds had soil contents of one or more metals exceeding the guideline values (GV) for As, Cd, Cu, Hg, Ni and Pb. One school playground in particular (N2) had Cd and Pb contents of 1.5 times the GV, while Cu and Hg contents were 2 and 3 times the GV, respectively. It should be noted that out of the three sampling points within this playground, only two had elevated metal contents. At a playground in a public park nearby (T5), Cd and Hg were found at contents slightly exceeding the GV. One playground (N1) had contents of As just above the GV, while playgrounds D3 and N3 had contents of Ni slightly exceeding the GV.

4. Discussion

No strong correlations were found between metal contents and pH or organic matter content. However, soil clay content correlated strongly with the metal content, especially As, Al, Cu, Fe, Ni, Pb and Zn. The strong correlation can be explained by the small size of clay particles and the resultant relatively large reactive surface area and permanent negative charge, facilitating a high adsorption capacity for cations compared to larger particles. This relationship was also evident from the strongly negative correlation between metal content and the larger sand particles, since a high sand content indicates low clay content.

At pH values above neutrality the metal ion activity in soil decreases but the total ion concentration in solution increases, which renders metal ions more mobile because of complex binding to dissolved organic carbon (DOC). However, when there is an absence of DOC, a higher pH renders metal ions less mobile and the effect of higher pH is less marked. The low organic matter content of the soils investigated could explain the absence of correlations between metal content and pH or organic matter content.

The highest contents of organic matter were found in the natural sites, corresponding to the presence of more vegetation. Another factor relating to the low organic matter content of the sampled sites was the recurrence of excessive trampling. The sampling sites were selected where the vegetation was eroded and since excessive trampling disturbs soil organisms and their corresponding incorporation of litter into the soil, low organic matter content at these sites was expected. The relatively high sand content of the sampled soils compared to the original clayey soils of the city suggests that foreign soil has been applied to many of the sites. The playgrounds located in the parks of the city center had the highest content of small particles, and these were also the sites that had not been altered in recent times, suggesting that the soil at these sites is the original Uppsala soil. Tungsten was found to correlate to the general parameters in the opposite way to the remaining elements, and was elevated at sites with low contents of the remaining metals. The difference can be explained by W being an anion in soil solution, and thereby attracting differently charged ions than the remaining metals, which are cations. The pattern of increased W contents where low contents of the other metals were found indicates a stronger association between metal content and chemical conditions on-site, than with input from surrounding pollution sources in Uppsala.

4.1. Metal distribution

There was no distinct distribution pattern of soil metal contents between the land use areas investigated. The land use category that differed most from the remaining categories was the city center, where the playgrounds had elevated metal contents, especially of Mn, Pb, W and Zn, metals commonly associated with traffic emissions. Instead of land use dependence, the rMMA values showed a pattern of increased metal content with increased age of the site, where the playgrounds in the city center parks were the oldest. High rMMAs were also found at the two natural playground sites which were built in the beginning of the 1900s. Since metals are rather immobile once they reach the soil system, metals disseminated onto the surface in the past are still present today. Since the transport of metals in soils is more likely to be physical than chemical, the time the soil has been left undisturbed in the urban environment to accumulate metal residues becomes significant for its metal content. Three out of the four city center parks were not purpose-built on-site, but were rather kept as green areas while the city expanded around them. The fourth city center site with high contents of metals was formerly a site for clay extraction to brickworks, and it is likely that the site, once extraction discontinued, was filled up with waste material. Another factor possibly contributing to the increased metal content of city park soil is the application of pesticides and fertilizers. Although age seemed to be one of the most important factors, clay content also contributed to the distribution pattern, which is apparent from Fig. 2. The one city center site with markedly lower metal content also had very low clay content. This site was built most recent out of the city center sites. Although the sampling category “natural land” may suggest that these sites are the oldest, the playgrounds within these sites were built in more recent times, and the category name only implies that the sites are not located near any point sources. Two playgrounds of this category were built in the beginning of the 1900s, whereas the remaining playgrounds were built after 1967. The oldest playgrounds had the highest rMMAs, further supporting the theory of metal accumulation over time, since it can be assumed that foreign soil has been added during construction of the more recent natural land playgrounds.

The clusters in Fig. 3 separated the sites with rMMAs above and below 160, with those having rMMAs above 160 representing sites with high contents of anthropogenic metals, with a few exceptions. The sites with elevated contents of anthropogenic metals, appearing in cluster B, also had elevated contents of the natural elements, explaining the similarity in distributions between the cluster analysis and the rMMA. This indicated that the metal content of the sampled sites was not mainly determined by point pollution sources, which would be apparent from a metal distribution by land use, but was more dependent on the soil characteristics on-site. Only one site was found to have elevated contents of anthropogenic metals but a low rMMA. It was located at the immediate side of a road, with the playground being built on a slope receiving runoff from the road. Although this site did not have any elevated contents, its main source of metals is traffic since there are no other sources nearby. This could explain its elevated contents of anthropogenic metals, but low contents of the remaining elements.

Although other authors have found land use to be a significant factor in the degree of soil metal contamination, it should be noted that Uppsala is mainly an academic city with few point pollution sources. In fact, its main pollution source by far is atmospheric deposition. The age of the land and characteristics of the soil on-site may therefore be more significant than land use in cities without major internal pollution sources.

4.2. Extreme values and guideline values

Elevated contents above the guideline value of Cd, Cu, Hg and Pb were found at one site located in the vicinity of a forest on the western outskirts of the city, but relatively close to a crematorium, known to release the above metals, particularly Hg. The crematorium is located about 2 km NW of the site and has been the largest source of Hg in Uppsala, emitting approximately 8.5 kg of mainly metallic Hg into the air annually up until the year 2000. At the present time, the release of Hg has decreased to 0.5 kg annually (R. Örnestav, pers. comm.). It is likely that the other elements also originate from the gaseous emissions of the crematorium, since there are no other sources nearby. The school was founded in 1909 and the crematorium in 1965 (K. Hedin, pers. comm.) It is important to note, however, that the extreme values were only exceeded at one of the three sampling sites for all elements except Pb. This sampling point was situated closest to the school building and it is possible that the soil at the two remaining sampling sites was altered during renewal of parts of the playground. The pH value at this point was also lower than at the two other sampling points at this site. One additional sampling point had elevated contents of Pb, but not exceeding the guideline value.

The guideline value for Ni was exceeded at a playground also located in the vicinity of a forest and in connection with an old school, dated 1901. There are no point sources nearby but the highest clay content of all sites investigated was found here. Apart from Ni, elevated contents of Al, Fe and Cr were also found here, although not exceeding any guideline values. A strong significant correlation was found between these four elements and the clay content of the soil, implying that soil clay content is of relevance in the absence of point sources. A third playground had As contents slightly exceeding the guideline value, but only at one sampling point. Elevated contents of Cu, Cr and W were also found at this site. Arsenic has previously been used in impregnation of wood for outdoor uses. In addition to As, impregnated wood is known to leave residues of Cr and Cu in the soil. This type of wood is no longer in use at playgrounds in Uppsala, but it is possible that it has been in the past, with residues left in the soil.

5. Concluding remarks

Current land use in Uppsala did not seem to be a major factor in determining the soil metal content at its playgrounds. Although the city center sites in general had higher metal contents than remaining land use areas, the age of the land seemed to be a more significant factor. The clay content also had a marked effect due to its relatively high adsorption capacity. The soil cover at the park and forest sites investigated with enriched metal contents has not been renewed, in comparison to playgrounds attached to day-care centers built during the latter part of the 20th century. Since Uppsala is not an industrial city and does not have many sources of metal pollution, the time the soil has been in place in the urban environment to accumulate metals seems to become more significant than surrounding land use.

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