JEE Journal of Ecological Engineering

Journal of Ecological Engineering 2023, 24(11), 82–90
https://doi.org/10.12911/22998993/171377
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Received: 2023.08.06
Accepted: 2023.08.22
Published: 2023.09.15

Elemental Composition of the Ultrafine Fraction of Road Dust in the Vicinity of Motorways and Expressways in Poland – Asphalt Versus Concrete Surfaces

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ABSTRACT

Air pollution in the vicinity of roads is a complex and growing problem. In urbanised areas, there are many sources of dust emissions, but one of the main ones is road traffic. Investigating and assessing the physical and chemical properties of road dust and, more specifically, dust collected from surface courses is one way of providing an opportunity not only to identify the contribution of the emitters concerned to the formation of dust air pollution in the vicinity of roads but above all the environmental risks associated with traffic emissions. The study aimed to analyse the elemental composition of dust with a fraction <0.1 mm, collected from asphalt and concrete roads characterised by the highest technical and service parameters in Poland. The samples were analysed using a Shimadzu EDX 7000 energy-dispersive X-ray fluorescence spectrometer, then the results were statistically analysed using the t-Welch test, and the enrichment factors EF were determined. It was shown that road dust with a grain size of less than 0.1 mm collected from asphalt surface course was extremely highly enriched in Cu, Cr, Pb and S, while that from the concrete surface course was enriched in Zn and Zr, indicating a strong anthropogenic origin of these elements; exhaust gases were identified as their source. Irrespective of the type of surface course, very high dust enrichment occurs for Ca, Mn, Ni, S, Ti and Y. These elements may originate from the abrasion process of vehicle tyres. For road dust collected from both road types, the most similar EF values were found for Fe, K, Mn, Si, Sr and Ti. The source of these elements is most likely the roadside soil. It follows that the type of road surface is not the main determinant of the composition of road dust with a fraction <0.1 mm.

Keywords: particulate matter, road dust, abrasion of the road surface, exhaust emissions

INTRODUCTION

There are many sources of dust emissions in the vicinity of roads. This is reflected in the wide range of types and concentrations of elements and their compounds in the material taken from the paving. The complex nature of road dust makes quantitative estimation of the contribution of specific elements to overall particulate emissions problematic and may not be resolved by chemical methods alone. The identification of non-exhaust-related road dust emitters is further complicated by their interaction.

Particulate matter (PM) particles from many different sources can settle on the road surface, making it very difficult to determine emissions from road wear and tear (Gunawardana et al., 2012). The particles generated during paving erosion can be in a wide size range and their composition is determined by the material that makes up the road surface layer (Penkała et al., 2023). Depending on the road design, the construction materials used and the gradient of the road lane, particulate pollutants can be transported, e.g., with the splash of water droplets, over distances of up to 10 m. In addition, very fine particles (PM) can move with the air in the direction of the wind over considerable distances, affecting areas even up to 250 m away (Czajka, 2017). Potentially toxic pollutants account for ca. 30% of the dust mass and are related to brake and tyre wear, exhaust emissions and the release of fly ash from asphalt. Heavy metals found in road dust, such as Zn, Cu, Pb, Ni, Cr and Cd, come mainly from traffic, while roadside soil is a significant source of Fe, Al and Mn (Al-Sareji et al., 2021; Gunawardana et al., 2012).

Although scientists have been fairly successful in defining groups of individual emission sources of selected elements, pinpointing the emitters of each is a much more complicated task. The large number of materials in use, or new materials (often of unspecified chemical composition), allow the identification of only the so-called major emission sources (Penkała et al., 2018; Świetlik & Trojanowska, 2014). For example, tread wear on car tyres is a clear emitter of Zn in road dust (Thorpe & Harrison, 2008; Zechmeister et al., 2005), however, on expressways, galvanised road infrastructure components can also be a major source of Zn (Hjortenkrans et al., 2006; Świetlik et al., 2013). Brake pads and discs are responsible for the emission of Cd, Cr, Cu, Fe, Ni, Pb and Zn (Zechmeister et al., 2005), but mainly for the high Cu content of road dust (Thorpe & Harrison, 2008). It is assumed that oils and lubricants are a source of Cd, Cu and Ni emissions, while exhaust gases enrich the environment with elements such as Cd, Cr, Cu, Ni, Pb and Zn (Zechmeister et al., 2005). Surface course abrasion is a source of road dust, the chemical composition of which is determined by the structure of the concrete or asphalt mix, aggregate and fly ash (if it was used in the production of the asphalt mix) (Penkała et al., 2019; Świetlik & Trojanowska, 2014).

The composition of roadside soil may also have a lot to do with material that becomes accumulated in the vicinity of traffic routes. It has been shown that, under certain conditions, road dust consists mostly of particles from the soil (60%), mainly quartz (40–50%). Approximately 2% is organic matter composed largely of plants (Gunawardana et al., 2012). Elements are also present in road dust which, when present in large quantities, are or may be toxic to living organisms, and the strength and type of effect depends mainly on the concentration and chemical form in which they are bound. The problem of the presence in road dust of such substances as well as other pollutants is increasingly being addressed by researchers worldwide (Kiebała et al., 2015). For example, the study of Konstantinova et al. (Konstantinova et al., 2022) investigated the levels and sources of road dust pollution collected in the city of Tyumen, a major transport centre with one of the highest motorisation rates in Russia. Analysing the proportion of elements in the samples, it was found that the concentrations of Cr, Ni and Co were higher as compared to the results from other major localities. Concentrations of Ni, Cr, Sb and Mo in road dust were higher than in roadside soils within the city, indicating that transport is the main source of these elements.

Only field and laboratory tests can provide specific information on dust emissions originating from road surface abrasion. However, it should be borne in mind that the results obtained can also be influenced by field/environmental factors and the proportion of vehicles travelling on a given road. There is a large body of work related to road dust in global literature, but most of it focuses on emissions from tyre abrasion, brake friction components and exhaust gases, while emissions from road surface abrasion still remain largely unexplored (Kiebała et al., 2015; Penkała et al., 2018, 2019, 2023; Thorpe & Harrison, 2008). In Poland, research of this type has been conducted to a very limited extent. For example, the work of Rybak and co-workers (Rybak et al., 2020) presents the distribution and possible sources of selected elements (Mn, Ni, Cu, Zn, As, Rb, Ba, Cr, Mg and Al) in road dust in two urban agglomerations: Wrocław (Lower Silesia) and Katowice (Upper Silesia) in Poland. Zn, As and Cu were proven to be the main elements present in the samples in both regions, probably from fuel combustion, traffic, home heating and industrial emissions. In addition, a potential source of Cu was the corrosion of metal alloys in vehicle covers and components or steel surfaces of road infrastructure.

Taking into account the above issues, the authors of the publication considered it extremely important to study the physical and chemical properties of the finest fraction of road dust (it can be identified with PM when the studied fraction consists of particles <100 μm), depending on the type of surface from which it was collected in the vicinity of motorways and expressways in Poland.

Although the scope of the work and the methodology applied are not new in global literature, it should be clearly emphasised that in the case of Poland, such research has not been conducted to date. However, in Poland, this problem is considered to be particularly significant due to the high values of concentrations of PM and some of its components in the atmosphere, which are still being considered to be even dangerously high. In this context, identifying the influence of road dust matter on PM concentrations and elemental composition appears to be extremely important. In addition, establishing the relationship between the quality of the road construction material and the elemental composition of the finest fraction of road dust (here equated with PM) may provide some new information relevant to research into new construction materials, additives or even technologies aimed at reducing road dust emissions.

METHODOLOGY

Dust samples were collected from sections of roads with the highest technical and performance requirements, i.e., motorways and express roads situated in the central and southern parts of Poland. Routes with two types of surfacing (asphalt and concrete) were selected. The list of roads is provided in Table 1 (GDDKiA, 2015). The distribution of measurement points on the map is shown in Figure 1.

The samples were collected at eight measurement points at similar heights on the left and right sides of the roadway (to obtain a representative sample that was not affected by wind or the direction of vehicle traffic). The dust for testing was collected each time by hand (using a broom) from an area of not less than 2 m^2 into sterile plastic containers. In total, samples from 16 points were analysed. Road dust was sampled in the summer months due to the reduction of the impact of de-icing agents in the material collected from the surface course and also due to the occurrence in Poland, in the ground, of the phenomenon of masking other sources of emissions by the then dominant municipal emissions (Majewski et al., 2018; Pastuszka et al., 2010; Rogula-Kozłowska et al., 2015)

Road dust samples were subjected to granulometric analysis with a Sieve Shaker mechanical sieve shaker. Using a standard type of sieve, seven fractions of material were obtained: 10–2 mm, 2–1 mm, 1–0.5 mm, 0.5–0.25 mm, 0.25–0.1 mm, 0.1–0.063 mm and <0.063 mm. Subsequently, all sieved samples with grain sizes <0.1 mm (total fraction 0.1–0.063 and <0.063 mm) were examined for

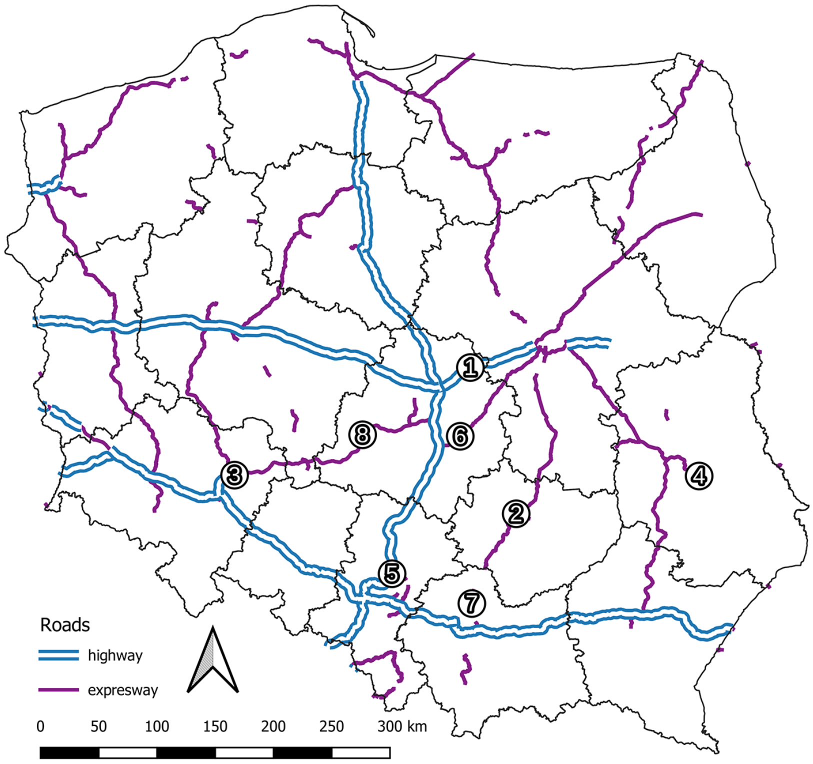

Figure 1. Map of Poland showing roads selected for the survey, road network based on (OpenStreetMap Contributors, 2023)

Table 1. Location of the road sections selected for the study

Measurement point	Road No.	Selected sections	Description of section	Chainage [km]		Surfacing type
				Beginning	End	
1	A2	Warszawa – Łódź	Interchange Łowicz – Interchange Skierniewice	385.952	398.096	asphalt
2	S7	Kielce by-pass	Interchange Kielce Zachodnie – Interchange Kielce Jaworznia	6.671	15.107	asphalt
3	S8	Wrocław – Sieradz	Interchange Wrocław Psie Pole/DK A8 and 98/ – Interchange Oleśnica Zachód /DW340/	29.219	51.664	asphalt
4	S17	Lublin – Piaski	Piaski/Przejście	0.000	1.040	asphalt
5	A1	Częstochowa – Katowice	Interchange Woźniki/DW789/ – Interchange Pyrzowice/S1/	395.336	399.837	concrete
6	S8	Warszawa – Piotrków Trybunalski	Interchange Wolbórz – Interchange Tomaszów Mazowiecki Południe	340.303	348.608	concrete
7	S7	Kraków – Widoma	Wesoła/Widoma/ – Kraków	642.514	657.897	concrete
8	S8	Sieradz – Wrocław	Interchange Złoczew – Sieradz Południe	148.668	168.594	concrete

elemental composition on a Shimadzu EDX 7000 energy-dispersive X-ray fluorescence spectrometer. Each time, approximately 15 g of pre-dried material was poured into a cup intended for solid samples (including bulk materials). The sample did not need to be crushed before testing. Instrument settings: 10 mm collimator, air atmosphere, total radiation exposure time to the sample 60 s. The results obtained were aligned to 100% (semi-quantitative analysis). The data obtained from the elemental composition analysis of road dust with a fraction <0.1 mm is summarised in Table 2.

To show whether the differences in elemental concentrations obtained in the road dust studies were statistically significant, a *t*-Welch test was performed. This is a statistical test of the equality of expected values in two populations. The *t* statistic is described by the following formula (Welch, 1947):

$$t = \frac{\overline{x_1} - \overline{x_2}}{\sqrt{\frac{s_1^2}{N_1} + \frac{s_2^2}{N_2}}} \tag{1}$$

where: x_i – average of the *i*-th sample; s_i^2 – variance in the *i*-th sample; N_i – *i*-th sample size.

Once the *t*-value has been calculated, it is possible, using a Student's *t*-distribution with the calculated number of degrees of freedom, to find the probability of the null hypothesis that the two populations have equal expected values (using a two-sided confidence interval) or the null hypothesis that the mean of one population is greater than or equal to the other (using a one-sided interval) (Welch, 1947).

Then, to investigate the influence of anthropogenic sources on the concentrations of all elements, the so-called enrichment factors (*EF*) were calculated. The enrichment factor values illustrate the relative increase or decrease in element content depending on the geochemical background and in relation to reference elements (Duczmal-Czernikiewicz & Suchan, 2015). In order to calculate enrichment factors, use was made of Mueller's equation (Muller, 1979):

$$EF = \frac{\frac{C_n}{C_{ref}}}{\frac{B_n}{B_{ref}}} \tag{2}$$

where: C_n – the content of the tested element in the test medium; C_{ref} – the content of the reference element in the test medium; B_n – the content of the tested component in the reference medium (background); B_{ref} – the content of the reference element in the reference environment

Aluminium was taken as the reference element, which is one of the typical markers of the dust of mineral origin (Lazo et al., 2018; Pastuszka et al., 2010; Sardans & Peñuelas, 2005). Values for the upper crust were taken as reference values for concentrations of B_n, B_{ref} (Wedepohl, 1995). The following are ranges of coefficient values for different degrees of enrichment (Liu et al., 2023):

- $EF \leq 1$ no enrichment,
- $1 < EF \leq 2$ slight enrichment,
- $2 < EF \leq 5$ moderate enrichment,
- $5 < EF \leq 20$ considerable enrichment,
- $20 < EF \leq 40$ very high enrichment,
- $EF > 40$ extremely high enrichment.

RESULTS AND DISCUSSION

The carried-out tests made it possible to determine the relative mass contribution of the elements present for each of the 16 samples. The results for dust with a fraction <0.1 mm taken at a given measurement point (1–8) from the left and right side of the road were averaged. In addition, average values for each element and type of surface course (asphalt/concrete) were also obtained (Table 2). Figures 3 and 4 show the groups of elements whose average mass contribution to road dust from all measurement points (1–8) The comparison was made according to the type of surface course asphalt (points 1–4) and concrete (points 5–8).

An analysis of Figures 2 and 3 shows that the average mass share of elements in road dust with a <0.1 mm fraction collected from the two surface course types is comparable for Cr, Mn, Ni, Rb, Sr, Y and Zr. For the asphalt surface courses, higher mass shares in road dust were obtained for Cu and V, while for the concrete surface courses, Pb and Zn. In addition, Tm was present in road dust collected only from the concrete surface course, and Pd in dust from the asphalt surface course. However, it is worth noting that the mass shares of Mn, Rb, Sr and Y are almost the same for dust from both surface course types, while for the other elements (Cr, Cu, Ni, Pb, V and Zr) the differences in the obtained mass shares are insignificant (except for Zn).

To show whether the differences in concentrations shown in Figures 2 and 3 were statistically significant, a *t*-Welch test was performed. The results are shown in Table 3. None of the -values obtained indicates statistically significant differences between asphalt and concrete surface courses. This may indicate that the average elemental composition of dust is not affected by surface course type.

Table 2. Mass share of elements identified in road dust with <0.1 mm fraction at eight measurement points for asphalt and concrete surface courses. Concentrations below the detection limit are denoted as <dl

Road dust with a particle size of <0.1 mm										
Measurement points	Asphalt paving (A)					Concrete paving (C)				
	1	2	3	4	Average	5	6	7	8	Average
Road No.	A2	S7	S8	S17		A1	S8	S7	S8	
Selected sections	Warszawa – Łódź	Kielce by-pass	Wrocław - Sieradz	Lublin - Piaski		Częstochowa - Katowice	Warszawa – Piotrków Trybunalski	Kraków - Widoma	Sieradz - Wrocław	
Elements [%]										
Al	2.178	1.927	3.442	2.517	2.516	3.036	1.890	2.148	2.740	2.453
Ca	24.889	42.974	24.954	23.978	29.199	18.849	29.350	43.071	24.439	28.927
Cr	0.128	0.070	0.143	0.080	0.105	0.060	0.136	0.046	0.119	0.090
Cu	0.218	0.087	0.205	0.108	0.154	0.085	0.176	0.085	0.205	0.137
Fe	17.592	9.306	17.672	9.851	13.605	9.705	17.216	9.088	16.946	13.239
K	4.138	3.557	4.509	5.385	4.398	6.549	3.566	3.827	4.742	4.671
Mn	0.398	0.335	0.429	0.304	0.366	0.278	0.428	0.332	0.403	0.360
Ni	0.035	<dl	0.052	<dl	0.044	<dl	<dl	<dl	0.055	0.055
Pb	0.071	0.046	0.095	<dl	0.071	<dl	0.093	<dl	0.091	0.092
Pd	<dl	0.116	<dl	<dl	0.116	<dl	<dl	<dl	<dl	<dl
Rb	0.040	0.038	0.046	0.040	0.041	0.052	0.035	0.039	0.053	0.045
S	1.627	0.551	1.260	0.922	1.090	0.894	2.256	0.493	1.384	1.257
Si	44.618	39.370	43.712	53.674	45.344	57.826	40.785	40.045	44.248	45.726
Sr	0.151	0.087	0.141	0.110	0.122	0.101	0.156	0.100	0.128	0.121
Ti	2.611	1.499	2.506	2.124	2.185	2.001	2.343	1.352	2.366	2.015
Tm	<dl	<dl	<dl	<dl	<dl	<dl	<dl	<dl	0.898	0.898
V	0.077	<dl	0.072	0.071	0.073	<dl	<dl	0.048	0.065	0.056
Y	0.024	0.013	0.029	0.017	0.021	0.018	0.021	0.012	0.025	0.019
Zn	0.930	0.205	1.039	0.444	0.654	0.122	1.609	0.148	1.459	0.834
Zr	0.745	0.258	0.517	0.488	0.502	0.442	0.630	0.309	0.524	0.476

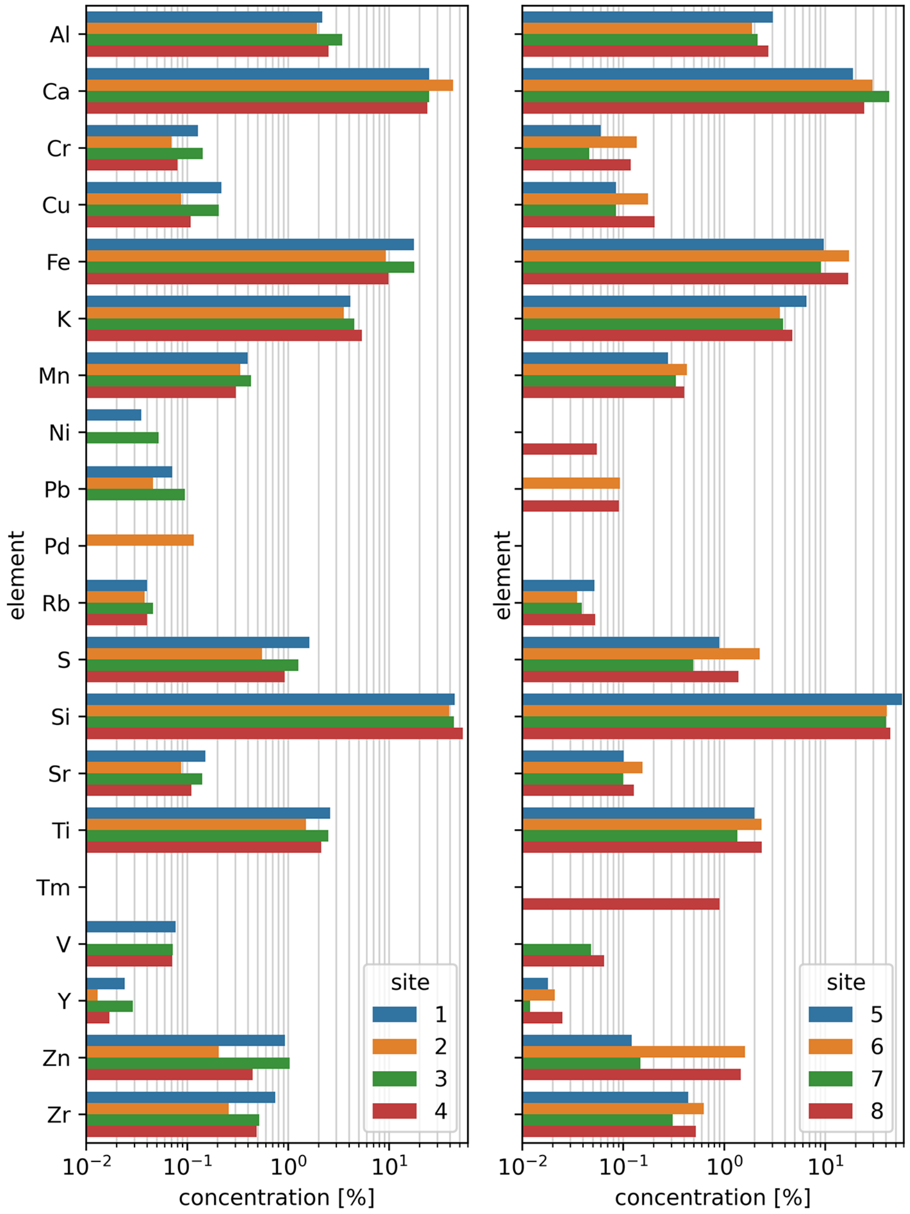

Figure 2. Average mass fraction (%) in road dust with fraction <0.1 mm for asphalt (left panel) and concrete (right panel)

In order to establish the influence of anthropogenic sources on element concentrations in road dust, an analysis of EF enrichment factors was carried out (Table 4).

Road dust of a <0.1 mm fraction collected from asphalt surface courses was extremely enriched in Cu, Cr, Pb and S, while from concrete surface courses in Zn and Zr. This demonstrates the strongly anthropogenic origin of these elements, which, according to (Aguilera et al., 2021; Rogula-Kozłowska et al., 2013) may originate from exhaust gases. In addition, irrespective of the type of surface course, a very high enrichment of dust has been recorded in Ca, Mn, Ni, S, Ti and Y, the source of which is most likely dust from vehicle tyre abrasion (Dziubak, 2021). For road dust collected from both types of road, the most similar EF values have been found for Fe, K, Mn, Si, Sr and Ti. The source of these elements may be roadside soil (Rogula-Kozłowska, 2015; Rogula-Kozłowska et al., 2013). It is worth noting that each of the elements tested (Ca, Cr, Cu, Fe, K, Mn, Ni, Pb, Rb, S, Si, Sr, Ti, V, Y, Zn and Zr) enriched road dust collected from both asphalt and concrete surface courses. It follows that the type of surface course is not the main factor determining the composition of road dust with a fraction <0.1 mm.

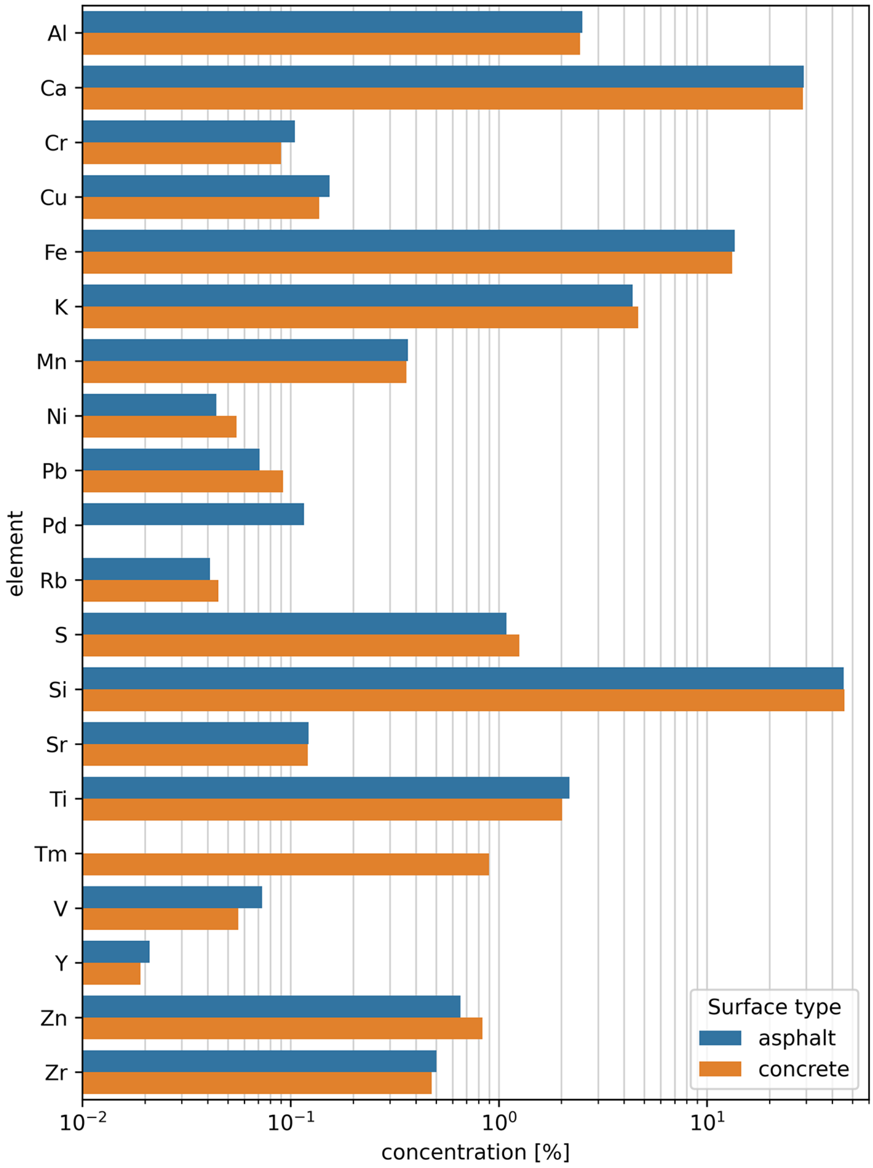

Figure 3. Average mass fraction (%) in road dust with <0.1 mm fraction for asphalt and concrete surface courses

CONCLUSIONS

Based on an analysis of the elemental composition of the dust of a <0.1 mm fraction collected from asphalt and concrete roads with the highest technical and performance characteristics, it was proven that the mass proportions of Al, Ca, Cr, Fe, Mn, Ni, Rb, Si, Sr, Y and Zr in the samples studied for both types of surface courses were almost the same. In the dust collected from the asphalt surface course, the higher mass proportions were found for Cu, Ti and V, while for the concrete surface course, the mass proportions were for K, Pb, S and Zn. The Tm was present in road dust collected only from concrete surface courses, and Pd in dust from asphalt surface courses. The mass shares of Ca, Fe, Mn, Rb, Si, Sr and Y in the road dust are comparable for both surface course types, while for the other elements (Al, Cr, Cu, K, Pb, S, Ti, V, Zn and Zr) the differences in the ascertained mass shares were insignificant (except for Zn).

In addition, road dust of a <0.1 mm fraction collected from asphalt surface courses was extremely highly enriched in Cu, Cr, Pb and S, while that from concrete surface courses was enriched in Zn and Zr, indicating a strongly anthropogenic origin of these elements; it is likely that exhaust gases are their source. Irrespective of the type of surface course, very high dust enrichment is found for Ca, Mn, Ni, S, Ti and Y; these may originate from the abrasion process of

Table 3. Results of the *t*-Welch test with equal means for dust with a fraction <0.1 mm from asphalt and concrete surface courses

Elements	Asphalt – Concrete
Al	0.38
Ca	0.55
Cr	0.07
Cu	0.18
Fe	0.42
K	0.62
Mn	0.53
Pb	0.73
Rb	0.20
S	0.39
Si	0.92
Sr	0.68
Ti	0.01
V	0.09
Y	0.14
Zn	0.71
Zr	0.74

Table 4. EF coefficients for elements determined in road dust with a fraction <0.1 mm, depending on the type of surface course (asphalt/concrete)

Elements	$EF_{Asphalt}$	$EF_{concrete}$
Ca	26	31
Cr	103	81
Cu	375	321
Fe	15	15
K	5	6
Mn	22	23
Ni	21	9
Pb	87	46
Rb	9	14
S	41	38
Si	5	5
Sr	12	12
Ti	23	21
V	18	12
Y	31	24
Zn	503	645
Zr	65	67
EF≤1	no enrichment,	
1<EF≤2	slight enrichment,	
2<EF≤5	moderate enrichment,	
5<EF≤20	considerable enrichment,	
20<EF≤40	very high enrichment,	
EF>40	extremely high enrichment	

vehicle tyres. For road dust collected from both types of roads, the most similar EF values were found for Fe, K, Mn, Si, Sr and Ti. The source of these elements may be roadside soil. Each of the elements obtained in the study (Al, Ca, Cr, Cu, Fe, K, Mn, Ni, Pb, Rb, S, Si, Sr, Ti, V, Y, Zn and Zr) enriched the road dust collected from both asphalt and concrete surface courses. It follows that the type of surface course is not the main factor determining the composition of road dust with a fraction <0.1 mm.

The conducted research will fill a gap in the current state of knowledge regarding the emission of pollutants generated during surface course abrasion and will contribute to at least a qualitative assessment of its contribution to road dust. The information obtained can be used to improve measures taken to reduce dust emissions accumulated in the vicinity of roads and contribute to the design of surface courses that are less susceptible to abrasion and therefore have a lower environmental impact.

It is evident that the information on road dust does not fully exhaust this issue. It follows that further research into this material is very important, especially as this topic is still under-recognised in Poland and needs to be analysed in a reliable manner.

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