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Original Research

Evaluation of Traffic-Related Heavy Metals Emissions Using Noise Barrier Road Dust Analysis

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Abstract

The impact of traffic emissions on the environment close to an arterial road often is assessed on the basis of heavy metal pollution of road dust collected directly from or close to the road surface. In our work we propose an assessment of a potential environmental hazard on the basis of an analysis of the content of heavy metals in the road dust trapped on vertical acoustic barriers installed directly by roads.

This kind of road dust appeared to be a good indicator of traffic related to heavy metals emissions. The pollution level of Cu and Zn reached the category of "extreme level." An increased level of Cu was found on a decelerating activity section. A good correlation between Cu, Mn, and Fe may imply that brake wear is also an important source of iron and manganese. It has been shown that high concentration of Zn is a result of its release from zinc-plated road furniture.

Keywords: traffic emission, road dust, heavy metals, noise barrier

Introduction

Motor transport is one of the most important sources of emissions of particulate heavy metals into the air. In densely populated countries of Western Europe transport PM emissions predominate in total atmospheric emissions, whereas coal combustion sources predominate in Eastern Europe. In Poland motor transport is responsible for emissions of 73.2 Gg TSP, which amounts to 18.6% of total TSP emissions [1]. In cities with limited ventilation, particularly in the most densely populated parts of the cities, the concentrations of toxic metals in the air are, to a large degree, affected by motor traffic. Similarly, in the close vicinity of arterial highways with intercity traffic, the air, soil, and surface waters are heavily contaminated with heavy metals. Budai and Clement estimated that highway wear emission is responsible for 57% of copper and 65% of zinc vehicle traffic emissions [2].

Road traffic involves numerous potential sources of metals, e.g. combustion products from fuel and oil; wear products from tyres, brake linings, bearings and clutches; corrosion products of vehicle components and road construction materials; and resuspension of soil and road dust. The significant reduction in the permissible lead concentration in petrol has led to a considerable decrease in exhaust lead emissions within the last two decades. However, the number of vehicles in operation increasing year by year and lengthening of trips have resulted in the emissions of larger amounts of metals (including Pb) originating from brake, tire, and road wear [3]. Landa et al. estimated that ca. 285,000 Mg of Zn was released from tire wear in the U.S. between 1936 and 1999 and that about 10,000 Mg of Zn was released in 1999, alone [4]. Copper emissions only from brake wear in Europe in 2000 is estimated at almost 2,400 Mg [5].

As environmental pollution by heavy metals creates hazards to human health and the environment, their concentrations in individual elements of the environment are monitored and emissions are inventoried.

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Traffic emission impact on the environment near a highway is often assessed on the basis of heavy metal pollution of roadside soil or runoff water [6, 7]. Road dust also has been used as an indicator of heavy metal pollution of the environment [8-10]. Road dust samples are collected directly from a highway surface [8, 10] or the surrounding area [9]. Another approach to estimation of heavy metal deposition resulting from road traffic consists in the application of biomonitoring methods [11].

In our work we propose an assessment of a potential environmental hazard on the basis of an analysis of the content of heavy metals in the road dust collected from vertical noise barriers installed directly by highways. Sample composition maps emissions of particulate metals in accordance with traffic and driving conditions averaged for a sufficiently long time.

Experimental

Object Description

The object of the studies was selected specifically to represent motor traffic of significant traffic density on a transit road with modern infrastructure. The E-77 dual carriageway is one of twelve international roads (Fig. 1). The road consists of two carriageways with two lanes each and two shoulders with an asphalt surface. Double guardrails separate the carriageways and single guardrails run along the right edge of both carriageways. The outside 0.3 m wide guardrails are fixed at a height of 0.5 m. Approximately 6 m high vertical absorptive noise barriers are installed in residential areas.

Annual average traffic density on the investigated stretch of the E-77 dual carriageway was 24,089±1,401 veh/h (max. 61,240 veh/h at the Raszyn-Janki section). The average traffic density on the whole road is 18,469 veh/h, which is close to the average traffic density on all international roads in Poland: 20,006 veh/h, and on dual carriageways 19,567 veh/h.

Fig. 1. Location of E-77 road and Jedlińsk-Falęcice section of sampling sites.

The mean contributions of passenger cars, light duty vehicles, heavy duty vehicles, buses and motorcycles in the vehicle fleet were 68.1%±1.2%, 10.7%±0.7%, 20.3%±1.1%, 0.74%±0.04%, and 0.17%±0.03%, respectively. The speed of the vehicles was 90-130 km/h. The traffic data were taken from the GDDKiA Report [12].

The research area was located in a typically agricultural region away from industrial and heat engineering sources of trace metals. The sampling points were located at three sites along the Jedlińsk-Falęcice section (Fig. 1). RD was collected for 2 years at the first two sites and for only 1 year at the latter site:

- Jedlińsk (road dust samples are denoted as RD-J): noise barriers (installed in 2009) are located on the eastern side 2.2 m from the carriageway, the width of the shoulder is 0.8 m and the sampling point was right before the traffic lights.
- Kamień (road dust samples are denoted as RD-K): noise barriers (installed in 2009) are located on the eastern side 2.5 m from the carriageway, the width of the shoulder is 1.2 m, steady-flow traffic.
- Falęcice (road dust samples are denoted as RD-F): noise barriers (installed in 2010) are located on both sides of the road. The eastern barrier is placed 4.4 m from the carriageway, the width of the shoulder is 2.8 m, steady-flow traffic.

Table 1 shows the characteristics of PM and heavy metal emissions on the investigated section of the road. The emission intensities were calculated as a sum of products of emission factors and traffic density for each vehicle type according to the methodology used for inventory of non-exhaust traffic emissions in Germany [13] and Denmark [14]. The exhaust emissions were obtained from the COPERT III model [15].

Sampling

The RD samples (ca. 20 g) were collected by means of a vacuum cleaner from the acoustic barriers on two surfaces: the 0.0-0.6 m×20 m bottom section, and the 1.8-2.4 m×20 m upper section (road dust samples are denoted as RD-(b) and RD-(u), respectively).

The soil samples used for the analyses allowing for the determination of contamination indexes were collected from the upper soil layer at two spots between road dust collection sites on the western side of the E-77 road.

All the samples were allowed to dry in air under laboratory conditions and then sieved through a nylon 1 mm screen and kept in tightly closed plastic containers before analysis.

Chemical Analysis

Sample Preparation

Microwave-assisted digestion of RD and soil samples was applied prior to AAS measurements. The weighed amounts of 0.5 g were placed in 100 mL Teflon vessels with 5 mL 65% HNO_3 and 1 mL 30% H_2O_2. The samples were

Table 1. Intensity of metal emission from E-77 road [kg·km^{-1}·year^{-1}].

Pollutant	Tire wear		Brake wear		Road abrasion		Fuel combustion	Total	
	GEFs	DEFs	GEFs	DEFs	GEFs	DEFs	COPERT III	GEFs	DEFs
PM	163.3	180	116.4	78.7	244.7	244.7	165.2	690	669
Cr	0.003	0.0006	0.021	0.011	0.017	0.006	0.034	0.08	0.05
Cu	0.005	0.003	41.32	3.02	0.001	0.003	1.157	42	4.2
Ni	0.003	0.005	0.068	0.009	0.009	0.005	0.048	0.13	0.07
Pb	0.026	0.014	0.975	0.366	0.001	0.014	1.507	2.5	1.9
Zn	29.36	2.11	5.21	0.91	0.02	0.02	0.68	35	3.7

GEFs – German emisson factors [13]
DEFs – Danish emission factors [14]

digested in a microwave oven (Milestone MLS 1200 Mega) according to the program given by the manufacturer's protocol [16]:

1) 6 min., 250 W
2) 1 min., 0 W
3) 6 min., 400 W
4) 6 min., 650 W
5) 6 min., 250 W
6) 5 min., ventilation

After cooling, the digests were transferred to polypropylene standard flasks and diluted to 50.0 mL with water.

Three subsamples were digested simultaneously for each sample.

Metal Determination

The metals (Al, Co, Cr, Cu, Fe, Mn, Ni, Pb, and Zn) were determined using an atomic absorption spectrophotometer AAS – Agilent Technologies 200 Series AA 240 FS AA with flame atomization. Standard solutions prepared by appropriate dilution of the stock solution 1000 µg/mL (J.T.Baker) were used to calibrate the device by means of the standard curve method. N_2O was used as an oxidizer only to determine Al, whereas the acetylene/air flame was used to determine the other metals.

Determination limits for the metals being examined were: Al – 6.1 mg/kg, Co – 2.6 mg/kg, Cr – 2.7 mg/kg, Cu – 1.2 mg/kg, Fe – 3.8 mg/kg, Mn – 1.6 mg/kg, Ni – 2.8 mg/kg, Pb – 3.0 mg/kg, and Zn – 1.0 mg/kg.

Results and Discussion

The E-77 dual carriageway is an efficient source of emissions of a number of metals, including toxic heavy metals (Table 1). It can be expected that the environment is, to the largest degree, loaded with copper and zinc, even though the estimated emission intensities of the two metals differ almost 7-fold (according to the German and Danish methodologies). The predicted emission intensities of Cr, Ni, and Pb are very similar, although the amounts of the released Ni and Cr are several times lower than those of Pb. The load of metals estimated in such a way only partially burdens the environment bordering with the traffic route, e.g. the initial dispersion of the emitted pollutants is effectively blocked by the absorbing noise barriers. PM concentration may decrease up to 50% behind the noise barrier [17]. Sanders et al. found that 3-30% of the debris falls on the road while 16-22% remains on the wheels and 8-25% is retained on the steering/suspension equipment [18].

As has already been mentioned, the RD trapped on acoustic barriers has certain characteristics that can be used to evaluate traffic-related heavy metal emissions. The RD samples represent an averaged pattern of exhaust emissions, brake wear, tire wear, road abrasion, and corrosion of road structure and automotive parts; also large-size particles that deposit close to the road or are disintegrated by vehicles and which undergo resuspension by wind and wake induced by traffic stream. The composition of the samples also reflects the density of the road traffic, its structure and driving conditions averaged for a relatively long time. It is also worth noting that the spectrum of emitted particles that are trapped on the active surface of the barriers should be fuller than the spectrum of particles that settle on flat surfaces near the road (traffic sediment samples are often used as indicators of environmental hazard from traffic pollutants).

Concentrations and Major Sources of Heavy Metals

The values of metal concentrations in the RD samples varied considerably from several mg/kg for Co to over 20,000 mg/kg for Al and Fe (Table 2). The mean concentrations of heavy metals were in the following order: Fe >Al >Zn >Mn >Cu ≥Cr >Pb >Ni >Co. The content of Zn exceeds even the permissible limit for soils in industrial areas and the permissible limit of Cu and Cr for agricultural soils [20].

These findings are consistent with the results of other road dust studies (Table 3). The fact that our values are higher than those published by other authors is an expected result in view of the kind of RD samples we have collected.

Table 2. Concentration of metals in the samples of road dust and soil.

Sample	Metal concentration [mg·kg-1]								
	Al	Co	Cr	Cu	Fe	Mn	Ni	Pb	Zn
RD-J(b)	11,968	4.19	191.38	233.25	19,906	492.42	22.13	42.26	3,257
	11,561	4.79	173.72	233.63	20,122	513.18	21.36	42.93	3,241
	11,403	4.18	179.35	227.18	20,418	510.16	21.72	42.43	3,220
x̄±SD	11,640±290	4.39±0.35	181.5±9.0	231.4±3.6	20,150±260	505±11	21.74±0.39	42.54±0.35	3,239±19
RD-J(u)	16,180	6.79	209.66	275.45	23,567	656.67	24.95	45.31	3,074
	16,511	5.59	211.66	265.57	23,468	640.97	26.76	44.53	2,999
	16,112	4.38	219.30	277.11	23,164	647.90	27.91	48.44	2,897
x̄±SD	16,270±210	5.6±1.21	213.5±5.1	272.7±6.2	23,400±210	648.5±7.9	26.5±1.5	46.1±2.1	2,990±89
RD-K(b)	10,410	6.29	56.91	101.84	14,763	442.29	24.96	56.11	2,880
	9,884	6.39	53.93	102.88	14,386	436.48	25.07	43.55	2,767
	9,785	6.39	58.96	102.94	14,231	448.73	26.38	44.67	2,763
x̄±SD	10,030±340	6.36±0.06	56.6±2.5	102.6±0.6	14,460±270	442.5±6.1	25.47±0.79	48.1±7.0	2,803±66
RD-K(u)	19,960	9.09	97.94	134.92	21,024	552.67	32.98	59.16	3,132
	20,488	8.99	99.94	139.92	20,054	575.65	31.78	55.77	3,143
	19,784	8.39	10.94	138.92	19,886	548.67	32.88	57.06	3,067
x̄±SD	20,080±370	8.82±0.38	99.6±1.5	137.9±2.6	20,320±610	559±15	32.55±0.67	57.3±1.7	3,114±41
RD-F(b)	13,561	15.94	139.49	81.70	16,885	394.58	28.49	35.47	3,420
	12,987	9.96	147.46	89.68	17,142	390.59	28.09	31.29	3,270
	13,383	7.97	159.49	77.75	17,512	384.77	28.31	33.69	3,138
x̄±SD	13,310±290	11.3±4.2	149±10	83.0±6.1	17,180±320	390.0±4.9	28.30±0.20	33.5±2.1	3,276±141
RD-F(u)	15,032	7.96	185.04	79.59	17,945	393.95	28.85	34.22	3,080
	15,362	9.94	188.94	83.53	18,463	413.68	28.64	34.61	3,194
	14,699	7.78	183.55	91.78	17,640	415.00	31.12	30.92	3092
x̄±SD	15,030±330	8.6±1.2	185.8±2.8	85.0±6.2	18,020±420	408±12	29.5±1.4	33.2±2.0	3122±63
RD (mean value ±SD)	14,400±3,400	7.5±2.8	148± 56	152±76	18,920±2,900	492±94	27.4±3.6	43.5±9.1	3092±176
Soil (x̄±SD)	4,220±250	2.87±0.15	9.04±0.41	4.17±0.34	4,440±150	107.7±4.9	5.86±0.38	7.13±0.19	20.94±0.82
Soil (background value) [19]	-	4.0	27.0	7.1	12,900	289	10.2	9.8	30.0

Metal concentrations in road dust particles are proportional to the emission intensity of these metals on a given section of the transit road. Thus, it can be expected that the areas adjacent to the E-77 expressway are, to the highest degree, exposed to pollution by Zn (3,092±176 mg/kg) and, to a much lesser degree, by Cu (152±76 mg/kg) and Cr (148±54 mg/kg). Pb pollution risk (regarded as a "hot issue" about 20 years ago) is small and comparable to Ni, 49±9.1 mg/kg and 27.4±3.6 mg/kg, respectively.

The concentration of Cu showed the highest variability (RSD = 50%). A considerable variability was shown by the concentrations of Cr and Co (RSD = 38% and 37%, respectively). In this case the concentration variability in individual samples reflects the effect of the parameters characterizing traffic flow on the emission of metals. As traffic density and traffic structure at all sampling sites were similar, the sources of the variable metal concentration in road dust can probably be found in traffic conditions on individual sections of the road. The Cu concentrations have confirmed that brake wear emission is the main source of Cu. The Jedlińsk sampling site was situated directly in front of the traffic lights, so the significant level of Cu identified in the road dust can be attributed to a high rate of brake abrasion from the increased stopping and the low speed of vehicles at this site.

Table 3. Concentrations of heavy metals in the road dust.

Site	Metal concentrations [mg/kg]								Reference
	Al	Cr	Cu	Fe	Mn	Ni	Pb	Zn	
E-77 Asphalt dual carriageway, Poland	14,400	148	152	18,900	492	27	44	3,092	This work
Asphalt highway, Korea			172			26	117	323	[8]
Highway, Turkey			208			33	212	521	[21]
Islamabad Highway, Pakistan,			52			23	104	116	[9]
Heavily trafficked tunnel, Sweden	7,810	36	264	13,500	170		74	496	[22]
Tema Highway, Ghana		152	44	35,900	356	6.5	117	213	[23]

Al concentration in the RD, considerably increased in comparison with soil samples (14,400 mg/kg vs. 4,200 mg/kg) which are also characterized by high variability, suggests a considerable share of anthropogenic-Al, e.g. from abrasion of aluminum rims of vehicle wheels. An analogous view can refer to emission sources of the other matrix metals: Fe and Mn.

Considering all the metals investigated, the determination results of Zn have a specific characteristic, high concentrations and very low variability (RSD = 5.7%; Table 2).

Zinc accounts for about 1% by weight of tire tread material and its release through tire wear has been often recognized as a significant source of Zn in road dust. Brake wear and fuel combustion also are regarded as important vehicular emission sources of Zn (Table 1). Our results seem to suggest that the problem is more complex. Zinc is the only metal that is present in all the samples at a constant level $[Zn]_{av.}$ = 3,092 mg/kg±176 mg/kg. The differences between zinc concentrations in the samples collected at different sites are not statistically significant: RD-J (3115 mg/kg±148 mg/kg), RD-K (2959 mg/kg±177 mg/kg) and RD-F (3199 mg/kg±129 mg/kg). An increased zinc content in the samples enriched with Cu due to intensive vehicle braking (RD-J) was not observed. Taking into account a significant variability of concentrations of the other metals, the reasons for the high and steady zinc level in the traffic dust can probably be found in non-vehicle components of road traffic emissions. In the case of the road considered, this is probably zinc-plated road furniture. Hence, the corrosion of zinc protective coating (periodically intensified by the application of de-icing agents) and road dust blasting of zinc coating (especially during winter road sanding) can be regarded as the most important sources of the traffic dust enrichment by zinc. Previously, Hjortenkrans et al. suggested that galvanized road furniture is probably the most dominant Zn source, which may obscure other correlations [24].

The lower concentrations of particulate metals in bottom samples (particularly Al, Cr, Cu, Mn, Fe, Ni, and Pb) than in upper samples probably result from higher concentrations of the metals in particles displaying a weaker tendency for sedimentation. Another possible explanation is probably a higher share of the mineral fraction thrusted from under the wheels in RD-(b) samples and characterized by the natural content of heavy metals. The latter factor is not predominant because the very high zinc content in RD samples caused to a high degree by road dust blasting of zinc coating does not practically depend on the RD-sampling level: bottom samples $[Zn]_{av.}$ = 3,107±240 mg/kg, and upper samples $[Zn]_{av.}$ = 3,075±87 mg/kg.

Assessment of Contamination

The degree of RD contamination by heavy metals is usually quantified by such parameters as geo-accumulation index (I_{geo}), pollution index (PI), integrated pollution index (IPI), and enrichment factor (EF) [8-10]. In this work EF was not used due to a lack of an appropriate reference element. Al and Fe used by other authors for normalization of heavy metal concentrations [23, 25] have a high occurrence variability in our RD samples due to their anthropogenic origin. I_{geo} was calculated according to equation:

$$I_{geo} = \log_2\left[\frac{C_n}{1.5B_n}\right]$$

...where: n-metal concentration in the RD was used as C_n, and n-metal concentration in unpolluted soil was used as B_n. Zinc pollution reached the extremely polluted ($I_{geo} > 5$) level, Cu – strongly polluted ($I_{geo} > 3$), Cr – moderately to strongly polluted ($2 < I_{geo} \leq 3$) (Fig. 2). The concentrations

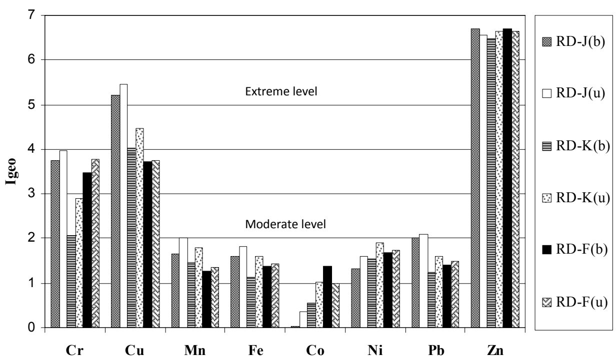

Fig. 2. Geo-accumulation index of heavy metals in the road dust of road E-77.

Table 4. Correlation matrix between metal concentrations in the road dust of E 77 road.

	Co	Cr	Cu	Fe	Mn	Ni	Pb	Zn
Co	1.00							
Cr	-0.20	1.00						
Cu	-0.63	0.55	1.00					
Fe	-0.28	0.70	0.81	1.00				
Mn	-0.43	0.29	0.85	0.83	1.00			
Ni	0.57	-0.21	-0.48	0.02	-0.03	1.00		
Pb	-0.23	-0.47	0.30	0.25	0.62	0.18	1.00	
Zn	0.47	0.43	-0.02	0.24	-0.20	0.01	-0.36	1.00

of the other metals studied did not exceed the moderately polluted level. The integrated pollution index, which is the mean value of the pollution index $(PI = \frac{C_n}{B_n})$ of n-metal in the RD samples studied, took a very wide range of values from IPI = 2.61 for Co (high level of pollution) to IPI = 148 for Zn (extreme level of pollution) (Fig. 3). The IPI value remains at a level of nearly 4 in the case of half of the metals considered (Mn, Fe, Ni, and Pb). The presence of Pb in this group of metals results undoubtedly from a significant reduction of Pb in fuels (max 5 mg/L, earlier 0.5-1 g/L).

On the basis of I_{geo} and IPI it can be said that the open environment along the E-77 road is heavily exposed to pollution by heavy metals, particularly Zn, Cu, and Cr.

Correlation Analysis

The Pearson's correlation coefficients for the metals studied are collected in Table 4. Significant positive correlations involve Mn, Fe, and Cu. Taking into consideration the unquestionable view that brake wear is the main source of Cu in road dust, a good correlation Cu with Mn and Fe indicates that brake wear is also an important source of iron and manganese [26]. These metals may be emitted during wear of the brake discs/drums, which are commonly made of steel or alloys. The potential sources of Fe also may be iron powder and steel wool used in semi-metalic and non-asbestos organic brake linings [27]. Therefore, the role of soil dust as a carrier of these metals in the RD samples is not predominant and this also confirms the assumption that the RD samples collected from the acoustic barriers are a good reflection of the properties of the particulate matter emitted during motor traffic.

Poor correlation coefficient values for the other metals indicate that these metals originate from a number of independent traffic emission sources.

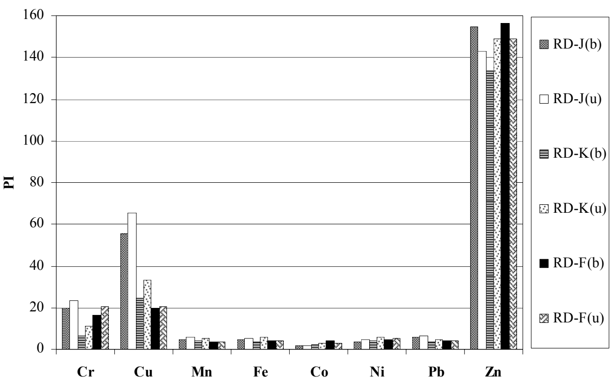

Fig. 3. Pollution index of heavy metals in the road dust of E-77 road.

Conclusions

The RD samples collected from acoustic barriers turned out to be a good indicator of traffic-related heavy metals emissions. An increased Cu level was found on a road section with a decelerating activity. A very high Zn concentration remaining at a steady level on the whole section of the road resulted from its release from zinc-coated road furniture and the important role of this emission pathway in the formation of a metals distribution pattern in the PM emitted. The pollution level of Cu and Zn reached the category of "extreme level." A good correlation between Cu and Mn, as well as between Cu and Fe, may imply that brake wear is also an important source of iron and manganese. The concentration values of the metals (Al, Cr, Fe, Mn, and Zn) in the RD trapped on acoustic barriers that we have obtained are higher than those published earlier. Without further research it will be difficult, however, to assess to what extent this results from a different characteristic of road traffic and to what extent it is a result of the fraction range of the traffic dust being generated, which is present in the RD samples analyzed in other laboratories.

Apart from the assessment of heavy metals emissions to the environment, the results of the determination of heavy metals in the RD trapped on acoustic barriers can be useful also for calibration of the methods used for the inventory of road transport heavy metals emissions.

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