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ORIGINAL PAPER

Spatial variation of contaminant elements of roadside dust samples from Budapest (Hungary) and Seoul (Republic of Korea), including Pt, Pd and Ir

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Abstract Roadside dusts were studied to explain the spatial variation and present levels of contaminant elements including Pt, Pd and Ir in urban environment and around Budapest (Hungary) and Seoul (Republic of Korea). The samples were collected from six sites of high traffic volumes in Seoul metropolitan city and from two control sites within the suburbs of Seoul, for comparison. Similarly, road dust samples were obtained two times from traffic focal points in Budapest, from the large bridges across the River Danube, from Margitsziget (an island in the Danube in the northern part of Budapest, used for recreation) as well as from main roads (no highways) outside Budapest. The samples were analysed for contaminant elements by ICP-AES and for Pt, Pd and Ir by ICP-MS. The highest Pt, Pd and Ir levels in road dusts were found from major roads with high traffic volume, but correlations with other contaminant elements were low, however. This reflects automobile catalytic converter to be an important source. To interpret the obtained multi-element results in short, pollution index, contamination index and geo-accumulation index were calculated. Finally, the obtained data were compared with total concentrations encountered in dust samples from Madrid, Oslo, Tokyo and Muscat (Oman). Dust samples from Seoul reached top level concentrations for Cd–Zn–As–Co–Cr–Cu–Mo–Ni–Sn. Just Pb was rather low because unleaded gasoline was introduced as compulsory in 1993. Concentrations in Budapest dust samples were lower than from Seoul, except for Pb and Mg. Compared with Madrid as another continental site, Budapest was higher in Co–V–Zn. Dust from Oslo, which is not so large, contained more Mn–Na–Sr than dust from other towns, but less other metals.

Keywords Contaminant elements · Pt · Pd · Ir · Road dusts · Urban environment

Introduction

In urban areas, various human activities lead to the formation of suspended aerosols and deposition of dust, of which the composition is different to the geochemical background. From this, there have been concerns about health risks for centuries and also significant changes of the composition of urban soils and downstream sediments from runoff and sewage.

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Permanent changes in the technologies for house construction, traffic and heating during cold seasons, as well as changes and improvements in the purification of industrial emissions, necessitate repeated monitoring of the composition of urban dust. Within a first step, representative sampling grids have to be established to find the hotspots of contaminations, prior to rather laborious and expensive size-fractionated sampling and microlocal analysis, which is the goal of this paper. Contrary to direct air dust sampling, deposited dust yields integrated contamination values over a long period of time, usually back to the last rainfall or street cleaning action. In addition to chemical toxicity, however, some particles promote catalytic reactions at their surface, which is not traceable by chemical analysis only. Direct health implications are due to the inhalable fraction, which is rather floating in the air than depositing at surfaces. Deposited dust directly imposes health hazards via consumption of unwashed food for dogs, which like to lick and sniff at dusty surfaces, and finally, the dust washout might harm green plants in parks, as well as river systems.

Many studies on the anthropogenic emissions, in particular, from automotive catalytic converters, the pollution levels and the evaluation of the platinum group metals (PGEs; Pt, Pd and Ir, etc.) have been undertaken using road soils and deposited dusts, and airborne particles (for example, Merget and Rosner 2001; Gomez et al. 2002; Ravindra et al. 2004; Rauch and Morrison 2008; Reith et al. 2014). Zereini and Alt (2000) edited an excellent comprehensive book on the anthropogenic platinum group element emissions and explain their impact on man and environment. Gomez et al. (2002) reviewed the levels and risk assessment for humans of PGEs in airborne particles and dusts of some European countries. Others also studied on environmental impact and relevance of PGEs and traffic-related elements, for example, Zereini et al. (2001) in Frankfurt am Main, Germany, Leopold et al. (2008) and Prichard et al. (2009) in Sheffield, UK, Pratt and Lottermoser (2007) in Cairns, Northern Australia, and da Silva et al. (2008) in Rio de Janeiro, Brazil.

Within this work, urban road dust from two capital cities, Budapest in Hungary and Seoul in Korea, has been analysed for main elements and a lot of metals, including Pt, Pd and Ir. Budapest and Seoul are substantially different in many aspects, like population density, car park and car driving, heating devices in winter time, industrial emissions, distance to the sea and geochemical background.

In Budapest, there live about 2.5 million inhabitants at 525 km^2, whereas in Seoul, there live 10.5 million inhabitants at only 605 km^2. In Budapest, cars move mainly with manual gears, and a substantial amount goes diesel powered, particularly the trucks, like usual in overall Europe, whereas in Seoul, most cars move gasoline powered with automated gears. Geologically, Budapest is situated at the edge of a basin filled with Tertiary and alluvial sediments; just some hills in the north-west direction are crystalline base rocks. The closest distance to the sea is 450 km, and this is the Adriatic Sea at Trieste, but influence from there is marginal. Seoul centre is about 37 km west to the sea, but due to the position of neighbouring Inchon harbour city, the area towards the coast is almost continuously inhabited. Seoul is placed on alluvial sediments of the Han River and surrounded by granite and gneiss hills.

Comparisons between road dust samples from these largely different urban areas might reveal common global urbanization effects and specific rather local influences. This can be ascertained from additional data per gram dust, available from other densely populated areas. The effect of urbanization might be traceable from differences between road dust samples from Budapest and from more rural Hungary.

Most of air pollution monitoring and studies available from the literature give data about concentrations in air and thus target on inhalable hazardous contaminants. In case the input to arable land is asked for, it is appropriate to consider deposited amounts per area. For reasons of simplicity, within this work, total dust was sampled without further grain size fractionation as a snapshot and analysed for as many elements as easily possible, to serve as a pilot study. In the future, a subset of elements for a more detailed investigation might be selected.

Metal contamination levels in soils and dusts of Seoul metropolitan city have been already published elsewhere (Chon et al. 1995, 1998). In addition, Lee et al. (2006, 2012) investigated the levels of Pt pollution in road dusts, roadside soils and tree barks sampled in 2005. The hotspots of Pt occurrence were found at major roads with high traffic volume, in particular corresponding to a high number of stop-and-go events. Combined chemical composition and scanning microscopic results identified automobile catalytic converters as Pt source. Within this work, additional samples taken in Seoul during summer time 2010 were analysed, as well as samples from Budapest, which might be contrasting Seoul in many ways. A suitable method was developed to analyse for Pd also, which needs separation from the matrix because of Sr and Y oxide interferences on the Pd-mass in the ICP-MS.

Experimental methods

Sampling

Road dust samples were obtained two times from traffic focal points in Budapest, from the large bridges across the River Danube, from Margitsziget (an island in the Danube in the Northern part of Budapest, used for recreation) as well as from main roads (no highways) outside Budapest. Similarly, road dust and roadside soil samples were collected from six sites of high traffic volumes in Seoul metropolitan city and from two control sites within the suburbs of Seoul, for comparison.

Road dust samples were collected by hand brushing with a nylon brush and plastic collection pan directly from the road surface. The road dust samples were dried at 80 °C for 40 min and then sieved in to 0.075 mm (−200 mesh) fraction.

Analysis

The level of Pt and other heavy metals in road dusts and roadside soils was analysed after acid digestion. Two grams of each road dust and roadside soil sample was weighed into a glass vessel equipped with a reflux condenser (behr Labor Technik GmbH Duesseldorf, Germany), to which 21 ml HNO_3, 7 ml HCl (reverse aqua regia) and 20 µl elemental bromine were added and the vessel left overnight. Then, it was digested for 30 min at 60 °C and for further 90 min at 140 °C in a heating block. Following digestion, samples were cooled to ambient temperature and filtered through black ribbon paper filters (100 S & S Rundfilter). Samples were then diluted with distilled water up to 100 ml.

For ICP-OES measurement, samples were measured diluted undiluted, 1 + 4 diluted, 1 + 24 diluted, and more, if necessary, in order to the optimum calibration range, and to see matrix effects. For ICP-MS measurement, the samples were diluted 1 + 4, and In/Rh was added and measured as internal standards. Pt–Pd–Ir–Au–Hg–Tl–Bi as well as Hf, Ta and Sr were read. Pt was manually corrected for Hf–O and Au for Ta–O. For Pd, correction would reach 80–100 %, and therefore, separation was necessary.

The separation procedure was modified after Jackwerth and Willmer (1976). Twenty millilitres of digest was mixed with 20 ml 2 M-HCl in an Erlenmeyer flask and evaporated to a few millilitres on a boiling water bath in order to expel the nitric acid and nitrous oxides. Then, 2 M-HCl was added, and finally, 2 ml 10 % Na-ascorbate and 2 ml 0.2 % dithizone were added in acetone. The dithizonates of Pt, Pd, Hg and Cu get slowly co-precipitated with excess reagent with dark green colour. The reagent is almost insoluble in aqueous acids, and thus, excess reagent is utilized as a scavenger. After standing overnight, the precipitate was filtered through paper and altogether digested with 4 ml HNO_3 suprapure in closed vessels by microwave-assisted heating. The digest was made up to 10 ml and measured by ICP-MS by the same program as above. As a control, if the precipitation was complete, the values obtained for Pt in the samples must be about equal to the result from the original sample solution, and there are no interferences from oxides.

Validation of the method was finally performed by analysis of the reference material BCR-723 (urban dust) for Pd, Pt and Ir, as well as for main and trace elements by regular participation in the IPE program (International Plant Exchange) of Wageningen University (The Netherlands) and the ALVA (Austrian Association for Food, Veterinary and Agricultural Items) for soils and feeds.

Results and discussion

Geochemical characteristics

Beneath known contaminants, Table 1 also contains information about main elements encountered. Because data in this work refer to digests in reverse aqua regia, comparability with other data is rather qualitative. Whereas carbonatic, oxidic, sulfidic and organic phases are usually recovered completely, it is well known that much of the silicate matrix remains in

Table 1 Comparision of the elements concentration in urban dust from Budapest and Seoul with other cities. Results of Madrid in 1990 and Oslo in 1994 (de Miguel et al. 1997), Tokyo (Furuta et al. 2005), Muscat in Oman (Yaghi & Abdul-Wahab 2004), Budapest and outside of the Budapest in 2010 (Kadar and Marton 2012) and Seoul in 2010 (Lee et al. 2012)

	Madrid (1990)	Oslo (1994)	Tokyo (2005)	Muscat (2004)	Budapest (2010)		Seoul (2010)	
					Range	Mean	Range	Mean
g/kg								
Al	43.8	59.5	50.7		4.4–10.8	8.5	13.9–22.1	17.9
Ca		42.7	50.6		22.4–87.3	67.6	31.2–58.6	44.6
Fe	19.3	51.5	47.4		15.3–87.5	24.8	25.7–37.9	33.9
K			12.9		0.50–3.92	2.18	4.4–7.9	7.4
Mg		13.2	13.5		3.9–17.7	12.7	5.9–9.3	7.8
Na		19.3	15.8		0.24–3.82	1.81	1.6-7.6	3.4
P		1.1			0.45–1.47	0.89	0.75–3.44	1.36
Ti			3.83		0.1–0.2	0.1	0.3–0.7	0.4
mg/kg								
As			5.9		8.2–15.8	11.6	15.2–31.2	24.4
Ba		526	530		68–961	353	217–716	523
Bi					0.3–1.8	0.7	1.6–6.3	2
Cd		1.4			0.31–1.89	0.83	1.4–6.7	3.5
Co	3	19	21.8		3.7–12.6	8.1	15.4–74.3	17.9
Cr	61		649	17–171	37.4–120.6	65.5	104–195	130
Cu	188	123	574	7–247	144–352	236	302–478	351
Hg					0.02–0.30	0.13	0.27–0.58	0.44
Li			156		5.5–21.7	13.9	25.0–43.0	29.5
Mn	362	833	814		345–1,011	417	541–681	639
Mo		4	14.7		0.3–8.8	4.4	6.7–18.6	13.7
Ni	44	41	540	43–3,033	19.2–49.9	27.5	42–109	62
Pb	1,927	180	257	20–288	49–1,891	408	130–284	214
Sb		6	31.5		1.1–20.4	8.4	10.0–60.1	44.3
Sn					3.3–31.0	14.7	21.1–50.4	39.7
Sr		344			47–166	111	69–113	90
V	17		98		14.7–29.6	25.1	23.4–43.2	34.7
Zn	476	412	1,610	3–2,333	317–2,110	891	1,075–2,065	1,476
µg/kg								
Pt					1.4–154	36.2	25.6–98.5	65.6
Pd					8.3–187	50	30.6–148	101.7
Ir					0.3–1.5	0.6	2.1–5.6	3.3

Data for Al, Fe, K, Mg, Ti, Ba, Cr, Li and V might be lower than total in this work because of incomplete dissolution in reverse aqua regia

the residue filtered off, and standardized conditions (boiling time and temperature) are needed to get reproducible results for the partially weathered silicates. Higher levels of Al, Fe and K indicate the acid granite rocks surrounding Seoul, as well as more Na due to the proximity to the seaside. Alkaline soils around Budapest tend towards more Ca and Mg, though these can derive from abrasion of buildings also. This leads to different estimated background levels used for the calculation of the contamination index (CI) (see below).

Platinum and palladium

Platinum occurs associated with Ir, Os, Pd, Ru and Rh, usually below 1 µg/kg (Ward and Dudding 2004). 0.4 µg/kg for both platinum and palladium is widely regarded as the geological background. Platinum and Pd concentrations encountered in urban dust samples usually show asymmetrical frequency distributions with a few high and many low values, but for this work, sampling was done mainly at presumable hotspots. In both cities, strong correlations between

Pt and Pd concentrations in street dust appeared because of the automotive catalyst as the common source. In spite of about four times higher population density in Seoul, Pt and Pd levels at traffic hotspots were about equal, whereas along main roads in the suburbs of Budapest, lower levels were encountered. In Seoul, Ir was significantly higher.

In Budapest, maximum Pt levels were found in dust collected at the central bridges across the River Danube, where urban traffic jams are focussed and more wind can be expected. Note the low level at the most northern bridge Margit Hid, which is close to a recreational area at Margitsziget (Margit Island). Pd maxima were rather met at the big squares in the east of Budapest.

For platinum group metals, catalyst materials used in cars are currently regarded as the main emission source. However, palladium emissions from industry cover a larger part than platinum, compared with emissions from traffic (Dirksen et al. 1999). Platinum metals may also be emitted from jewellery production, and gold–palladium and palladium-based alloys have been used in dental laboratories and surgeries, which enter the urban dust as abrasion from pedestrians (Helmers et al. 1998; Jackson et al. 2010).

Iridium and ruthenium are not constituents of the 3-way catalyst, and they originate from noble metal-processing industries (Dirksen et al. 1999). For municipal sludge in England, it is unlikely that Ir, Ru and Os originated from automobile catalysts (Jackson et al. 2010). But in addition to Pt–Pd–Rh, the use of Ir as a novel active metal in catalytic converters ought to be reflected in road dusts and roadside soils in the future (Fritsche and Meisel 2004).

Hazardous metals and metalloids

Beneath Pt and Pd, multi-element methods permit the determination of a lot of other hazardous metals and metalloids from the same digest. This enables to establish possible relations between the rather new contaminants Pt and Pd, with others items investigated for a longer period of time.

Main anthropogenic sources of urban dust are combustion processes (heating in winter, cooking), industrial emissions, abrasion of buildings and streets, and traffic. Some metals can form volatile oxides and halogenides during combustion (Sager 1999), leading to condensation aerosols till deposition and adsorption at dust particles. Note that all samples were taken during the non-heating period.

Antimony

Whereas Sb levels in road dusts around Budapest were met at about background levels, some accumulation in the city occurred. In Seoul dusts, levels were higher throughout. Note that the sampling site at the SNU (Seoul National University) is rather calm without traffic jams. Note that for the upper continental crust, an As/Sb proportion of about six can be expected (Wedepohl 1995), which is widely met in soils and sediments also (Sager, unpublished).

Enrichment of antimony in dusts has been observed in many countries, even in Arctic ice. Due to health risks, asbestos brake parts have been generally replaced by nonasbestos organic type pads. Within a model testing series, each deceleration event caused emissions of 34 µg Sb, assuming a car of 2,000 kg running at 40–60 km/h. Higher disc T caused emission of coarser particles, i.e. 0.8 µm at 100° and 2.0 µm at 400°. Abrasion dusts from brake pads contained 1.5 % antimony (Iijima et al. 2008) and also 3.1 % Fe, 15 % Cu, 1 % Zn, and 12 % Ba. Sb is a component of brake linings and a flame retardant in vulcanization of rubber (Fujiwara et al. 2011).

Arsenic

In Budapest street dust, arsenic was at background levels expected for soils, and in Seoul, it was only slightly elevated. Fossil fuels, in particular coal burning, are main sources for As in dust, but there are also some emissions from metal processing (Ajmone-Marsan and Biasoli 2010). At least in the USA, Pb from wheel weights contains 5 % of As (Hays et al. 2011).

Barium

For soils, total digestion might yield about 4 times higher Ba concentration than aqua regia, and thus, reference data should be read with care. Nevertheless, Ba met in street dust was higher than expected for agricultural soils (in aqua regia). For Seoul, no background level for Ba was available, and therefore, contamination levels cannot be classified. Abrasion dusts from brake pads contained 12 % barium (Iijima et al. 2008). Microanalysis of single dust grains sampled in Prague city revealed that the abrasion of brakes produces coarse aerosol particles of high Cu–Ba–Fe–Zn, and Sb–K–Ti. Sometimes $K_2O.nTiO_2$ is used to improve the thermal resistance and to lower the brakes wear (Ondraček et al. 2011).

Cadmium

Whereas in Budapest dusts Cd was slightly above soil levels, Seoul dusts contained more and obviously not correlated to traffic intensity. In urban areas, main cadmium sources are Ni–Cd batteries, electronics, photography, pigments and plastics (Ajmone-Marsan and Biasoli 2010). Phosphate minerals and emissions from smelters might be less important. Cd tends to accumulate in roadside dust soils, plants and trees (Panwar et al. 2007, 2010, 2011).

Chromium

Total digestion yields about three times more Cr than aqua regia, but the recovery from organic soils and composts can be higher. The background value in Table 2 refers to aqua regia. Chromium is used in metallurgic and galvanic industry, and is a likely component of engine metal alloys (Hays et al. 2011). Motor vehicle exhaust and waste incineration have been regarded as the main sources for urban dust (Ajmone-Marsan and Biasoli 2010).

Copper

Copper is likely recovered from any digest and found remarkably enriched in the samples investigated. Electronic waste and electrical wearing (Ajmone-Marsan and Biasoli 2010) are well known for copper, but also abrasion dusts from brake pads contained 15 % copper (Iijima et al. 2008).

Lead

With respect to mean crust and mean soil values, Pb was found significantly enriched in street dust; it was the only element which was higher in hotspots of Budapest than in Seoul. After cease of using leaded gasoline, main sources are brake wear and the loss of Pb wheel weights (Fujiwara et al. 2011; Hays et al. 2011).

Manganese

Manganese was found at about ambient levels. In urban areas, main manganese sources are fertilized agricultural organic soils around the cities for horticultural and crop production. Mn tends to accumulate in roadside dust soils, plants and trees (Marton 2012; Marton et al. 2011; Petr et al. 2011).

Molybdenum

Enrichment of Mo in dust samples was significant. Molybdenum has been used as a component of brake linings (Fujiwara et al. 2011).

Nickel

Though recovery of Ni is not complete from aqua regia for all sample types, concentrations met in the dust samples were not much elevated. Nickel is a component of stainless steel and thus present in engine metal alloys, but it has been also used as a tracer for oil combustion. Apart from this, appreciable amounts are found in Ni–Cd batteries (Hays et al. 2011; Ajmone-Marsan and Biasoli 2010; Fujiwara et al. 2011). In the Oman, higher nickel levels in outdoor dust have been traceable to enrichments in adjacent serpentinite rocks (Yaghi and Abdul-Wahab 2004).

Sodium

Mean Na concentration in street dusts seems to decline with increasing distance to the seaside. Thus, Na in Oslo dust was higher than in Seoul dust, and Na in both were higher than in Budapest dust.

Zinc

Zn is usually completely recovered in aqua regia, and it is highly enriched in urban dust and can come from many sources. Zinc is widely used in the metallurgic and galvanic industry, as well as for tires, batteries, electronic equipment and alloys (Ajmone-Marsan and Biasoli 2010). Traffic-related sources in particular are tyre rubber, brake pads, safety fences and oil additives for wear protection (Fujiwara et al. 2011). Abrasion dusts from brake pads contained 1 % zinc (Iijima et al. 2008). In the USA, Zn is used in oil additives and is a major component of galvanized metals and many paints

Table 2 Results of urban dust samples and soil background values

	Budapest		Outside Budapest		Seoul		Background (BG)	
	Range	Mean	Range	Mean	Range	Mean	Budapest	Seoul
g/kg								
Al	4.4–10.2	8.5	8.4–9.8	9.0	13.9–22.1	18.2		
Ca	22.4–87.3	67.6	26.7–86.7	50.2	31.2–58.6	43.5		
Fe	15.3–87.5	24.8	12.3–16.8	14.7	25.7–37.9	33.6		
K	0.6–3.9	2.0	1.5–2.7	1.8	4.4–7.9	6.5		
Mg	3.9–17.7	12.7	7.6–17.4	14.7	5.9–9.3	7.8		
Na	0.3–3.1	2.0	0.3–2.6	0.7	1.6–7.6	3.5		
P	0.6–1.5	0.9	0.6–0.9	0.8	0.8–1.5	1.1		
mg/kg								
As	8.2–15.8	11.6	6.7–11.1	9.1	15.2–31.2	24.9	7.3	6.8
Ba	137–961	304	53–448	162	217–716	570	95	
Bi	0.3–1.8	0.7	0.2–0.4	0.3	1.6–6.3	2.0		
Cd	0.4–1.9	0.8	0.3–0.7	0.5	1.4–6.7	3.5	0.5	0.3
Co	4.8–12.6	8.0	4.9–6.7	5.7	15.4–74.3	17.9	9	
Cr	37.4–120.6	65.5	21.0–33.7	27.5	104–195	130	21	25.4
Cu	144–352	236	122–274	164	302–478	351	19	15.3
Hg	0.1–0.2	0.1	0.1–0.2	0.1	0.3–0.6	0.5	0.1	
Li	5.5–21.7	13.9	12.0–15.8	13.1	25.0–43.0	29.5		
Mn	345–1,011	417	284–361	329	541–681	639		
Mo	1.3–8.8	5.0	0.2–1.7	0.9	6.7–18.6	13.7		
Ni	19.2–49.9	27.5	9.7–15.8	13.4	42–109	62	22	17.7
Pb	49–1,891	408	32–129	78	130–284	214	17	18.4
Sb	2.8–20.4	10.3	1.7–3.6	2.5	10.0–60.1	44.3		
Sn	6–31	20.9	4.1–9.1	6.6	21.1–50.4	39.7		
Sr	47–166	111	42–159	67	69–113	90		
Ti	0.1–0.2	0.1	0.1–0.12	0.1	0.3–0.7	0.4		
V	14.7–29.6	25.1	17.4–22.6	20.2	23.4–43.2	34.7		
Zn	317–2,110	891	202–840	403	1,075–2,065	1,476	65	54.3
µg/kg								
Pt	1.4–154	36.2	4.1–32.5	10.6	25.6–98.5	65.6		
Pd	8.3–187	50	8.6–36.7	17.9	30.6–148	101.7		
Ir	0.3–1.5	0.6	0.2–0.6	0.5	2.1–5.6	3.3		

(Hays et al. 2011). In dust grains sampled in Prague, the broad size distribution of Zn having also an ultrafine mode reflects various other sources than from tyres, like additives of engine oils (Ondraček et al. 2011).

Classification of results via indices

In order to get an overview about contamination levels and hazards from multi-element analysis, indices have been developed to get a rapid overview and to compare data worldwide.

Within Table 4, detailed results of the platinum metal concentrations together with main contaminant metals as well as the pollution index (PI) and the CI are shown.

The contamination index (CI)

Like the geo-accumulation index, the CI in Table 4 refers to the background values (Table 2), which are slightly different between Seoul and Hungary. It summarizes the multi-element approach to one number, thus smoothing individual outliers of a few contaminants only. Based on data from Yoon et al. (2009), it was calculated for Seoul as:

$$\begin{aligned}\mathrm{CI} = \{&(\mathrm{As}/6,83) + (\mathrm{Cd}/0,29) + (\mathrm{Cr}/25,4)\\ &+ (\mathrm{Cu}/15,3) + (\mathrm{Ni}/17,7) + (\mathrm{Pb}/18,4)\\ &+(\mathrm{Zn}/54,3)\}/7\end{aligned}$$

for Hungary, it was calculated as:

$$\begin{aligned}\mathrm{CI} = \{&(\mathrm{As}/7,3) + (\mathrm{Ba}/95) + (\mathrm{Cd}/0,5) + (\mathrm{Co}/9)\\ &+(\mathrm{Cr}/21) + (\mathrm{Cu}/19) + (\mathrm{Hg}/0,1) + (\mathrm{Ni}/22)\\ &+(\mathrm{Pb}/17) + (\mathrm{Zn}/65)\}/10\end{aligned}$$

Gondi et al. (2004) determined the background values of soils from the Hungarian Planes. These background values are similar to those of the east Austria's soil type "above alluvial sediments" (Danneberg 1999). These background values were determined after aqua regia digestion.

The CI shows higher contamination levels met in Seoul throughout; the high CI at the Margit Hid might be due to Pb-painting.

The geo-accumulation index (GAI)

The geo-accumulation index (GAI) has been defined by Müller 1979, 1981) as the logarithm (based on 2) of the measured concentrations over the geochemical background times 1.5. This has to be calculated for every element analysed and shows the enrichment versus the original geogenic level met on site. The GAI classifies metal enrichment as it is usual for the microbial saprobial status (class 1–5); the concentration space of each class doubles with increasing class number, which approximates the effect of environmental hazard Müller (1979, 1981).

A GAI around zero means uncontaminated, around one means moderately contaminated, around three means heavily contaminated and larger than five means extremely contaminated (Table 3). For the calculation of the GAI, baseline values similar to the CI were used (Table 2). From this, Seoul road dust is strongly enriched in almost everything. Road dust sampled outside Budapest still contains significantly more Cu, Zn and Pb than the background.

Table 3 Mean and standard deviations of calculated geo-accumulation indexes (GAI)

	Budapest	Main roads around Budapest	Seoul traffic hotspots
As	0.09 ± 0.32	−0.23 ± 0.30	1.22 ± 0.33
Ba	1.30 ± 1.03	0.22 ± 1.22	
Cd	0.22 ± 0.69	−0.60 ± 0.60	3.05 ± 1.28
Cr	1.18 ± 0.77	−0.13 ± 0.41	2.17 ± 1.06
Cu	3.01 ± 0.55	2.74 ± 0.77	4.02 ± 0.25
Hg	−0.15 ± 0.50	−0.89 ± 1.10	
Ni	−0.03 ± 0.74	−1.21 ± 0.49	1.64 ± 1.32
Pb	3.88 ± 1.63	1.56 ± 1.16	2.90 ± 0.36
Zn	3.14 ± 0.90	2.33 ± 0.98	4.23 ± 0.29

Explanation: geo-accumulation <0,5 non-accumulated
0.5–1.5 moderately accumulated
1.5–2.5 moderately heavy accumulated
2.5–3.5 heavily accumulated
> 3.5 very heavy accumulated

The pollution index (PI)

The PI refers to the tolerable levels, which have been set by legal advices, based on health impacts. Because thresholds might differ, the PI has to be calculated for each country separately. For our case, it is:

$$\begin{aligned}\mathrm{PI} = \{&(\mathrm{As}/20) + (\mathrm{Cd}/3) + (\mathrm{Cr}/100) + (\mathrm{Cu}/100)\\ &+(\mathrm{Sb}/5) + (\mathrm{Pb}/100) + (\mathrm{Zn}/300) + (\mathrm{Hg})\\ &+(\mathrm{Tl}/2)\}/8\end{aligned}$$

the PI indicates even more differences between the sampling areas than the contamination index. In the suburbs of Budapest, health hazards seem to be significantly lower (Table 4).

Table 5 indicates possible correlations between platinum metal and "traditional" metal contaminations in urban dust samples. At the bridges and the centre of Budapest, these are quite strong, contrary to the situation in Seoul and more rural areas the Hungarian. In short, in Budapest more Pd/Pt means less other contaminant metals. Pollution and contamination indices correlate well for Hungary, but not so well for Seoul.

Comparison with data from other regions

Concentrations in deposited dust samples

Unknown deposition velocities and surface adhesion reactions make it almost impossible to transfer µg/m^3, into µg/m^2 and into µg/g dust properly. Therefore,

Table 4 Detailed results for platinum metals and other contaminants. Pollution index (PI) and contamination index (CI) were calculated for other contaminants than Pt, Pd and Ir of Budapest and Outside of the Budapest in 2010 (Kadar and Marton 2012), and Seoul in 2010 (Lee et al. 2012)

	Cd mg/kg	Cu	Pb	Zn	Sb	Sn	Bi	Hg	Pt µg/kg	Pd	Ir	PI	CI
Budapest centre													
Szel kalman Square	0.9	258	330	1,029	21.2	29.5	1.7	0.1	100.3	166.2	0.9	2.2	6.7
West R. St.	0.8	192	545	1,355	3.2	9.1	0.6	0.1	7	29.2	0.4	1.4	7.4
East R. St.	2	150	434	2,179	12.3	20.7	0.7	0.2	31	46.8	0.5	1.9	8.8
Great circle	0.8	253	164	654	12.3	28.4	1.2	0.2	107	170	0.6	1.3	4.8
Budapest bridges													
Margit	1.2	206	2,790	886	11	15.6	0.7	0.1	1.9	12.8	4.5	4.2	27.1
Szabadsag	0.9	357	779	958	15.7	25.9	0.9	0.2	90	91	0.5	2.9	10.6
Petofi	0.6	329	787	495	8.8	21.7	0.6	0.1	133	122	0.5	1.9	8.3
Erzsebet	0.7	311	292	460	9.6	19.9	1.5	0.1	128	138	0.9	1.3	5.1
Margit Is. (Recr. A.)	0.5	187	81	628	2.9	6.8	0.4	0.2	2.7	20.6	0.4	0.6	3.3
Outside budapest													
Monor	0.3	422	30	196	2.3	6.3	0.2	0.1	9.1	15.4	0.4	0.4	3.2
Torokbalint	0.5	201	62.5	277	3.7	7.5	0.4	0.3	37	39.4	0.6	0.5	2.7
Szentendre	0.7	179	259	719	2.7	5.9	0.3	0.2	8.3	11.3	0.8	0.8	4.9
Vecses	0.5	134	88	989	2.9	9.3	0.3	0.1	21.5	19.9	0.5	0.7	3.5
Dunaharaszti	0.4	189	80	403	1.4	4.9	0.2	0.1	3.6	23.1	0.2	0.4	2.7
Ferihegy airport	0.8	214	96.5	1,040	3.8	9.8	0.5	0.1	29.5	23.3	0.7	0.9	4.5
Seoul													
Department SNU	16.9	335	268	1,676	10	39.2	6.3	0.6	25.6	81.8	2.2	2.7	20
Daerim area	6.7	390	217	1,340	28.5	50.4	1.6	0.5	33.9	30.6	2.7	3.8	13.7
Art centre	1.5	302	284	1,075	42.4	24.6	1.8	0.3	54.1	52	3.2	3.9	10.2
Seodaemun cross	2.9	478	213	2,065	58.4	40.2	2.8	0.5	49.4	82.2	2.1	7.8	15.4
Gyodae cross	4.1	475	215	1,454	60.1	705	5.7	0.4	98.5	128	3.3	7.7	14
Gangnam cross	6.2	336	183	1,497	35.6	21.1	1.9	0.4	77	125	4	4	13.3
Samsung cross	1.5	331	178	1,434	48	43.2	2.1	0.5	79.3	121.2	3.4	4.6	10.6
Zamsil cross	1.4	365	130	1,880	46.2	30.6	1.7	0.3	98.4	148	5.6	5.9	21.8

according to the simple sampling procedure done within this pilot study, the presented data from Seoul and Budapest can be just compared with concentrations with respect to urban street dust mass found in solid dust samples (Table 1), which were available from Madrid and Oslo (de Miguel et al. 1997), Tokyo (Furuta et al. 2005), and Muscat in Oman (Yaghi and Abdul-Wahab 2004). Table 2 contains a more detailed compilation of data from Seoul and Budapest. Among these, dust sampled in Seoul reached top levels for Cd–Zn–As–Co–Cr–Cu–Mo–Ni–Sn. Just Pb was rather low in Seoul because unleaded gasoline was introduced in Korea as compulsory in 1,993. In former times, the main source of Pb was petrol, leaded at different levels, e.g. 0.4 g/l for Madrid and 0.0375 g/l for Oslo (de Miguel et al. 1997). Concentrations in Budapest dust samples were lower than from Seoul, except for Pb and Mg. Compared with Madrid as another continental site, Budapest was higher in Co–V–Zn. Dust from Oslo, which is not so large, contained more Mn–Na–Sr than dust from other towns, but less other metals.

Street dust from Seoul contained higher concentrations of As, Ca, Mg, Mo, Ni, Sb and Sn and also higher trends for Cd and Zn, whereas Al, Cu, Fe, Mn and Pb were in about the same range (Fujiwara et al. 2011). Compared with Oslo, street dust from Seoul was higher in Cd, Cu, Mo, Ni, Sb and Zn, and it was lower in Mn–Na–Sr and in the same range for Ba–Co–P–Pb–Ca. Compared with samples from Madrid, Seoul was higher for Co–Cr–Cu–Mn–Ni–V–Zn and lower for Pb.

Table 5 Correlation coefficients between pollution index (PI), contamination index (CI) and platinum metals

	Pt	Pd	Ir	PI
Budapest centre				
PI	−0.0115	−0.1313	0.3703	
CI	−0.1994	−0.3098	−0.4090	0.6249
Budapest bridges				
PI	−0.7620	−0.8050	0.7506	
CI	−0.7652	−0.7626	0.8343	0.9695
Outside budapest				
PI	0.1531	−0.2213	0.6491	
CI	−0.0333	−0.2863	0.5266	0.9240
Seoul				
PI	0.5416	0.3930	0.0909	
CI	−0.0017	0.3193	0.2874	−0.0930

Continental Budapest contained higher concentrations of As, Ca, Mg and higher trends for Pb, whereas it was lower in Fe and Al, and within the same range for Cd, Cu, Mn, Mo, Ni, Sb, Sn and Zn. Samples from Budapest were higher for Cu–Pb–Zn–Ca than from Oslo, lower for Co–Mn–Ni–Sr–Na than from Oslo and within the same range for Ca–Cd–Mo–P and Sb. Samples from Budapest were higher for Co–V–Zn than samples from Madrid, lower for Ni–Pb and within the same range for Cr–Cu–Mn (de Miguel et al. 1997). In Raleigh of North Carolina(Hays et al. 2011), atmospheric particulates sampled near a highway contained less Li–Mg–Ca–Mn–Fe–Co–Cu–Pb–Zn than the Budapest or the Seoul dust samples, but more Sb, V, Pd and Pt, whereas Cr, Ba and P were within the same range and Ni–Tl were as high as in Seoul.

In outdoor dust Muscat in Oman (Yaghi and Abdul-Wahab 2004), high Ni could be clearly assigned to the high abrasion of adjacent serpentinite rocks, whereas high Zn came presumably from rubber tyres and Cr from corrosion of automobile parts. Cu levels from outdoor and indoor dust samples were the same, and thus, there was no additional outdoor source.

Grain size effects

Data referring to atmospheric dust can be given either per volume of air, as deposition per area, or as concentration in the dust particles. Usually, condensation processes from the gas phase lead to the formation of small grain size particles, whereas abrasion from soil and buildings rather coarser particles, and thus, the concentration changes largely with grain size (e.g. see Furuta et al. 2005 as shown for Tokyo). Elements forming volatile chlorides or oxides (Sager 1999) get accumulated in the inhalable fraction <2 µm, which comprises As, Cd, Pb, Sb, Se, V and Zn, e.g. for Tokyo (Furuta et al. 2005). Similarly, in Prague city, sampling of dust particles near a freeway and subsequent microlocal analysis identified groups of elements with similar size distributions, which might indicate identical origin. Particles of about 2.5 µm containing mainly Fe–Cu–Mn–Zn were assigned to the abrasion of different vehicle parts. Si–Al–Ca and Ca–K particles of about 5 µm were interpreted as resuspension of road dust, long-range transport or the regional background (Ondraček et al. 2011).

Assignment to sources

In dust samples from Oslo as well as from Madrid, Ba–Cd–Mg–Pb–Sb–Ti–Zn were identified as “urban elements” because they emanate from traffic or from buildings. If cement would be a major source, the proportion Mg/Ca was assumed close to 0.14. The group Ga–La–Mn–Sr–Th–Y was assigned mainly to soil particles, and a third group Cs–Ni–Ca–Cu–Fe–Mo–Sr of mixed origin (de Miguel et al. 1997). Under conditions met in USA, Br–Sb–Zn were recommended as markers for motor vehicle emissions, As–Se–S for coal fired power plants, V–Ni–REE for oil fired power plants, Mn–Al-Sc for soil, K for wood burning, Fe–Mn for steel making and Na–Cl for the proximity to the seaside (Huang et al. 1994). Close to a highway near Raleigh in North Carolina, USA, resuspended surface soil was regarded as a likely source of Al–Fe–Ca–Mg–K, whereas Fe–Ba–Zn–Cu–Sb were linked to the brake lining emissions and to a lesser extent to tire wear (Hays et al. 2011).

A diesel particulate filter can significantly reduce the particulate mass emissions at >90 % by means of filtration and trap also parts of volatile fractions (Hu et al. 2009). Alternatively, diesel engines may be equipped with a V_2O_5–TiO_2 catalyst for NO_x reduction. Emissions of Ni and Cr significantly increased with higher exhaust temperatures (Hu et al. 2009). During cruise style, higher engine load resulted in high exhaust temperature and thus in increased emissions of components of the lubricants occurs (Hu et al. 2009) about fivefold compared with baseline.

Special emissions from traffic

When composition data of atmospheric particulates from different parts of the globe are compared, traffic emissions as one of the main sources may differ due to vehicle construction, fuel and lubricant composition, use of catalysts, as well as composition of tyres and brake pads. In addition, geogenic background, proximity to the sea side, facades of buildings and choice of heating devices may impose certain variations. Last but not least, population density of Seoul is about four times that of a European city. Just concentrations of P and Ba seem to be the same in urban dust worldwide.

In the USA, vehicles go 95 % gasoline powered (Hays et al. 2011), whereas in Europe all trucks and a significant amount of smaller vehicles go diesel. Replacement of leaded gasoline by catalyst-equipped vessels changed the compositions of the particles. Al, La, Ce and Pt–Pd are supposed to increase, whereas apart from Pb, also Mn–Fe–Br were supposed to decrease in concentrations. At cold starts in particular, more La–Ce–Cl were emitted within a special test series (Huang et al. 1994). In the USA, lubricating oils contained about 0.15 % Zn, 0.12 % P, 0.35 % S and 0.35 % Ca, whereas other concentrations were just in the mg/kg range (Hu et al. 2009).

Urban soil samples

The concentrations met in urban soils collected in 0–10 depth in Vienna as well as in many other cases have been reviewed by Ajmone-Marsan and Biasoli (2010). In these cases, dust gets diluted with original soils, as well as with garden moulds and the like, which have been added to maintain green plants growing. With respect to the levels found in rural areas, urban soils are generally enriched in anthropogenic elements like Pb, Cu, Zn and Cr, because they may get a lot of additional runoff from sealed areas besides increased atmospheric deposition, but urban road dust surely contains even more hazardous metals.

Conclusions

This study presents the first results of Pt, Pd, Ir in road dusts collected from inside Budapest, as well as from major roads around this city, together with metallic contaminants and main elements. According to the geo-accumulation indices calculated for the respective metals, Budapest urban dust was heavily polluted with Pb, Zn and Cu, as well as moderately polluted with Cr and Ba. Pollution levels along main roads around Budapest remained high for Cu and Zn, but were rather low for others.

As a contrast, street dusts collected at traffic hot spots in the 4 times more densely populated Seoul metropolitan area were heavily polluted with Zn, Cu, Cd, Pb and Cr, and to a lesser extent with As and Ni (Table 3).

Platinum and Pd pollution were rather independent from other contaminant metals, except for samples from the bridges situated in Central Budapest, which can be shown by the correlation coefficients of the respective contamination indices with Pt and Pd concentrations encountered (Table 4, 5).

The obtained results indicate that highest Pt, Pd and Ir levels in road dusts were found from major roads with high traffic volume. Significant difference between heavy traffic roads in PI and CI from Budapest (Szél Kálmán Square, West Railway Station, East Railway Station, Great Circle, Danube Bridges) and Seoul (Crosses: Seodaemun, Gyodae, Gangnam, Samsung, Zamsil) (2.14 and 9.1; 5.98 and 15.00), and light traffic roads (0.61 and 3.31; 3.5 and 14.6) reflects that an important source of Pt, Pd and Ir in roadside environment is automobile catalytic converter. Road dusts with high Pt, Pd and Ir level were enriched in traffic-related contaminant elements also. Among Madrid, Oslo, Tokyo and Muscat (in Oman) data, dust samples in Seoul reached top levels for Cd–Zn–As–Co–Cr–Cu–Mo–Ni–Sn. Just Pb was rather low in Seoul because unleaded gasoline was introduced as compulsory in 1993. Concentrations in Budapest dust samples were lower than from Seoul, except for Pb and Mg. Compared with Madrid as another continental site, Budapest was higher in Co–V–Zn. Dust from Oslo, which is not so large, contained more Mn–Na–Sr than dust from other towns, but less other metals.

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