

ANNA HOŁTRA (ORCID: 0000-0001-7999-3719)<sup>1</sup>

DOROTA ZAMORSKA-WOJDYŁA (ORCID: 0000-0002-0111-1816)

## SINGLE- AND MULTI-ELEMENTAL POLLUTION INDICES AS A USEFUL TOOL FOR THE EVALUATION OF ROAD DUST CONTAMINATED BY TRACE ELEMENTS

The challenge still is to quantify the impact of traffic volume, road type, age of the road, and the presence of road infrastructure on the content of trace elements in road dust. Samples of road dust were collected at the intersections and roundabouts of the main roads of Wrocław and the county, where traffic was accelerated. This research aimed to identify the level of Cu, Zn, Pb, Cr, Ni, and Cd in road dust, and determine the quality of road dust using individual and integrated pollution indices. A strong anthropogenic influence was noted, which causes a heterogeneous distribution of metal concentrations in the tested road dust. The principal component analysis (PCA) analysis showed that traffic intensity affects mainly the metal content in road dust. The strong Spearman's correlations indicate the connection of trace elements to road traffic. Individual and integrated pollution indices confirm the enrichment of road dust with tested elements, although the permissible metal concentrations according to the Polish standard were not exceeded. A medium or high probability of road dust toxicity was recorded.

### 1. INTRODUCTION

Road dust is used as an indicator of potential environmental pollution with trace elements. It is a conglomerate of various substances in different amounts, containing particles from brake pad and disc wear, road surface wear, and winter tires, which are more susceptible to abrasion than summer tires, metal chains applied to car tires, pavement paints, anti-knock agents added to fuels, fuel leaks, car exhaust emissions, engine oil and lubricants leaks, and exhaust catalyst wear [1–3]. De Miguel et al. [4] presented the classification of road dust based on its chemical composition. The first group of dust

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<sup>1</sup>Wrocław University of Science and Technology, Faculty of Environmental Engineering, Wybrzeże Wyspiańskiego 27, 50-370 Wrocław, Poland, corresponding author A. Hołtra, email address: [anna.holtra@pwr.edu.pl](mailto:anna.holtra@pwr.edu.pl)

includes natural elements (Ga, La, Mn, Sr, Th, Y), the origin of which should be seen in the erosion of urban soils and also in the mineralogical composition. The second group includes chemical elements found in urban areas (Ba, Cd, Co, Cu, Mg, Pb, Sb, Ti, and Zn) of different anthropogenic origins, e.g., from the linear emission of communication pollutions, construction sites, and the demolition of buildings and structures containing cement and other construction materials. The third group of dust includes elements such as Ca, Ni, Fe, Mo, Cs, Rb, Sr, and U, which are both of natural and anthropogenic origin.

In Poland, the emission of the following eight trace elements is inventoried: As, Cd, Cr, Cu, Hg, Ni, Pb, and Zn. The transport sector in Poland is responsible for 37% of copper emissions, which comes mainly from tribological processes (brake pads wear). The increase in copper emissions is related to a 287% increase in the number of vehicles since 1990 and, consequently, an increase in vehicle kilometers traveled. In 2019, the share of the transport sector in the total emission of other monitored trace elements in Poland was as follows: 10% of Cr, 7% of Zn, 3% of Pb, 1% of Hg, more than 0.5% of Ni, As and Cd. Comparing the data from 2019 and 1990, there was an increase in the emission of trace elements in the air from the transport sector: fourfold for Cu (75.30 Mg), Zn (27.73 Mg), and As (0.11 Mg); threefold for Hg (0.11 Mg); 2.5 times for Cd (0.05 Mg); and twofold for Ni (0.57 Mg) and Cr (3.56 Mg). The exception was lead, with a 94% decrease in its emission [5]. The main cause of zinc emissions is the wear of car tires and galvanized road infrastructure elements. The increase in truck share in road traffic contributes to the emission of road dust with a higher zinc content compared to copper. Lead emissions are responsible for exhaust gases, despite the use of unleaded gasoline, and the wear processes of brake linings and car tires occurring during car traffic [6]. The sources of chromium and nickel emissions are more diversified and are released into the environment with exhaust gases, wear of the tread of car tires, wear of friction elements of the brake system and clutch, and abrasion of road surfaces [7]. Furthermore, the increased level of chromium in road dust is caused by corrosion of the chassis and body parts of the car [8, 9]. Currently, antimony from brake pads, manganese from fuels, and platinum metals from catalysts are noted on new roads [10]. To a lesser extent, environmental pollution with trace elements is associated with losses of fuels, oils, lubricants, and operating fluids. A serious source of metals is abrasion and leaching of road surfaces, corrosion of metal elements of road equipment, and the use of road maintenance measures that intensify metal release processes, e.g., salt promotes corrosion of vehicle elements and the road infrastructure [11].

According to Budai and Clement [12], the emission from the wear of vehicle components and road surfaces is responsible for 57% of total copper and 65% of total zinc from road transport in Hungary. In 1999, it was estimated that in the USA 10 Gg of zinc was released as a result of the abrasion of car tires. In the period of 63 years (from 1936 to 1999), 285 Gg of zinc was released [13]. Hulskotte et al. [14] report that in Europe, the annual copper emission from the wear of brake system components is estimated at

almost 2.4 Gg. According to Pulles et al. [3], road transport in the European Union countries was responsible for 6% of the total lead emissions into the air. In 2019, Poland presented that 3.30% of Pb originated from motor transport (auto tire and brake wear) [5]. Many authors confirm the fact that metals released from road transport occur in road dust and pollute the soil in the road area [15–17].

The study aimed to assess the contamination level of road dust by trace elements at the busiest intersections, roundabouts, and exits from main roads in the city of Wrocław and the surrounding area with various conditions of car traffic and periods of road use, and using individual and integrated pollution indices.

## 2. MATERIALS AND METHODS

*Selected roads and a monitored area.* Transport routes of varying age and traffic volume located in Wrocław (western Poland) were selected for this study. Municipal traffic monitoring shows that the least traffic is on the Eastern Bypass of Wrocław No. DW372 (in Polish: *Wschodnia Obwodnica Wrocławia*, WOW), because the road is narrow with one lane in both directions [18]. At the WOW, the traffic volume is an average of 19 536 vehicles per day (median of 19 868; minimum of 17 630; maximum of 21 109). The WOW runs along the southern and eastern borders of Wrocław, occasionally passing through the city area. At the time of the study, the WOW had been in use for 10 years.

The traffic intensity on the other discussed roads in Wrocław is similar but higher than on the WOW. Along the Wrocław Downtown Bypass (in Polish: *Śródmiejska Obwodnica Wrocławia*, SOW), surrounding the oldest part of the city, there were an average of 40 418 vehicles per day (median of 41 571; minimum of 26 224; maximum of 55 715). The SOW has been in use for over 50 years at selected sampling points. It is a two-lane road and is used to distribute internal car traffic between the districts of Wrocław city. Between the communication routes are tram tracks and a green belt. There are additional driving lanes in the places of exits and intersections.

The average daily traffic volume at the Wrocław Motorway Bypass No. A8 (in Polish: *Autostradowa Obwodnica Wrocławia*, AOW) had 35 107 vehicles (median of 35 110; minimum of 25 648; maximum of 48 384). At intersections of access roads, the traffic volume was 39 447 vehicles per day (median of 36 509; minimum of 16 140; maximum of 64 945). The AOW, which is three-lane in each direction to the four-lane at the road exit, has been used for 12 years and runs along the western and northern borders of the Wrocław agglomeration. The AOW and the WOW are to soon create a closed O-ring with a diameter of 15–20 km around Wrocław to reduce the car traffic in the city. In the monitored area, selected roads cut through the agricultural land, and there are no significant industrial plants in the area.

Furthermore, the dust sampling was carried out on the oldest of the roads in Wrocław city, Legnicka Street, which is the main exit artery of Wrocław and connects the western part of the city with the city center. At Legnicka Street, the traffic volume is an average of 39 447 vehicles per day (median of 36 509; minimum of 16 140; maximum of 64 945). This road was rebuilt after World War II and is currently three-lane in each direction, with traffic lights configured to create a green wave.

The road surface on all roads is asphalt, and the road lanes are separated from the roadside by a curb. Along the road, there is a surface water runoff separated by a curb. On the WOW and AOW, transit traffic is accelerated.

*Sampling points.* A total of 8 locations along Legnicka Street in Wrocław city (Nos. 1–8), 10 locations at the SOW (Nos. 9–18), 7 locations at the WOW (Nos. 19–25), and 5 locations at the AOW (Nos. 26–30) were selected (Fig. 1). Road dust samples were collected in areas where there is a slowdown in motor vehicle traffic due to the presence of roundabouts or large intersections, with or without traffic lights, as well as at the exits from main roads.

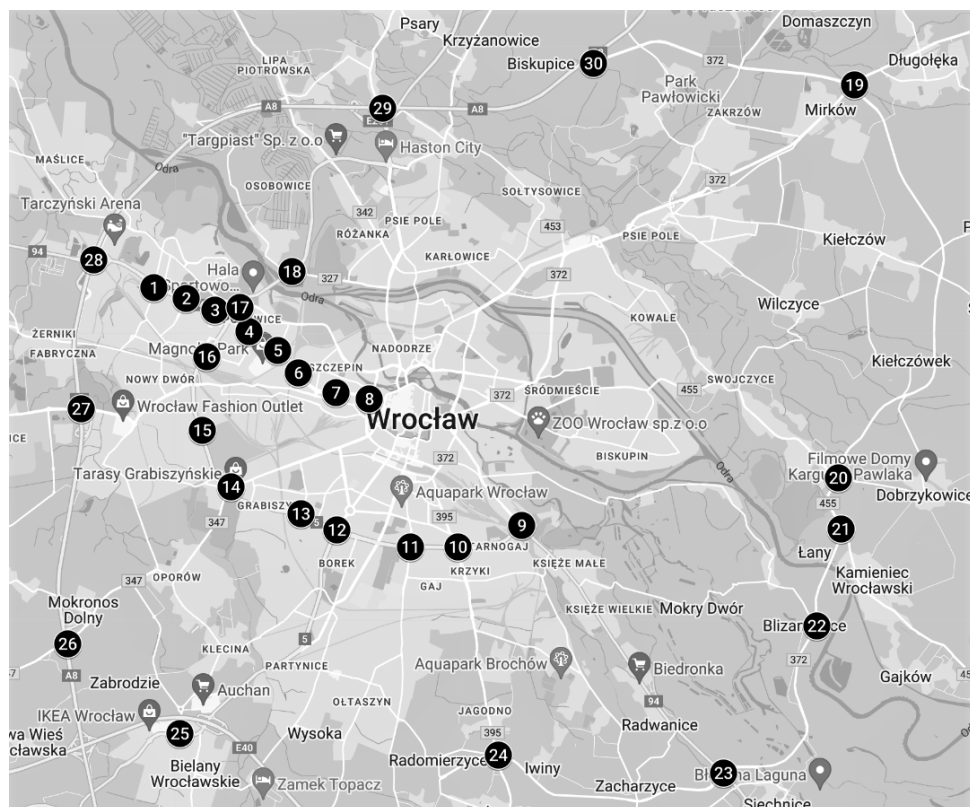


Fig. 1. Google map with road dust sampling sites

*Laboratory analysis.* Road dust sampling took place in the period February–March 2022, when a deficit of precipitation and no snow cover were recorded. Samples were collected at the edge of the road lane, separated by a curb from the roadside, on road sections with a maximum length of 5 m. Three samples were collected at each intersection or roundabout. After homogenization, they weighed around 150 g. The collection of the research material was carried out using a nylon broom, and road dust was scooped up on a plastic dustpan. The air-dried road dust was sieved through a nylon sieve with a mesh of 1 mm to remove larger fractions. A total of 90 representative road dust samples were collected on the main roads of Wrocław County.

The wet microwave digestion of road dust was carried out in the TFM vessels with a volume of 100 cm<sup>3</sup>. About 0.2 g of the homogenized sample was digested in 8 cm<sup>3</sup> of concentrated HNO<sub>3</sub>, 65% Suprapure (Sigma-Aldrich). Each batch of samples for the microwave digestion system (START D, Milestone) contained a reagent sample without any test material. The digestion system was operated in two steps: the first digestion step was carried out for 2 min at 140 °C, and the second one for 15 min at 220 °C. The process of mineralization in a closed system was operated under a pressure of 20 MPa and was aided by microwaves with the power of 800 W. Solutions after mineralization were quantitatively transferred to 50 cm<sup>3</sup> volumetric flasks, with the filter paper being washed with deionized water. Obtained filtrates were analyzed for six trace element contents (Cu, Zn, Pb, Cr, Ni, and Cd) of the FAAS (flame atomic absorption spectrometry) or GFAAS (graphite furnace atomic absorption spectrometry) methods using a Thermo Solaar iCE 3500 device (Thermo Scientific) according to PN-ISO 11047: 2001 [19]. The CRM solutions of selected metals (Sigma Aldrich) were used in this research for the calibration curve method, as components in the method of standard addition, and also for the validation of analytical methods. All measurements and analyses were carried out three times, and the acidic microwave digestion of road dust was performed twice. The reagent blank samples were used to check the instrument readings. The limits of detection were estimated based on three times the standard deviation for digestion blanks. The accuracy of the determination was controlled using the method of standard addition. The percentage of the recovery was 94–98%. The AAS results were verified through the standard deviation, the coefficient of variation, and the confidence interval. The accepted results of the coefficient of variation for a given point were within 10%. Statistical analysis was made based on the *t*-student test, with the number of degrees of freedom equaling 5 and the *p*-value being 0.05.

*Statistical analysis.* The contents of trace elements in the soil were calculated as average values with the standard deviations for six independent environmental samples from one sampling site. The hierarchical cluster analysis (HCA) was performed with Ward's algorithm and the squared Euclidian distance. The average values of the pollution index for a given location were used. The statistical analysis was carried out using the program Statistica. The data was checked for a normal distribution of the Shapiro–Wilk's *W*-test. Tests of significance were made at the 95% confidence level. The Mann

–Whitney  $U$ -test was used for the data which did not show a normal distribution. The Spearman's rank correlation coefficients were calculated for elemental concentrations for all samples. The correlations were considered strong when higher than 0.7, and the  $p$ -value was 0.05. Pearson's correlation coefficient ( $p < 0.05$ ) was used to explore relationships between the total content of the analyzed trace elements.

Table 1

Single-elemental and multi-elemental pollution indices [20]

Parameter	Formula	Class	Classification
Geoaccumulation index	$I_{\text{geo}} = \log_2 \frac{C_n}{1.5B_n}$	0	<0 unpolluted
		1	0–1 unpolluted to moderately polluted
		2	1–2 moderately polluted
		3	2–3 moderately to highly polluted
		4	3–4 highly polluted
		5	4–5 highly to extremely polluted
		6	>5 extremely highly polluted
Pollution index	$PI = \frac{C_n}{B_n}$	1 2 3 4 5	>1 no pollution 1–2 low pollution 2–3 moderate pollution 3–5 strong pollution >5 very strong pollution
Average pollution index	$PI_{\text{avg}} = \frac{PI_{\text{sum}}}{n}$		
	$PI_{\text{sum}} = \sum_n PI$		
Vector modulus of pollution index	$PI_{\text{vec}} = \left( \frac{\sum PI^2}{n} \right)^{1/2}$		
Pollution load index	$PLI = (PI_1 PI_2 \dots PI_n)^{1/n}$		
		2	1 baseline levels of pollution
		3	>1 deterioration of quality
Nemerow pollution index	$PI_{\text{Nem}} = \left( \frac{PI_{\text{avg}}^2 + PI_{\text{max}}^2}{2} \right)^{1/2}$	1	>0.7 clean
		2	0.7–1 warning limit
		3	1–2 slight pollution
		4	2–3 moderate pollution
		5	>3 heavy pollution
Background enrichment factor	$PIN = \sum_n \frac{PI_{\text{class}}^2 C_n}{B_n}$	1	0–7 clean
		2	7–95.1 trace contaminated
		3	95.1–518.1 lightly contaminated
		4	518.1–2548.5 contaminated
		5	≥2548.5 highly contaminated
Potential ecological risk	$RI = \sum E_r$	1	<90 low
		2	90–180 moderate
		3	180–360 strong
		4	360–720 very strong
		5	>720 highly to strong

Table 1

Single-elemental and multi-elemental pollution indices [20]

Parameter	Formula	Class	Classification
Ecological risk factor	$E_r = T_r \cdot PI$	1	<40 low
		2	40–80 moderate
		3	80–160 considerable
		4	160–320 high
		5	>320 very high
Contamination security index	$CSI = \sum_n \left( W \left( \left( \frac{C_n}{ERL} \right)^{1/2} + \left( \frac{C_n}{ERM} \right)^2 \right) \right)$	1	Contamination severity: <0.5 uncontaminated
		2	0.5–1 very low
		3	1–1.5 low
		4	1.5–2 low to moderate
		5	2–2.5 moderate
		6	2.5–3 moderate to high
		7	3–4 high
		8	4–5 very high
		9	>5 ultrahigh
Probability of toxicity	$MERMQ = \frac{\sum C_n}{n \cdot ERM}$	1	>0.1 low risk level 9%
		2	0.1–0.5 medium risk level 21%
		3	0.5–1.5 high risk level 49%
		4	>1.5 very high risk level 76%

$C_n$  – concentration of a trace element, mg/kg.

$B_n$  – reference UCC background, mg/kg: 28 (Cu), 67 (Zn), 17 (Pb), 47 (Ni), 92 (Cr), 0.09 (Cd) after [21].

$n$  – number of studied trace elements ( $n = 6$ ).

$T_r$  – toxic-response factor: Cu (5), Zn (1), Pb (5), Ni (5), Cr (2), Cd (30) after [22].

$W$  – computed weight of an element: 0.075 (Cu, Zn), 251 (Pb), 215 (Ni), 134 (Cr), 0.25 (Cd).

$ERL$  (effect range) low/ $ERM$  (effect range medium): 34/270 (Cu), 150/410 (Zn), 46.7/218 (Pb), 20.9/51.6 (Ni), 81/370 (Cr), 1.2/9.6 (Cd)

*Pollution indices.* A set of commonly used environmental pollution indices (Table 1) was discussed by Kowalska et al. [20]. They were calculated based on the elemental composition of the upper continental crust (UCC), as recommended by Rudnick and Gao [21]. The values of the UCC reference background and pollution indices provide a universal character for the presented results. In our research, trace elements (Cu, Zn, Pb, Cr, Ni, and Cd) were selected in road dust, which contribute to the high contamination of soils in the vicinity of middle-aged to older-aged roads.

### 3. RESULTS AND DISCUSSION

#### 3.1. LEVEL OF CONTAMINATION AND ASSESSMENT

The results of the principal component analysis (PCA) were based on 30 cases describing the sampling points of road dust.

Table 2

The loadings of environmental parameters on the first four axes of PCA

Environmental variables	<i>AX1</i>	<i>AX2</i>	<i>AX3</i>	<i>AX4</i>
Cu, mg/kg	<b>0.872919</b>	0.355787	0.092869	0.012386
Zn, mg/kg	<b>0.616453</b>	0.538978	0.187054	0.056380
Pb, mg/kg	0.034393	<b>0.584643</b>	<b>0.589084</b>	0.449970
Cr, mg/kg	0.364966	0.436277	0.270137	<b>0.732808</b>
Ni, mg/kg	<b>0.781318</b>	0.214361	0.235088	0.048772
Cd, mg/kg	<b>0.701332</b>	0.387113	0.325472	0.176989
Traffic volume, cars/day	<b>0.703942</b>	0.533559	0.310088	0.122117
Road age, year	0.250536	<b>0.765994</b>	0.463538	0.137394

The values with the highest variation percentages are bold.

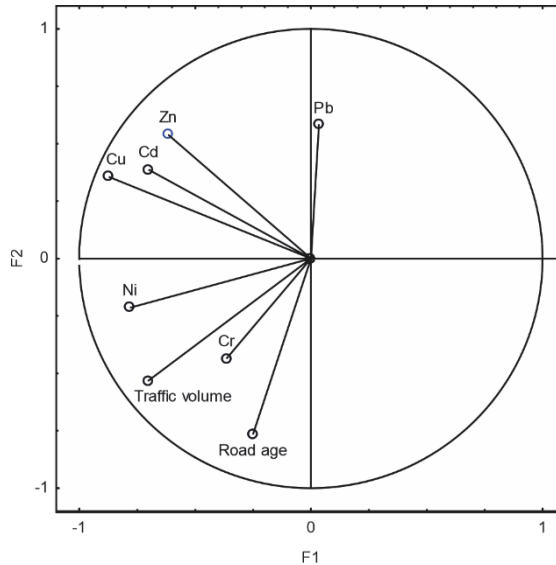


Fig. 2. Results of principal component analysis performed on 8 environmental characteristics

The principal component analysis, based on 8 environmental variables, shows two main gradients ( $F1$  and  $F2$  factors) of the environment (Table 2, Fig. 2) for the study area. The first gradient (factor  $F1$ ), describing 37% of the variance in environmental characteristics, was associated with a strong influence of heavy metal concentrations in road dust (Cu, Ni, Cd, and Zn) and with the parameter characterizing traffic volume. The first ordination axis ( $AX1$ ) was negatively correlated with all the mentioned variables. The second gradient (factor  $F2$ ) was associated with the content of Pb in road dust and the road age, which means how long it had been used. The second ordination axis ( $AX2$ ), describing 25% of the variance, was positively correlated with Pb content and



negatively correlated with the time of road use. The third ordination axis ( $AX3$ ), describing 12% of the variance, was inversely correlated with the second ordination axis ( $AX2$ ) in terms of Pb content. The fourth ordination axis ( $AX4$ ), describing 10% of the variance, was negatively correlated with Cr content in road dust.

The results of the principal component analysis indicate that road age was not a factor affecting the distribution of trace element concentrations in road dust. It can be explained by the short time of deposition of road dust at the edge of the road due to leaching by rainfall and mechanical cleaning of the road surface. Traffic volume affects the content of metals in road dust, except for lead, due to the use of unleaded fuels today.

### 3.2. CORRELATION ANALYSIS

There was strong ( $CV > 45\%$ ) or average ( $CV > 25\%$ ) spatial variability of trace metal content in road dust along main roads in Wrocław. Very strong variability ( $CV > 100\%$ ) was recorded for Cr at Legnicka Street, Pb at the SOW, and Zn at the AOW, pointing to a high heterogeneity. It means the accumulation of road pollutants along the road lane. A low coefficient of variation ( $CV < 25\%$ ) shows similar concentrations of metal in road dust and suggests one common source of the element's emission.

The lack of spatial homogeneity is also confirmed by the  $p$ -value ( $< 0.05$ ) of the Shapiro–Wilk's  $W$ -test. The normal distribution can only be passed for Pb at Legnicka Street (the  $p$ -value equal to 0.475), for Cu at the SOW (0.161), and Ni at the AOW (0.389). The frequency histograms of studied metals in road dust, except for the previously mentioned cases (metals), confirm the deviation from the Gaussian curve. The positive kurtosis in most cases indicates a concentration of scores around the average value and the presence of a few outliers. The positive skewness values suggest that the arithmetic mean values are higher than the median ones. The exceptions are metals with negative kurtosis and/or negative skewness, with small absolute values for both parameters, which is confirmed by a distribution close to the normal for these elements.

In the studied road dust, there were significant correlations ( $p < 0.05$  of Mann–Whitney's  $U$ -test) between the total metal content (Table 3). A strong (0.7–0.9) or moderate (0.4–0.7) Spearman's correlations were identified between all pairs of metals at the WOW, for 50% of metals pairs at the SOW and 30% of metals pairs at the AOW, which may suggest that these metals have a common source of origin related to road traffic. Weak relationships were obtained at Legnicka Street, except for the Zn–Pb (0.58) and Zn–Ni pairs (0.46). Świetlik et al. [9] indicated that the source of copper in road emissions is the abrasion of brakes, tires, and surfaces, and the source of zinc is exhaust emissions from diesel engines and the corrosion of safety fences. The significant Pearson's correlation values are 0.55 for the Cu–Cr pair and 0.43 for the Zn–Cr pair in road dust collected from the noise barrier by the E77 road in Poland. Wawer et al. [23] confirm that the strong Pearson's correlations between analyzed metal pairs from road emissions to soils were due to mainly the wear of brake linings and tires.

Table 3

Spearman's correlation analysis ( $p < 0.05$ )

Location		Cu	Pb	Cr	Ni	Cd
Legnicka Street	Zn	0.14	0.58	-0.08	0.46	-0.23
	Cu	-	0.36	0.21	-0.38	0.04
	Pb	-	-	-0.07	0.33	0.03
	Cr	-	-	-	0.30	-0.30
	Ni	-	-	-	-	-0.22
		Cu	Pb	Cr	Ni	Cd
SOW	Zn	0.72	0.64	-0.02	0.71	0.67
	Cu	-	0.48	-0.25	0.45	0.44
	Pb	-	-	-0.21	0.11	0.26
	Cr	-	-	-	0.29	-0.20
	Ni	-	-	-	-	0.64
		Cu	Pb	Cr	Ni	Cd
WOW	Zn	0.97	0.48	0.87	0.71	0.67
	Cu	-	0.48	0.88	0.70	0.67
	Pb	-	-	0.40	0.78	0.61
	Cr	-	-	-	0.70	0.71
	Ni	-	-	-	-	0.56
		Cu	Pb	Cr	Ni	Cd
AOW	Zn	0.68	0.54	0.34	-0.11	0.68
	Cu	-	0.84	0.02	-0.74	0.69
	Pb	-	-	-0.30	-0.81	0.33
	Cr	-	-	-	0.35	0.25
	Ni	-	-	-	-	-0.17

### 3.3. INDIVIDUAL POLLUTION INDICES

The analysis of individual pollution indices (Table 1) confirms the heterogeneous distribution of metals in the studied area (Tables 4, 5). The highest median values, except for Pb, were recorded at the SOW in the center of Wrocław and the AOW (highway bypass) due to the intensive use of both of these roads (Table 4). The values of the median for pollution indices in road dust for the next two roads are even 2.7 times lower. Legnicka Street with slightly higher metal levels than the WOW correlates with the traffic intensity recorded on these roads [18].

The median values of individual indices ( $PI$  and  $I_{geo}$ ) are ranked as follows:  $Cu > Zn$  or  $Cd > Pb > Ni > Cr$ . The maximum values for both indices indicate the dominance of Cu, and then Cd or Zn in the order, and the further arrangement of  $Pb > Ni > Cr$  is similar to the median values of the  $PI$  and  $I_{geo}$  indices.

The assessment of the degree of road dust pollution based on the median values, considering the geochemical values (the UCC reference background), indicates a very strong, strong or moderately polluted level of Cu (the 5<sup>th</sup> to 3<sup>rd</sup> class of  $PI$ ) or highly to moderately polluted level of Cu (the 3<sup>rd</sup> to 1<sup>st</sup> class of  $I_{geo}$ ) (Tables 4, 5).

Table 4

Descriptive statistics of individual pollution indices

Location	Index	Metal	N	Avg	Med	Min	Max	Quartile		Std. dev.	CV [%]	Skewness	Kurtosis
								Lower	Upper				
Legnicka Street	PI	Cu	24	3.14	2.93	1.81	5.72	2.58	3.27	1.09	34.74	1.60	2.24
		Zn	24	2.83	2.39	1.70	5.44	2.16	3.22	1.13	39.83	1.50	1.38
		Pb	24	1.01	0.99	0.69	1.27	0.95	1.11	0.13	12.67	-0.08	0.63
		Cr	24	0.87	0.45	0.29	3.93	0.40	0.54	1.17	134.52	2.39	4.13
		Ni	24	0.83	0.81	0.57	1.12	0.69	0.99	0.18	21.88	0.11	-1.55
		Cd	24	1.57	1.60	1.26	1.73	1.53	1.66	0.13	8.45	-1.38	1.42
SOW	PI	Cu	30	7.10	7.05	3.43	9.84	6.12	8.57	1.79	25.18	-0.43	-0.26
		Zn	30	4.09	3.58	1.33	7.27	3.09	5.32	1.83	44.74	0.58	-0.69
		Pb	30	0.98	0.63	0.19	4.53	0.29	1.04	1.11	113.02	2.44	5.21
		Cr	30	1.03	1.02	0.55	1.91	0.77	1.11	0.36	34.54	1.20	1.55
		Ni	30	1.60	1.50	0.90	2.65	1.21	1.95	0.52	32.43	0.67	-0.49
		Cd	30	4.55	3.98	1.73	9.11	2.68	5.51	2.42	53.30	0.78	-0.66
WOW	PI	Cu	21	3.17	2.64	1.46	6.69	2.16	4.03	1.64	51.73	1.40	1.03
		Zn	21	2.01	1.74	1.02	4.02	1.34	2.74	0.98	48.99	1.19	0.26
		Pb	21	1.35	1.25	0.46	3.07	0.69	1.59	0.82	60.53	1.10	0.51
		Cr	21	0.48	0.40	0.29	0.86	0.31	0.56	0.19	39.83	1.08	0.20
		Ni	21	0.68	0.62	0.36	1.35	0.50	0.78	0.30	43.54	1.44	1.25
		Cd	21	2.81	1.85	0.82	7.98	1.31	4.03	2.34	83.13	1.56	1.23
AOW	PI	Cu	15	6.11	4.72	2.99	13.58	3.62	5.66	3.97	64.90	1.45	0.51
		Zn	15	8.56	3.82	2.35	29.09	3.57	4.02	10.59	123.71	1.66	0.88
		Pb	15	1.51	1.27	0.91	2.13	1.20	2.05	0.49	32.10	0.24	-1.82
		Cr	15	0.64	0.67	0.54	0.74	0.56	0.69	0.07	11.44	-0.18	-1.79
		Ni	15	0.97	0.94	0.71	1.21	0.92	1.08	0.15	15.91	-0.24	-0.69
		Cd	15	3.53	2.90	1.74	7.52	2.53	2.99	2.09	59.17	1.47	0.60
Legnicka Street	I <sub>geo</sub>	Cu	24	0.99	0.97	0.27	1.93	0.78	1.12	0.44	44.45	0.75	1.06
		Zn	24	0.83	0.67	0.18	1.86	0.52	1.09	0.50	60.08	0.95	0.21
		Pb	24	-0.58	-0.60	-1.12	-0.24	-0.66	-0.43	0.19	-32.70	-0.61	1.75
		Cr	24	-1.41	-1.74	-2.35	1.39	-1.92	-1.48	1.12	-79.36	2.09	3.25
		Ni	24	-0.88	-0.90	-1.41	-0.43	-1.11	-0.60	0.32	-36.30	-0.07	-1.52
		Cd	24	0.06	0.10	-0.25	0.20	0.03	0.14	0.13	207.44	-1.54	1.81
SOW	I <sub>geo</sub>	Cu	30	2.19	2.23	1.19	2.71	2.03	2.51	0.41	18.93	-1.10	1.02
		Zn	30	1.30	1.25	-0.18	2.28	1.04	1.83	0.68	52.43	-0.35	-0.12
		Pb	30	-1.18	-1.25	-2.97	1.60	-2.37	-0.53	1.20	-102.12	0.70	0.31
		Cr	30	-0.61	-0.56	-1.45	0.35	-0.96	-0.43	0.47	-76.36	0.26	0.22
		Ni	30	0.02	0.00	-0.74	0.82	-0.31	0.38	0.46	2282.62	0.14	-0.75
		Cd	30	1.41	1.41	0.21	2.60	0.84	1.88	0.76	54.16	0.14	-1.10
WOW	I <sub>geo</sub>	Cu	21	0.93	0.82	-0.04	2.16	0.53	1.42	0.66	70.96	0.61	-0.07
		Zn	21	0.28	0.21	-0.55	1.42	-0.16	0.87	0.63	229.03	0.64	-0.62
		Pb	21	-0.39	-0.26	-1.72	1.03	-1.12	0.08	0.86	-221.34	0.08	-0.85
		Cr	21	-1.76	-1.89	-2.38	-0.80	-2.28	-1.41	0.53	-30.12	0.58	-0.78
		Ni	21	-1.24	-1.26	-2.08	-0.15	-1.57	-0.95	0.55	-44.44	0.74	0.14
		Cd	21	0.53	0.31	-0.87	2.41	-0.19	1.43	1.02	191.32	0.68	-0.36



All road dust samples exceeded the geochemical background for Cu, Zn, and generally for Cd (Table 4). The maximum exceeding of the UCC background was 14 times for Cu, 29 times for Zn, and more than 9 times for Cd. In 20% of road dust samples, a high ecological risk ( $E_r > 270$ ) of cadmium contamination (Table 5) is also due to the high toxic-response factor ( $T_r$ ) of 30 for this element (Table 1). The remaining samples indicate considerable (25%) or moderate (53%) ecological risk due to cadmium levels. The chemical elements such as Pb, Cr, and Ni do not contaminate the examined road dust in many locations, which is confirmed by the median values of the  $PI$  and  $I_{geo}$  indices, which are close to or below one.

The differences between the  $I_{geo}$  and  $PI$  classes result from different ways of calculating these indices. For the  $I_{geo}$  index, the conversion factor of 1.5 and the logarithmic operation are used to eliminate background variability (Table 5). The pollution indicators show that road dust is contaminated with tested trace elements, even though the permissible metal concentrations for soils from the IV group: 15 (Cd), 500 (Ni), 600 (Cu, Pb), 1000 (Cr), and 2000 (Zn) mg/kg, are not being exceeded, according to the Polish regulations [24].

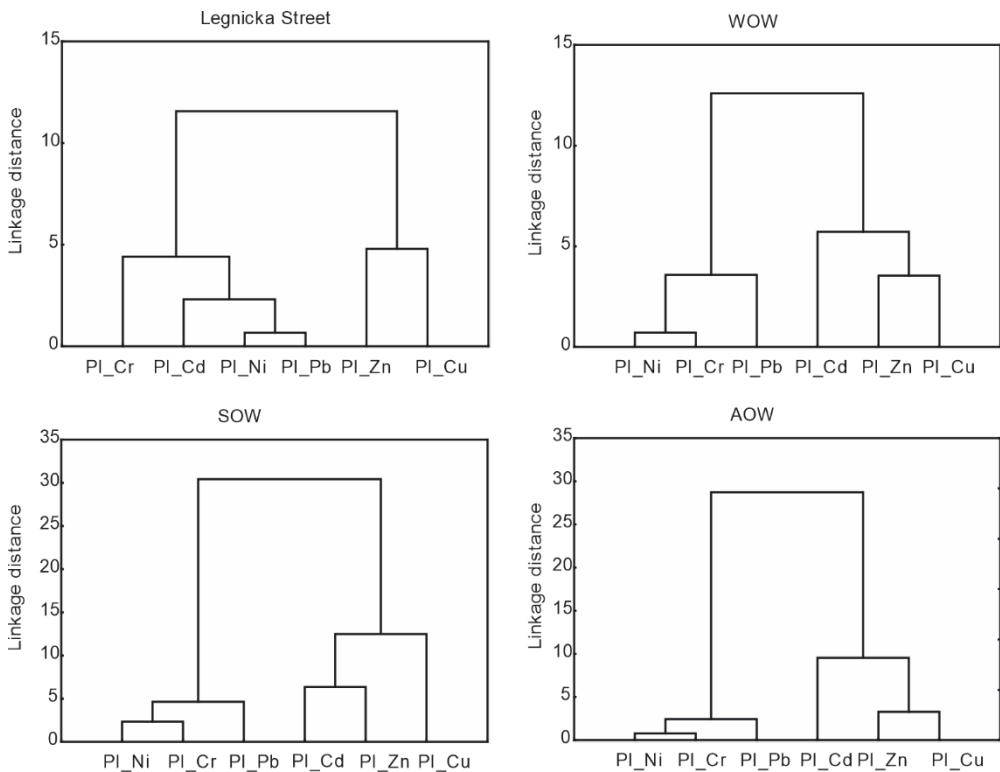


Fig. 3. Dendrograms of single pollution indices (median values)

Identification of trace element clusters in road dust along the communication routes using the HCA method shows a strong aggregation for copper and zinc associated with one main group (Fig. 3). Cadmium at the SOW, AOW, and WOW is associated with this group (Cu and Zn). Legnicka Street with separated cadmium has a different system, which forms a subgroup with the remaining elements (Cr, Ni, and Pb). These three trace elements with much lower *PI* values and a high level of aggregation form a separate group, which is further divided into associated subgroups. The binding of metals in the dendrograms corresponds to the amount of road dust enrichment with trace elements from transport, and their strong Spearman's correlations confirm the common origin of metals (Table 3).

### 3.4. INTEGRATED POLLUTION INDICES

Based on the median of the *CSI* index, there was a little deterioration of road dust quality with trace elements on the studied roads. The *PIN* index confirms this fact, suggesting a higher level of metal contamination in the samples. The defined scale for the *PLI* index does not differentiate sufficiently from the degree of sample contamination; therefore, regardless of the amount of background value exceedance, this parameter classifies each road dust as contaminated with metals. According to the *RI* index, road dust along Legnicka Street and the WOW is slightly polluted with metals, while road dust at the AOW and SOW is moderately polluted.

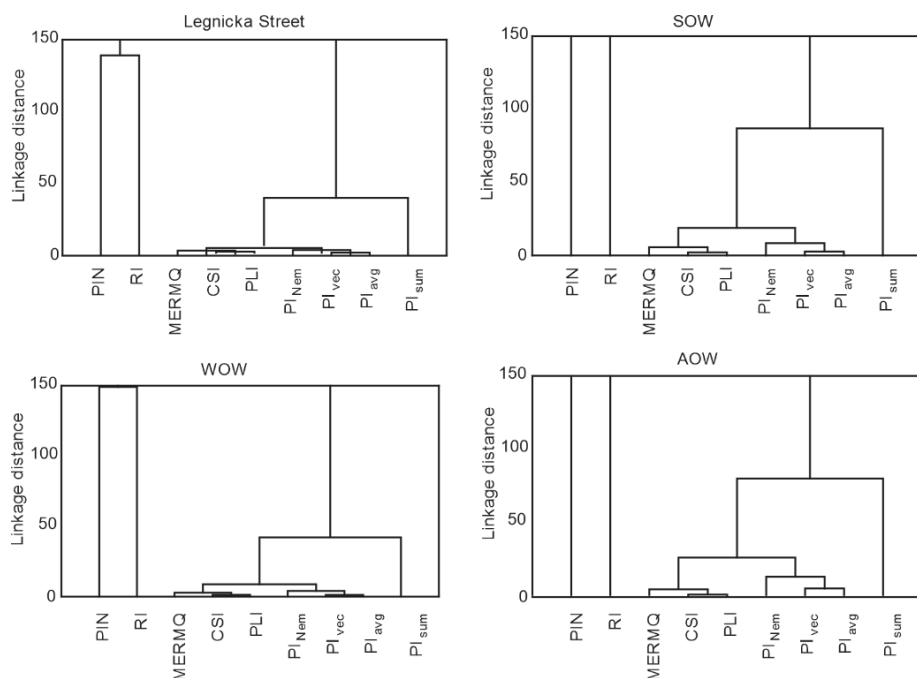


Fig. 4. Dendrograms of integrated pollution indices (median values)

Table 6

Descriptive statistics of integrated pollution indices

Location	Index	N	Avg	Med	Min	Max	Quartile		Std. dev.	CV [%]	Skewness	Kurtosis
							Lower	Upper				
Legnicka Street	$PI_{sum}$	24	10.26	9.93	7.80	12.66	8.68	12.11	1.76	17.19	0.10	-1.56
	$PI_{avg}$	24	1.71	1.66	1.30	2.11	1.45	2.02	0.29	17.19	0.10	-1.56
	$PI_{vec}$	24	2.06	1.94	1.52	2.69	1.71	2.49	0.43	20.77	0.32	-1.32
	$PLI$	24	1.36	1.35	1.05	1.82	1.14	1.49	0.23	17.00	0.66	-0.42
	$PI_{Nem}$	24	2.94	2.71	2.01	4.28	2.31	3.61	0.81	27.43	0.65	-0.96
	$RI$	24	76.66	76.27	61.43	90.76	74.29	80.08	7.57	9.87	-0.30	0.55
	$PIN$	24	101.36	88.55	39.21	177.84	63.38	145.15	49.05	48.39	0.52	-1.06
	$CSI$	24	1.03	1.00	0.85	1.34	0.88	1.13	0.16	15.78	0.69	-0.69
$MERMQ$	24	0.31	0.31	0.23	0.43	0.24	0.36	0.07	22.68	0.56	-1.00	
SOW	$PI_{sum}$	30	19.35	20.46	9.11	27.32	15.98	21.68	5.36	27.72	-0.32	-0.51
	$PI_{avg}$	30	3.23	3.41	1.52	4.55	2.66	3.61	0.89	27.72	-0.32	-0.51
	$PI_{vec}$	30	4.05	4.19	1.82	5.60	3.43	4.67	1.13	27.87	-0.50	-0.40
	$PLI$	30	2.24	2.28	1.13	3.29	1.70	2.63	0.61	27.38	-0.12	-0.74
	$PI_{Nem}$	30	5.78	6.32	2.66	7.67	4.80	6.73	1.44	25.02	-0.87	-0.06
	$RI$	30	190.97	172.34	79.87	330.54	133.33	242.50	77.21	40.43	0.42	-0.92
	$PIN$	30	363.53	380.17	77.45	595.73	281.09	436.61	156.37	43.01	-0.18	-0.60
	$CSI$	30	1.70	1.61	1.06	2.78	1.36	1.95	0.48	28.26	0.78	0.25
$MERMQ$	30	0.54	0.54	0.31	0.83	0.46	0.62	0.14	26.28	0.34	-0.01	
WOW	$PI_{sum}$	21	10.50	8.80	4.59	18.59	6.97	17.00	5.02	47.81	0.68	-1.12
	$PI_{avg}$	21	1.75	1.47	0.76	3.10	1.16	2.83	0.84	47.81	0.68	-1.12
	$PI_{vec}$	21	2.11	1.67	0.86	3.88	1.35	3.69	1.12	52.84	0.76	-1.13
	$PLI$	21	1.35	1.20	0.66	2.43	0.92	1.83	0.57	42.28	0.80	-0.41
	$PI_{Nem}$	21	2.96	2.19	1.17	6.00	1.73	5.20	1.75	58.89	0.90	-0.94
	$RI$	21	113.36	77.97	38.65	273.05	61.49	174.28	77.35	68.23	1.24	0.27
	$PIN$	21	116.67	46.06	12.09	308.81	33.68	289.76	121.27	103.94	0.93	-1.11
	$CSI$	21	0.95	0.89	0.64	1.52	0.72	1.05	0.27	29.02	1.03	0.30
$MERMQ$	21	0.26	0.23	0.15	0.49	0.16	0.29	0.11	42.89	1.33	1.03	
AOW	$PI_{sum}$	15	21.33	14.09	9.77	53.71	13.57	15.41	16.86	79.07	1.62	0.81
	$PI_{avg}$	15	3.55	2.35	1.63	8.95	2.26	2.57	2.81	79.07	1.62	0.81
	$PI_{vec}$	15	4.76	2.81	1.82	13.48	2.61	3.12	4.52	94.85	1.63	0.83
	$PLI$	15	2.19	1.87	1.42	3.99	1.79	1.91	0.94	42.95	1.52	0.68
	$PI_{Nem}$	15	7.05	3.73	2.41	21.52	3.24	4.39	7.49	106.25	1.64	0.84
	$RI$	15	158.71	123.21	81.04	338.63	122.63	129.25	93.97	59.21	1.52	0.68
	$PIN$	15	382.63	167.29	60.93	1270.40	158.05	245.61	461.15	120.52	1.61	0.79
	$CSI$	15	1.61	1.16	1.11	3.37	1.12	1.30	0.91	56.30	1.65	0.87
$MERMQ$	15	0.54	0.37	0.32	1.24	0.36	0.40	0.36	67.01	1.65	0.86	

However, the calculation formula of the  $PI_{Nem}$  index shows a greater degree of pollution, assessing road dust at Legnicka Street and the WOW as moderately polluted, and at the AOW and SOW as very polluted. The  $MERMQ$  index shows the medium risk level for roads in Wrocław, with a 21% probability of toxicity for tested road dust, was determined (Tables 6, 7). However, a high level of risk was also reported, with a 49% probability of toxicity, mainly at the SOW.

Table 7

Classification of dust pollution based on multi-elemental pollution indices in 30 sampling sites

Location	No.	<i>PLI</i>	<i>PI<sub>Nem</sub></i>	<i>PIN</i>	<i>RI</i>	<i>CSI</i>	<i>MERMQ</i>	Legend
Legnicka Street	1							<i>PLI</i>
	2							1, 2 classes
	3							3 class
	4							
	5							<i>PI<sub>Nem</sub></i>
	6							<i>PIN</i>
	7							<i>RI</i>
	8							1 class
SOW	9							2 class
	10							3 class
	11							4 class
	12							5 class
	13							
	14							
	15							<i>CSI</i>
	16							1 class
	17							2 class
WOW	18							3 class
	19							4 class
	20							5 class
	21							6 class
	22							
	23							
	24							<i>MERMQ</i>
	25							1 class
AOW	26							2 class
	27							3 class
	28							4 class
	29							
	30							

Dendrograms in Fig. 4 indicate an aggregation of multi-elemental pollution indices (Table 1) based on their median values. All HCA diagrams show two major groups of integrated pollution indices for the discussed roads in Wrocław, with a high linkage distance level of these clusters ranging from approx. 680 to 2400.

The first group includes the *PIN* and *RI* indices at a linkage distance of 140 to 670. The values of both indices are even up to 100 times higher than the other integrated pollution indices discussed, which results from their calculation formulas.

The second group of indices is further divided into three subgroups based on the concentration parameter, or the *PI* index. All these indicators indicate a strong association at a distance level below 90. The first subgroup comprises the *MERMQ*, *CSI* and *PLI* indices. The second subgroup forms the *PI<sub>Nem</sub>* index, which further connects to the



$PI_{\text{vec}}$  and  $PI_{\text{avg}}$  indices. A separate third subgroup includes the  $PI_{\text{sum}}$  index, due to its high values compared to the above-mentioned indices.

From the group of multi-elemental pollution indices, it is worth using the  $PIN$  index (Fig. 4), which is based on the pollution classes of the  $PI$  index, and also suggested by Kowalska et al. [20]. The second comprehensive pollution index is the  $PLI$  index, which is the geometric mean of the  $PI$  index and was used by Wróbel et al. [25] and Rybak et al. [16].

In our opinion, it is appropriate to use the  $PI_{\text{Nem}}$  index, which considers the average and maximum values of the  $PI$  index in the equation and also it has well-defined criteria for assessing pollution [26]. Moreover, the  $MERMQ$  index, assessing the probability of toxicity, can be independently applied.

#### 4. CONCLUSIONS

The research showed that road dust samples collected at the intersections and roundabouts of main roads in the city of Wrocław and the Wrocław county in Poland are a good indicator of heavy metal emissions from road traffic. As expected, the increased metals content in road dust is related to traffic intensity. This parameter affects the lack of spatial homogeneity of heavy metals in road dust. The strong correlations between analysed metal pairs, and also dendrograms confirm a common source of chemical elements from road transport emissions. The levels of tested heavy metals in road dust do not exceed the permissible values of these elements for soils in the road area, according to the Polish Regulation. However, the highest enrichment of road dust with metals is observed at the SOW in the centre of Wrocław and at the AOW highway bypass in the Wrocław county in relation to the UCC geochemical background. The exceedances of the UCC background are the highest for copper, zinc and cadmium, enriching the road dust with metals at least a moderate degree. Unfortunately, it might lead to the occurrence of a potentially high ecological risk. The amount of the remaining trace elements fluctuates at the level of the geochemical background.

Our research confirms that different indices can similarly assess the level of samples' contamination. Therefore, it is not necessary to use all discussed pollution indices during the assessment of the environmental component, such as road dust. It is worth choosing indices with clearly defined criteria for assessing the degree of pollution. The integrated  $PLI$  index response correlates with the  $I_{\text{geo}}$  index response in an assessment of the pollution degree of tested road dust, especially when a single heavy metal is the most polluting component of the sample. It is also noteworthy that the  $PI_{\text{Nem}}$  index assesses the degree of pollution of the analysed road dust in Wrocław similarly to the  $PI$  index. Additionally, the ecological risk factor ( $E_r$ ) is consistent in response with the  $PI$  index for cadmium, mainly due to the high toxic-response factor ( $T_r$ ) for this element. The high values of the  $PI$  index for copper were observed in some locations, increasing the values of the ecological risk factor to a moderate level.

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