



Article Spatiotemporal Assessment of Soil and Vegetation Pollution with Toxic Metals from Road Traffic along the First Romanian Highway

Luoana Florentina Pascu[†], Bogdan Adrian Stănescu[†], Marian Gabriel Gâsnac and Lidia Kim *D

National Research and Development Institute for Industrial Ecology—ECOIND, Drumul Podu Dambovitei Street, No. 57-73, District 6, 060652 Bucharest, Romania; ecoind@incdecoind.ro (L.F.P.);

bogdan.stanescu@incdecoind.ro (B.A.S.); gasnac.marian@gmail.com (M.G.G.)

Correspondence: lidia.kim@incdecoind.ro; Tel.: +40-723-529-206

⁺ These authors contributed equally to this work.

Abstract: The present study is dedicated to the assessment of soil and vegetation pollution with toxic metals on a section of the oldest highway in Romania, which has been in operation for more 50 years. In the assessment, the limits of the national legislation were complied with, and the analysis was related to locally identified reference levels at a distance from the road, the main potential source of pollution. The average concentrations of toxic metals in the investigated soils had wide amplitudes, and the variations were quite large. The recorded values of the concentrations were at high or moderate levels compared to those in previous research. The contamination factor, geo-accumulation index and other calculated indices indicate the presence of pollution at a moderate level. The concentrations of toxic metals in the range 0.15–0.42 mg/kg d.m. for Cd; 2.00–6.04 mg/kg d.m. for arsenic, 16.20–76.27 mg/kg d.m. for Cu, 17.40–28.40 mg/kg d.m. for Ni, and 149.00–535.00 mg/kg d.m. for Zn exceeded the reference levels in the soil in at least one studied area. The concentrations of Zn at two observation points exceeded the alert threshold, indicating the presence of pollution caused by road traffic. Among the factors that can influence pollution, the following can be distinguished: the specifics of the areas and the activities carried out, the increase in cars transiting the highway, and the location in relation to the entrance or exit areas of the highway.

Keywords: road traffic; toxic metals (As, Cd, Pb, Cu, Ni, and Zn); soil and vegetation; environmental pollution assessment; ecological and health risk

1. Introduction

Highways and frequently traveled roads are major contributors to the release of emerging pollutants, which contaminate the air, soil, and biota. In some cases, these pollutants can also affect surface and underground water. Among the pollutants specific to road traffic, toxic metals pose a significant risk to both the environment and human health. Toxic metals are not biodegradable and reach the food chain relatively quickly due to their mobility and potential for accumulation in plants and vegetation [1-5]. The most well-known toxic metals from road traffic are Pb, Cu, Ni, Cd, Cr, and Zn. Several studies have discussed the sources of these metals and the links among them. It has been established that brake and tire wear is a primary cause of toxic metal contamination in areas adjacent to road traffic. Cu, Zn, Cd, and Pb are the toxic metals closely associated with this source [6,7]. Other studies have identified potential contamination sources, including the paint used in coatings (containing Cr and Ni), as well as the corrosion of metal parts at high temperatures, which can release toxic metals such as Pb, Cd, Cu, and Zn [8,9]. Additional identified sources include residues resulting from incomplete fuel combustion, oil leaks, and fuel additives [1]. In addition to the metals mentioned, the presence of Hg, As, and some platinum metals in areas with heavy road traffic has been monitored in recent



Citation: Pascu, L.F.; Stănescu, B.A.; Gâsnac, M.G.; Kim, L. Spatiotemporal Assessment of Soil and Vegetation Pollution with Toxic Metals from Road Traffic along the First Romanian Highway. *Environments* **2024**, *11*, 21. https://doi.org/10.3390/ environments11010021

Academic Editors: Gianniantonio Petruzzelli, Meri Barbafieri and Marco Vocciante

Received: 8 December 2023 Revised: 10 January 2024 Accepted: 17 January 2024 Published: 20 January 2024



Copyright: © 2024 by the authors. Licensee MDPI, Basel, Switzerland. This article is an open access article distributed under the terms and conditions of the Creative Commons Attribution (CC BY) license (https:// creativecommons.org/licenses/by/ 4.0/). years [2,10–13]. Arsenic and mercury are metals that are highly toxic to the environment and human health. Arsenic is a very mobile toxic metal, and its bioavailability is high in sediments and contaminated soils [8,14,15]. Nonetheless, there can be a considerable increase in the concentration of toxic metals in commercial or industrial areas adjacent to road traffic [16]. These sources contribute to increases in concentrations, suggesting an anthropogenic influence [17,18]. The adsorption of toxic metals on microplastics in urban roadside soils was also studied.

The results showed that the highest concentrations of toxic metals, including As, Pb, Ni, Cr, Cu, and Cd, were found to be adsorbed on PET, PP, PE (Cd and Ni), and PVC (Cr and Cu) [19]. Specific indices have been proposed in the literature to evaluate the level of pollution caused by toxic metals, determine the degree of contamination, and estimate the ecological and health risks associated with these. These indices provide information about the extent of toxic metal contamination, sources of pollution, and the estimation of carcinogenic and non-carcinogenic risks to human health [3,11,12,18,20-22]. Metal's bioavailability in the soil constantly changes and depends on specific chemical, biological, and environmental factors. In soils that are contaminated with toxic metals, the absorption of these can hinder plant growth. Some plant species can accumulate high levels of toxic metals, potentially threatening animals and humans [23,24]. Toxic metals can reach plants through bioaccumulation from polluted soil or deposition on plant leaves from dust particles [15,25,26]. According to Regulation 2011/574/EU, Annex I, regarding unwanted substances in feed products, the maximum allowed concentrations for toxic metals in feed products are 2 mg/kg of As, 1 mg/kg of Cd, and 30 mg/kg of Pb [27]. Considering these aspects, estimating the level of pollutants in plants and vegetation is necessary. If the estimation was based solely on the total trace element content in the soil, it would not be easy to establish a clear relationship between the concentration of toxic metals in the plant and that in the soil because not all forms of toxic metal are available to plants. Therefore, the partition coefficient/transfer factor and the biological accumulation coefficient can be used to accurately determine the amount of toxic metal transfer from soil to plants [15,28]. The A1 highway under study is part of Pan-European Transport Corridor IV, consisting of five functional sections. The first highway section that is the subject of this study is 109.6 km long and connects Bucharest to Pitesti, the seat of Arges County. This section was the first highway in Romania, built between 1967 and 1972. In 1997-1999, the highway was rebuilt and modernized in a proportion of over 80%. The old foundation was excavated, and a new foundation with wider lanes was made; the concrete road surface was abandoned, and the entire highway was asphalted. This highway was chosen for this study, on the one hand, due to its age and the changes that have taken place during its maintenance, and on the other hand, because the highway is intended for car traffic only, with strict traffic and safety restrictions imposed. These factors can provide additional information for evaluating the soil and vegetation environmental components. In addition, the advantage of the highway as an investigation area is its location in an open area that allows the taking of witness samples to compare the results, the necessary distance being at least 50 m from the road safety zone [29]. From the statistical data investigated, in 2015, an average of 15,387 cars/day traveled on the highway [30]. In 2022, the number of cars traveling on the highway increased to 23,240 cars/day [31].

The current study focused on assessing the contamination of soil and vegetation with toxic metals (As, Cd, Pb, Cu, Ni, and Zn) along the A1 highway. The main objectives of the study were the following: (1) to evaluate the contamination of the soil in the vicinity of the highway by referring to the legislation in force and based on the pollution indices; (2) to identify the dependency between toxic metals from soil and vegetation through an assay of the concentration of toxic metals from the soil with the distance from the road embankment; (3) to determine the variation in the concentration of toxic metals over time, taking into account the increase in the number of cars transiting the highway; and (4) to identify potential sources that can influence the concentration of toxic metals in the investigated area.

2. Materials and Methods

2.1. Location of Sampling Points

In total, 9 observation points (OP) were established on the section of the A1 Bucharest– Pitesti highway in Romania in order to evaluate the soil and vegetation quality. The points for taking soil samples were marked with S, and those for vegetation were marked with V. In choosing the areas, we took into account the assurance of representativeness, and chose key areas, such as the entry and exit points on the highway where road traffic is intense, and areas provided for parking, where there are service spaces in the category of fuel distribution stations. Also, specially designed areas where the highway is crossed by other roads, industrial, or commercial areas in the proximity of the highway, and areas without any development (free zones) were taken into account.

For each observation point, soil samples were taken at distances of 2 m and 5 m from the freeway shoulder (from the safety zone), and then at a distance of 60 m from the safety zone as reference samples. The outline of an observation point regarding the distribution of the sampling points can be seen in Figure 1.



Figure 1. Distribution of soil and vegetation sampling points on the A1 highway; the traffic direction from Bucharest to Pitesti (source of map: Google Earth Pro, 2023, modified).

At the European level, the average background concentrations of toxic metals were identified in areas farthest from the road embankment and at least 30 m away. The distance at which contaminants spread varies depending on vehicle speed, wind speed, rainwater runoff, and highway maintenance. The highest concentrations of toxic metals were found within 5 m of the road embankment. Though high concentrations of toxic metals were also found at distances of 25 and even 150 m, the concentrations of toxic metals generally dropped below the background level after a distance of 50 m [1]. All the samples were taken from the topographic surface at depths from 0 to 10 cm.

Herbaceous vegetation (grass) samples, contaminated with dust, were taken at distances of 2 m and 60 m (reference level). Several sampling campaigns were carried out in the spring and summer of 2017, and, subsequently, in the summer of 2022. For the sampling of the soil, manually operated pedological sampling equipment was used, as was the soil sampling kit (Hand Auger Set) by Royal Eijkelkamp B.V., Gelderland Province, The The locations of the observation points for taking soil and vegetation samples are presented in Figures 2 and 3, and the geographic coordinates are in Table 1.



Figure 2. Location of soil and vegetation sampling points along the A1 highway; direction of traffic from Bucharest to Pitesti (source of map: Google Earth Pro, 2023, modified).



Figure 3. Location of soil and vegetation sampling points along the A1 highway; direction of traffic from Pitesti to Bucharest (source of map: Google Earth Pro, 2023, modified).

Observation Points (OP)	GPS Coordinates with Garmin, Model Montana 610 Lat/Long WGS'84	Details of Sampling Locations	Photos (Source of Images: Google Street View)
	A1 Hig	hway, Bucharest–Pitesti Direction	
OP 1	44°26.595′ N 25°55.109′ E	Sampling point located at km 15 on the highway (after A1 entrance area)	
OP 2	44°31.932′ N 25°31.984′ E	Sampling point located at km 49 on the highway (in the vicinity of the fuel distribution station)	
OP 3	44°38.869′ N 25°22.789′ E	Sampling point located at km 67 on the highway (near a paved road, DC77, which crosses the highway)	
OP 4	44°42.676′ N 25°8.986′ E	Sampling point located at km 87 on the highway, direction Bucharest–Pitesti, a free zone)	
OP 5	44°47.874′ N 24°58.669′ E	Sampling point located at km 104 on the highway in the vicinity of an industrial area	
	A1 Hig	hway, Pitesti-Bucharest Direction	
OP 6	44°45.259′ N 25°5.137′ E	Sampling point located at km 94 on the highway, an area with logistics warehouses	

Table 1. GPS coordinates and description of sampling points.

Observation Points (OP)	GPS Coordinates with Garmin, Model Montana 610 Lat/Long WGS'84	Details of Sampling Locations	Photos (Source of Images: Google Street View)
OP7	44°42.000′ N 25°13.685′ E	Sampling point located at km 80 on the highway, with service areas (fuel distribution station)	
OP8	44°28.602′ N 25°41.179′ E	Sampling point located at km 35 on the highway (free zone after crossing the Arges river)	
OP9	44°26.217′ N 25°57.470′ E	Sampling point located at km 10 on the highway (exit from the highway), a commercial area	

Table 1. Cont.

2.2. Preparation of Soil and Vegetation Samples

To determine the total content of toxic metals, the soil samples were air-dried, grounded, sieved, and homogenized, retaining a fraction of <150 µm for analysis because its homogeneity is much better and allows the standard deviation of the repeatability to decrease [32]. From each soil sample, 0.5 g was weighed and mineralized with a mixture of HCl and HNO₃ at a ratio of 3:1 in a closed system by microwave digestion. The reagents used in the experiments were of a grade of ultrapure quality, purchased from Sigma-Aldrich, Taufkirchen, Germany. After digestion was completed, the solution was filtered and washed with distilled water, and the obtained filtrate was collected in a 50 mL volumetric flask (SIST ISO 11466/99—soil quality—extraction of microelements soluble in aqua regia) [33]. The distilled water was produced with Ultrapure Water System Millipore Milli-Q, Merk Millipore, Germany.

The vegetation samples were spread out and left at room temperature to dry (covered with filter paper) for 2 days. The samples were finely ground and weighed ($0.5 \text{ g} \div 1 \text{ g}$). Then, 10 mL of HNO₃ and 2 mL of H₂O₂ (reagents of ultrapure quality) were added. The glasses covered with a watch glass were left for 16 h to destroy the organic matter (cold mineralization). The next stage consisted of hot mineralization; the samples were digested on an electric stove until the remaining liquid became clear. The samples were filtered and put into a 50 mL volumetric flask. Then, the residue was washed with ultrapure water, and the resulting water was collected in the volumetric flask.

The toxic metal contents of the soil and vegetation samples were determined via inductively coupled plasma mass spectrometry on ICP-MS equipment (type Agilent 7900, Agilent Technologies, Santa Clara, CA, USA). The toxic metals analyzed in the soil and vegetation samples were Cd, Cu, Ni, Pb, Zn, and As. For the analysis of toxic metals from the two types of samples, calibration curves were drawn in the range of

 $10 \ \mu g/L-50 \ \mu g/L$. To verify the results, a certified reference material, SRM 2710—Montana Soil (Sigma Aldrich, Taufkirchen, Germany)—traceable to NIST, was used.

2.3. Assessment of Soil and Vegetation Pollution and Ecological Risk

In order to evaluate the pollution of the soil and vegetation with toxic metals in the areas adjacent to the A1 highway, a series of specific evaluation indices were used. In addition to these indices, the results obtained were reported to the reference values for traces of chemical elements in the soil from Romanian Order 756/1997 (approval of the regulation regarding the assessment of environmental pollution) for sensitive land use types (Table 2) [34].

Table 2. Reference values for toxic metals in soil (mg/kg d.m.)—types of sensitive uses.

Toxic Metals	As	Cd	Pb	Cu	Ni	Zn
Normal values	5	1	20	20	20	100
Alert thresholds	15	3	50	100	75	300
Intervention thresholds	25	5	100	200	150	600

To evaluate the degree of pollution with toxic metals, the following were determined: the contamination factor (CF_m) and the degree of contamination (CD); the geo-accumulation index (*Igeo*); the pollution load index (*PLI*); the risk factor (*RF*), and the ecological potential risk index (*PER*).

The evaluation of contamination with toxic metals in the soil was carried out by determining the contamination factor (CF_m) of each toxic metal in the samples and the degree of contamination (CD) for all six toxic metals in the samples, in accordance with the formulas given by Hakanson [22]:

$$CF_m = \frac{C_m s}{C_m b} \tag{1}$$

$$CD = \sum CF_m \tag{2}$$

where $C_m s$ is the metal concentration in the contaminated soil; $C_m b$ is the metal concentration in the reference soil (uncontaminated). In the literature, the background concentrations of metals in the Earth's crust determined by Taylor and McLennan were considered for the reference soil [35]. In our study, considering that we wanted to identify the contamination specific to road traffic, the metal concentrations in the soil 60 m away from the road safety zone were considered reference values.

Muller introduced the bioaccumulation index (*Igeo*) for assessing toxic metal contamination in surface aquatic sediments [36]. Later, the *Igeo* was also used to evaluate toxic metal contamination in soils and roadside dust from road traffic [3,8,18,20].

$$Igeo = log_2 \left[\left(\frac{C_m s}{K \times C_m b} \right) \right]$$
(3)

where *K* is the correction factor that takes into account the variations in metal traces in the natural background as a result of lithogenic effects (K = 1.5) [35].

In accordance with the method introduced by Tomlinson et al., using the pollution load index (*PLI*), the pollution levels and the distribution of pollution in different sites are compared [37].

$$PLI = \sqrt[n]{(CF_m)_1 \times (CF_m)_2 \times (CF_m)_3 \times \dots \times (CF_m)_n}$$
(4)

$$(PLI)_{zone} = \sqrt[m]{(PLI)_1 \times (PLI)_2 \times (PLI)_3 \times \dots \times (PLI)_m}$$
(5)

where CF_m is the contamination factor; *n* is the number of metals analyzed; *m* is the number of observation points.

The risk factor (*RF*) and the potential ecological risk index (*PER*) was introduced by Hakanson [22] and is calculated in accordance with the following formulas:

$$RF = CF_m \times Tf \tag{6}$$

$$PER = \sum RF \tag{7}$$

where RF is the risk factor for each metal; Tf is the toxic response factor for each metal; PER is the potential ecological risk index for all metals from each observation point.

The toxic metal accumulation from the soil in the vegetation was calculated using a single index (the bioaccumulation index—*BAI*) [38]:

$$BAI = \frac{CM_v}{CM_s} \tag{8}$$

where CM_v is the metal concentration in the vegetation, and CM_s is the metal concentration in the soil.

In the Table 3, the limits and levels of pollution for *CF_m*, *CD*, *Igeo*, *PLI*, *RF*, *Tf*, and *PER* are given.

Table 3. The indices of the limits and levels of pollution.

Index	Value	Category							
Contamination factor (CF)	CF < 1 $1 \le CF < 3$ $3 \le CF < 6$ CF > 6	Low contamination Moderate contamina Considerable contam Very high contamina	Low contamination Moderate contamination Considerable contamination Very high contamination						
Contamination degree (CD)	CD < 6 $6 \le CF < 12$ $12 \le CF < 24$ CF > 24	Low contamination of Moderate contamina Considerable contam Very high contamina	legree tion degree iination degree tion degree						
Index	Value	Category							
Pollution load index (PLI)	PLI < 1 PLI = 1 PLI > 1	Unpolluted level Baseline level of pollution The presence of pollution							
Index	Value	Class	Pollution level						
Geoaccumulation Index (Igeo)	≤ 0 0-1 1-2 2-3 3-4 4-5 >5	0 1 2 3 4 5 6	Unpolluted lev Unpolluted lev Moderate pollu Moderate pollu High pollution High pollution Very high pollu	rel rel to moderate po ation level ation level to high level level to very high ation level	ollution level n pollution level h pollution level				
Index	RF	PER	Ecological risk						
Risk factor (<i>RF</i>) and Potential ecological risk (<i>PER</i>)	$\begin{array}{l} RF < 40 \\ 40 \leq RF < 80 \\ 80 \leq RF < 160 \\ 160 \leq RF < 320 \\ RF \geq 320 \end{array}$	$\begin{array}{c} PER < 150 \\ 150 \leq PER < 300 \\ 300 \leq PER < 600 \\ \hline \\ PER \geq 600 \end{array}$	Low ecological Moderate ecologica High ecologica Very high ecologica Extremely high	risk ogical risk I risk ogical risk 1 ecological risk					
Toxic metal	As	Cd	Pb	Cu	Ni	Zn			
Toxic response factor (<i>Tf</i>)	20	30	5	5	6	1			

3. Results

The concentrations of toxic metals analyzed in the soil samples (for the three sampling campaigns and their median) are presented in Table 4 (expressed in mg/kg). For the statistical interpretation of the results, the median and the absolute deviation from the median (MAD) were used, these being more robust against extreme values compared to the arithmetic mean and the standard deviation [39].

Observation Points	Sampling Points		As	Cd	Pb	Cu	Ni	Zn
OP1	S1 (2 m)	Min.	5.32	0.14	20.70	19.40	12.92	239.00
		Max.	5.74	0.19	41.63	46.79	28.40	246.00
		Median	5.74	0.19	41.63	46.79	28.40	246.00
		MAD	0.42	0.05	20.93	22.69	2.88	7.00
	S1 (RL)	CV (%) Median	7.32	0.30	50.28 12 71	48.49	10.14	2.85 146.00
<u>OP</u> 2	62 (2 m)	Min	2.25	0.06	12.0	17.20	20.64	155.00
OF2	52 (2 III)	Max	5.55 7.15	0.08	17.00	47.30	20.64	444.00
		Median	6.04	0.14	17.59	28.25	25.40	182.00
		MAD	1.11	0.08	0.34	10.95	1.10	27.00
		CV (%)	18.38	55.00	1.93	38.76	4.33	14.84
	S2 (RL)	Median	4.73	0.16	15.81	26.16	22.30	83.15
OP3	S3 (2 m)	Min.	2.20	0.04	34.10	20.50	21.80	91.90
		Max.	5.51	0.79	88.92	32.38	32.18	177.00
		Median	3.47	0.27	44.90	26.78	25.66	147.00
		CV(%)	36.62	0.23 84 87	24.05	20.91	15.00	20.41
	S3 (RL)	Median	1.68	0.25	14.99	9.18	20.07	80.31
OP4	S4 (2 m)	Min	1.80	0.06	18.90	12.61	13.17	387.00
011	01(211)	Max.	4.46	0.53	33.84	42.14	29.09	665.00
		Median	3.68	0.20	21.92	19.20	22.80	535.00
		MAD	0.78	0.14	3.02	6.59	6.29	130.00
		CV (%)	21.20	68.00	13.78	34.32	27.59	24.30
	S4 (RL)	Median	1.58	0.14	11.47	15.74	20.34	117.00
OP5	S5 (2 m)	Min.	1.82	0.07	14.90	12.60	14.47	87.80
		Max.	6.32	0.61	57.65	20.19	20.72	164.00
		Median	4.73	0.42	20.83	16.20	17.40	149.00
		CV(%)	33.62	45 24	28.47	22.00	2.95	10.00
	S5 (RL)	Median	1.84	0.16	5.12	5.64	22.26	78.40
OP6	S6 (2 m)	Min.	1.46	0.05	28.19	42.00	15.91	276.00
		Max.	4.33	0.89	47.95	88.18	24.00	453.00
		Median	3.94	0.66	31.50	76.27	18.10	347.00
		MAD	0.39	0.23	3.31	11.91	2.19	71.00
		CV (%)	9.87	34.85	10.51	15.62	12.10	20.46
	S6 (RL)	Median	0.99	0.18	8.90	56.20	14.77	89.50
OP7	S7 (2 m)	Min.	2.28	0.10	17.60	22.80	21.29	105.00
		Max. Modian	8.50 5.78	0.66	77.65	/1.20	45.81	231.00
		MAD	2 72	0.17	20.45	18.88	4 41	230.00
		CV (%)	47.06	62.59	53.75	45.30	17.16	0.43
	S7 (RL)	Median	3.94	0.20	22.83	23.77	17.82	104.00
OP8	S8 (2 m)	Min.	1.51	0.11	26.80	25.74	16.07	113.00
	· · /	Max.	5.40	0.74	50.05	36.80	23.78	207.00
		Median	3.94	0.18	44.90	30.03	20.20	186.00
		MAD	1.46	0.07	5.15	4.29	3.58	21.00
	S8 (RL)	CV (%) Median	37.08 1.99	0.13	11.47 31.13	14.29	17.72	11.29 67.60
000	SQ (2 m)	Min	1.76	0.10	23.10	26.02	17.80	110.00
017	59 (Z III)	Max	5 25	0.10	42.12	20.21 96.22	37.61	246.00
		Median	2.00	0.15	34.10	31.50	22.30	192.00
		MAD	0.24	0.05	8.18	5.29	4.41	54.00
		CV (%)	12.00	33.33	23.99	16.79	19.78	28.13
	S9 (RL)	Median	2.44	0.16	21.00	20.12	17.61	69.30

Table 4. Toxic m	etal concentrat	tions in soil sa	amples studied.
Tuble II Tokie III	ietui concentiuu	ciono ni bon be	ampies studied.

Min.: minimum value (mg/kg); Max.: maximum value (mg/kg); Median (mg/kg) MAD; median absolute deviation, CV (%): coefficient of variation; RL: reference level.

As can be seen in Table 4, at a distance of 2 m from the road embankment, cadmium was found in the soil samples in low concentrations that varied between 0.14 mg/kg (S2) and 0.66 mg/kg (S6) (as its median values). The obtained values were in the same order as that of the Cd concentrations in the reference levels (RL), which indicates the presence of Cd in the area in the natural background of the soil. In the literature, the concentration of Cd studied in areas with road traffic varies between the limit of the method and approximately

Legend:

2 mg/kg [3,20,21]. The median arsenic concentration at 2 m varied between 2.00 mg/kg in S9 and 6.04 mg/kg in S2. At sampling points S3, S5, and S6, the arsenic concentrations were at least two times higher than the reference values, indicating contamination due to road traffic. The median concentrations of Pb, Cu, and Ni varied at a distance of 2 m between 17.59 mg/kg (S2) and 44.90 mg/kg (S3 and S8) for Pb, 16.20 mg/kg (S5) and 76.27 mg/kg (S6) for Cu, and 17.40 mg/kg (S5) and 28.40 mg/kg (S1) for Ni. At all the sampling points, the concentrations of Pb and Cu were higher than the reference levels, which indicates contamination induced by road traffic. In the case of nickel, the median concentrations analyzed in the soil samples were close to those of the reference levels. The highest concentrations were obtained for zinc, varying between 147 mg/kg (S3) and 535 mg/kg (S4). Regarding the reference levels, the concentrations of Zn in the analyzed samples were much higher at sampling points S4 and S6, exceeding the reference levels fourfold, and indicating significant soil contamination with Zn due to road traffic.

Table 5 shows the median concentrations of the toxic metals analyzed in the soil samples (at a 2 m distance from the road embankment) relative to the reference values established by Order 1997/756.

Table 5. Concentrations of toxic metals relative to reference values of Order 756/1997
--

	S 1	S2	S 3	S 4	S 5	S 6	S 7	S 8	S 9	NV	AT	IT
As (2 m)	5.74	6.04	3.47	3.68	4.73	3.94	5.78	3.94	2.00	5.00	15.00	25.00
As (RL)	3.41	4.73	1.68	1.58	1.84	0.99	3.94	1.99	2.44	5.00	15.00	25.00
Cd (2 m)	0.19	0.14	0.27	0.20	0.42	0.66	0.27	0.18	0.15	1.00	3.00	5.00
Cd (RL)	0.30	0.16	0.25	0.14	0.16	0.18	0.20	0.13	0.16	1.00	3.00	5.00
Pb (2 m)	41.63	17.59	44.90	21.92	20.83	31.50	38.05	44.90	34.10	20.00	50.00	100.00
Pb (RL)	12.71	15.81	14.99	11.47	5.12	8.90	22.83	31.13	21.00	20.00	50.00	100.00
Cu (2 m)	46.79	28.25	26.78	19.20	16.20	76.27	41.68	30.03	31.50	20.00	100.00	200.00
Cu (RL)	10.93	26.16	9.18	15.74	5.64	56.20	23.77	16.09	20.12	20.00	100.00	200.00
Ni (2 m)	28.40	25.40	25.66	22.80	17.40	18.10	25.70	20.20	22.30	20.00	75.00	150.00
Ni (RL)	18.99	22.30	20.07	20.34	22.26	14.77	17.82	14.23	17.61	20.00	75.00	150.00
Zn (2 m)	246.00	182.00	147.00	535.00	149.00	347.00	230.00	186.00	19 2 .00	100.00	300.00	600.00
Zn (RL)	146.00	83.15	80.31	117.00	78.40	89.50	104.00	67.60	69.30	100.00	300.00	600.00

The pollutant concentration value corresponds to the normal value of reference (NV)

The pollutant concentration value corresponds to the value of the alert threshold (AT)

The pollutant concentration value corresponds to the value of the intervention thresholds (IT)

By reporting the concentrations of toxic metals to the reference values established in accordance with Order 1997/756 for soils with sensitive use (Figure 4), it can be seen that the concentrations of As and Cd were within normal values for all the analyzed samples, except for As at points S1, S2, and S7, where the concentrations exceeded the reference values. The concentrations of Pb in most samples exceeded the normal reference values. The Cu and Ni concentrations exceeded the normal reference values at all sampling points, but less so in S4 and S5 for Cu and in S5 and S6 for Ni. The concentrations of Zn in all samples exceeded the normal reference values, and in S4 and S6, the concentrations exceeded the alert threshold. In the following, an analysis of the maximum values of the lead content in the soil was carried out for all the observation points (Figure 4), and for comparison, the average values for the lead content in the soil at the reference points located at approximately 60 m from the highway were recorded (Figure 5).



Figure 4. Maximum values of lead in all soil samples.



Figure 5. Reference values (medians) of lead at all observation points.

It was observed that the maximum value of the lead content in the soil was recorded at OP 3, located on along the A1 highway in the Bucharest to Pitesti direction. The elevated content of lead in the soil is explained by the long period of operation of this highway, especially long periods of time in which leaded gasoline was used. The normal value of the national legislation was exceeded, namely with the value of 20 mg/kg d.m. in eight of the nine observation points, which reinforces that the possible source of lead in the soil was emissions from motor vehicles as a result of the use of leaded gasoline.

It was observed that the median values in the reference areas mostly fell within the normal limits for lead content in the soil. At only three of the nine observation points, the value of 20 mg/kg d.m. was slightly exceeded.

Similarly, representations were made for zinc; the maximum values are shown in Figure 6, and the reference values are shown in Figure 7.



Figure 6. Maximum values of zinc in all soil samples.



Figure 7. Reference values (median) of zinc in all observation points.

There are large amplitudes between the maximum recorded values and the reference values 60 m away. It is important to note that the normal values for the zinc content in the soil at all observation points exceeded the value of 100 mg/kg d.m.

It was observed that for the vast majority of reference points, the median values were within the limit of the normal values. Only in three out of nine observation points were the normal values exceeded, without any other exceeding the alert threshold. The large differences in concentrations from the points in the vicinity of the A1 and up to only 60 m away from it suggest that the source of the recorded zinc concentrations is the road traffic on the highway and the long duration of circulation on it.

Sampling Points	As (n	ng/kg)	Cd (n	ng/kg)	Pb (m	g/kg)	Cu (m	g/kg)	Ni (m	ıg/kg)	Zn (n	ng/kg)
	2 m	(RL)	2 m	(RL)	2 m	(RL)	2 m	(RL)	2 m	(RL)	2 m	(RL)
V1	0.95	0.53	0.21	0.25	4.13	3.66	7.56	4.11	12.37	10.70	47.86	31.16
V2	0.61	0.75	0.11	0.15	3.84	3.31	6.41	4.78	6.52	7.74	44.03	24.72
V3	0.69	0.50	0.05	0.19	3.48	3.33	7.05	5.34	5.30	10.78	45.69	32.65
V4	0.43	0.73	0.24	0.16	3.57	4.94	7.81	6.02	9.01	9.35	40.72	30.03
V5	0.51	0.59	0.31	0.25	3.31	3.41	7.33	3.76	7.69	9.79	39.94	16.33
V6	0.74	0.34	0.18	0.09	2.77	2.52	7.61	4.12	8.41	6.83	45.37	32.04
V7	0.78	0.85	0.06	0.11	3.22	3.06	5.11	2.95	5.74	5.11	38.81	18.15
V8	1.03	0.88	0.18	0.15	4.78	3.66	12.08	4.19	6.83	7.71	89.00	19.58
V9	0.52	0.77	0.19	0.08	4.66	3.80	8.30	5.15	6.78	7.34	73.98	28.73

Table 6. Toxic metal concentrations in vegetation samples studied.

the road embankment.

Table 6 shows that the arsenic content in the vegetation samples was in the range of 0.43–1.03 mg/kg, and the cadmium content was between 0.05 and 0.31 mg/kg. The lead content varied between 2.77 and 4.78 mg/kg, and the concentrations of Cu and Ni were found in the same range, with 5.11–12.08 mg/kg for Cu and 5.30–12.37 mg/kg for Ni. Similar to the soil samples, in the vegetation samples, the Zn content was the highest, varying between 38.81 and 89.00 mg/kg. It was observed that there were no significant differences between the concentrations in the samples and the reference levels, except for copper in sample V8 and Zn in samples V5, V7, V8, and V9. Table 7 shows the results obtained for the bioaccumulation index (BAI) of toxic metals in the soil and vegetation (samples taken at a distance of 2 m from the road embankment).

Table 6 shows the results obtained for the concentration of toxic metals (median for the spring/summer 2017 sampling campaigns) in the vegetation samples 2 m away from

Vegetation/Soi	l As	Cd	Pb	Cu	Ni	Zn
V1/S1	0.16	1.10	0.10	0.16	0.44	0.19
V2/S2	0.10	0.75	0.22	0.23	0.26	0.24
V3/S3	0.20	0.20	0.08	0.26	0.21	0.31
V4/S4	0.12	1.20	0.16	0.41	0.40	0.08
V5/S5	0.11	0.73	0.16	0.45	0.44	0.27
V6/S6	0.19	0.27	0.09	0.10	0.46	0.13
V7/S7	0.13	0.24	0.08	0.12	0.22	0.17
V8/S8	0.26	1.00	0.11	0.40	0.34	0.48
V9/S9	0.26	1.27	0.14	0.26	0.30	0.39

Table 7. BAI values for toxic metals in the investigated area.

The highest values for the BAI (Table 7) were obtained for Cd in most of the sampling points, which indicates a dependence between the soil and vegetation. For the other toxic metals, the BAI values did not vary significantly, suggesting that the concentrations in plants are not a dependent function of the concentrations in the soil. The concentrations of toxic metals in the soil samples taken 2 m from the road embankment were monitored over time. Figure 8 shows their evolution over three campaigns during spring/summer 2017 and after five years in summer 2022.

9.00 8.00 concentration (mg/Kg) 7.00 6.00 5.00 4.00 3.00 2.00 As 1.00 0.00 562 55/2 Sla 50 Sampling point Summer 2017 Spring 2017 Summer 2022





(e) Ni





(**d**) Cu



Figure 8. Monitoring the temporal evolution of toxic metal concentrations (2017–2022).

By monitoring the soil over time at the sampling points 2 m and 60 m (RL) from the road embankment (Figure 8), it can be seen that the lowest concentrations were obtained in the sampling campaign of spring 2017 in most of the analyzed samples. This indicates that in the spring, the concentrations of toxic metals on the soil surface were lower due to the soil surface being washed with water from the snow accumulated during the winter. The toxic metal concentrations varied in the other two sampling campaigns (summer 2017 and 2022). In the summer 2022 campaign, a decrease in the Cd and Pb concentrations and a slight increase in the As, Cu, and Zn concentrations were observed. This increase could be associated with the increase in cars transiting the highway. Figure 9 shows the concentrations of toxic metals obtained in the soil samples taken at a distance of 2 m, 5 m, and 60 m from the road embankment.

For most of the sampling points, it was observed that the concentrations of toxic metals decreased with distance from the road embankment (Figure 9). For lead, at sampling points S1, S3, S5, S6, and S7, a significant decrease in the concentrations at 2 m was



observed compared to the concentrations at 5 m and 60 m. For Cu, Pb, and Cd, the highest concentrations at a distance of 2 m were obtained at points S1, S3, and S9.

Figure 9. Monitoring the evolution of toxic metal concentrations with distance.

Figure 10 shows the values obtained for the *CF* for the six toxic metals studied in the soil and vegetation at each sampling point and the degree of contamination for each point.



Figure 10. Contamination factor and contamination degree of soil and vegetation. (**a**) Contamination factor and contamination degree of soil. (**b**) Contamination factor and contamination degree of vegetation.

Referring to the contamination factor (Figure 10a), it can be seen that for most of the analyzed toxic metals, the contamination was moderate ($1 \le CF \le 3$). At sampling points S4 and S6, there was considerable contamination with zinc ($3 \le CF \le 6$). The degree of contamination was in the interval of 7.67–17.65, which indicates a moderate degree of contamination at S2, S3, S7, S8 and, S9, and a considerable degree of contamination at the other studied points (Table 3). In most vegetation samples (Figure 10b), contamination with toxic metals was low for Cd and Ni and moderate for the other analyzed toxic metals, with zinc being the only one that induced considerable contamination at sampling point V8. The degree of contamination of the vegetation samples was moderate for all investigated points (Table 3).

Table 8 shows the results obtained for the *Igeo*, PLI, and PLI zones for the studied toxic metals (median concentrations) in the soil samples taken at a distance of 2 m from the road embankment.

Sampling Point –		Igeo						
	As	Cd	Pb	Cu	Ni	Zn	– PLI	PLI Zone
S1	0.17	-1.24	1.13	1.51	0.00	0.17	1.83	
S2	-0.23	-0.78	-0.43	-0.47	-0.40	0.55	1.22	
S3	0.46	-0.47	1.00	0.96	-0.23	0.29	1.89	
S4	0.63	-0.09	0.35	-0.30	-0.42	1.61	1.84	
S5	0.78	0.81	1.44	0.94	-0.94	0.34	2.21	2
S6	1.41	1.29	1.24	-0.14	-0.29	1.37	2.63	
S7	-0.03	-0.15	0.15	0.23	-0.06	0.56	1.63	
S8	0.40	-0.12	-0.06	0.32	-0.08	0.88	1.75	
S9	-0.87	-0.68	0.11	0.06	-0.24	0.89	1.38	

Table 8. Igeo values of toxic metals in the studied points—the area adjacent to the A1 highway.

Regarding the *Igeo* (Table 8), arsenic (S6), Cd (S6), Pb (S1, S3, S5, and S6), Cu (S1), and Zn (S4, and S6) induced a moderate level of pollution (class 2). Arsenic (S2, S7 and S9), Cd (for all investigated points except sampling points S5 and S6), Pb (S2, and S8), Cu (S2, S4, and S6), and Ni (for all investigated points except sampling point S1) classify the analyzed soils as class 0 (unpolluted level). At all other points, the analyzed toxic metals classify the soils as class 1 (an unpolluted level to a moderate pollution level). Both the PLI and PLI zones indicate the presence of pollution at the investigated points (PLI and PLI zones > 1).

Figure 11 presents the results obtained for the risk factor and the potential ecological risk of toxic metals in the soil.



Figure 11. Evolution of the risk factor and the potential ecological risk in soil.

The ecological risk factor indicates a moderate ecological risk at sampling points S3, S4, S5, and S6 (for arsenic) and at points S4, S5, S7, and S8 (for Cd), and a high ecological risk at point S6 (for Cd). The RF was low for all other analyzed toxic metals (Figure 11). The potential ecological risk was moderate for points S5 and S6. For all other investigated points, the *PER* was low (Table 3).

4. Discussion

This study is a contribution to elucidating how pollutants from the category of toxic metals, with high toxicity, can remain in the soil or accumulate through phytoextraction in spontaneous vegetation for long periods of time. According to the specific literature, the anthropogenic input of Pb and Zn in areas with road traffic has been confirmed at the European level. More than 82% of 64 publications related to the anthropogenic input of Pb were dedicated to road traffic. More than 62% of 48 publications were devoted to the anthropogenic supply of Zn in areas with road traffic [5]. The details presented in this evaluation, in the case of lead and zinc, confirm this research with concrete data from our studies. The large amplitude between the concentration levels recorded from the vicinity area from the highway to the reference area at approximately a 60 m distance only confirms the polluting contribution of road traffic on the highway, to which must be added the long period of its exploitation. Also, with a significant contribution, those particularities specific to the entrance and exit areas of the A1 highway can be mentioned, where both accelerations and decelerations of vehicles imply an increase in gas emissions from vehicles, and in recent years, in the case of the entry flow towards Bucharest, there were cases of congestion or traffic jams up to the exit on the ring road, extending for several kilometers. In the latter situation, frequent starts and stops and traveling at a very low speed result in additional pollutants being produced due to increasing emissions. As a rule, heavy cars are the ones that are forced to exit the A1 towards the ring road, and thus significant emissions are from these categories of vehicles. The number of two-axle trucks has decreased from 767 per day in 2015 to 526 per day in 2022. Similarly, the number of three- and four-axle trucks has declined from 308 per day in 2015 to 244 per day in 2022. On the other hand, the number of heavy trucks of the TIR type has increased from 2714 per day in 2015 to 4574 per day in 2022 [30,31]. The regular acceleration and deceleration of highway vehicles generates more toxic metal-contaminated dust from wheels and other car parts [3]. We should not neglect the synergistic contribution of particulate matter with toxic metal content from areas outside the limits of Bucharest, where real estate, logistics, and industrial developments are registered, which introduce an additional contribution of pollutants to the soil level. Also, numerous cases of uncontrolled waste storage and the incineration of some waste in the peripheral area of Bucharest have been reported, which induce additional emissions with pollution output. The assessment carried out along the Bucharest-Pitesti highway sector of the A1 highway highlights the fact that the highest concentrations of toxic metals, such as Cd, As, Cu and Zn, which exceeded the reference levels in the investigated areas, are located at OP 6. At this point, there is a built-up area with some logistic warehouses, where heavy vehicles transit for loading and unloading operations (Table 1). Practically, we can record an additional output of emissions from these heavy transport vehicles outside of those on the highway. The presence of deposits in the vicinity of the highway may, to some extent that was not evaluated in this study, determine changes in the dispersion mode of pollutants at the ground level. High concentrations of toxic metals, as expected, were recorded at the observation points at the entrances and exits of the highway, found at points OP1 and OP9, respectively, in the Bucharest-Pitesti and Pitesti-Bucharest directions. It was recorded at OP4 that there was significant contamination with Zn (535.00 mg/kg), which exceeded the reference level by four times. Since OP4 is a free area without other anthropogenic sources, this contamination can be attributed to road traffic, in the absence of other information from the history of this area. Among all toxic metals analyzed, the concentrations of zinc at OP4 (free area) and OP6 (logistic storage) exceeded the alert

threshold for sensitive-use soils. This aspect indicates the presence of pollution due to road traffic but also due to the long period of exploitation of these highways in Romania.

5. Conclusions

The activities carried out within this study aimed to fill a gap regarding the evaluation of the soil and vegetation in the areas located near the oldest highway in Romania, which has been in use for more than half a century. Over the years, numerous rehabilitations have been recorded in its various sections. The flow of vehicles has increased in the last 30 years, and in many sections, new functional areas have appeared, including areas with logistics warehouses, industrial areas, areas with fuel distribution stations, etc. It should be stated that the evaluation complied with the national legislative provisions, specifically those stipulated in Order 756/1997, regarding the evaluation of environmental pollution with subsequent modifications and additions, and the analysis of soil samples considered a reference by positioning sampling sites approximately 60 m away from the main road traffic on the highway. In this way, it was possible to better detect the contribution of road traffic as a potential source of soil pollution in the immediate vicinity of the highway. Other factors that can influence the pollution output were not neglected either; these refer to the specifics of the sampling areas, the activities carried out in those areas, the positioning of the sampling points compared to that of the entry or exit areas from the highway, and other influencing factors. Regarding the hypotheses from which the study started, we can confidently say that they have been confirmed, in the sense that there is a proven causal connection between the intense traffic on the highway, and the increased potential of pollution with toxic metals in the immediate vicinity due to exhaust gas emissions. On an old highway, as in the case of the A1 Bucharest-Pitesti highway, there are additional potential pollutants, including long periods of use of fuels with lead content in internal combustion engines, as well as periods of exploitation, with the role of the accumulation of toxic metals over long periods of time leading to residual pollution. Although a history could not be established for each observation point in the present study, all data could be correlated with the current specifics of the monitoring areas. Here, we must mention the entrance and exit areas of the highway, where there are additional polluting ports that could be well-documented. This evaluation carried out on the Bucharest–Pitesti highway section of the A1 highlights the fact that there is at least one area where toxic metal concentrations, such as those of Cd, As, Cu, Ni, and Zn, exceed the reference levels in the investigated areas.

The results obtained in this paper have applicative potential for the elucidation of aspects related to the behavior of toxic metals accumulated in soil and vegetation, when environmental assessment is necessary in areas of roads or highways that have been in use for a long time. The behavior of toxic metals was monitored over time and by comparison with reference levels of toxic metals (samples taken 60 m from the highway embankment). The approaches of the study can be extended for similar situations from different countries, on the condition that they will be correlated with other relevant information regarding the potential sources of pollution or particularities specific to the analyzed areas.

Author Contributions: Conceptualization, L.F.P., B.A.S. and L.K.; methodology, B.A.S. and L.K.; data curation, L.F.P., B.A.S. and L.K.; formal analysis, M.G.G. and L.K.; validation, B.A.S. and L.K.; investigation, B.A.S., M.G.G. and L.K.; resources, L.F.P. and L.K.; writing—original draft preparation, L.K.; writing—review and editing, B.A.S. and L.K.; visualization, L.F.P. and M.G.G.; supervision, B.A.S. All authors have read and agreed to the published version of the manuscript.

Funding: This work was carried out through Core Programs with the support of the Ministry of Research, Innovation and Digitalization, within the National Research Development and Innovation Plan 2022–2027 (contract 3N/2022, project code PN 23 22 02 02 and PN 23 22 03 02).

Data Availability Statement: All raw data can be obtained from the authors upon request.

Conflicts of Interest: The authors declare no conflicts of interest.

References

- Werkenthin, M.; Kluge, B.; Wessolek, G. Metals in European roadside soils and soil solution—A review. *Environ. Pollut.* 2014, 189, 98–110. [CrossRef] [PubMed]
- 2. An, S.; Liu, N.; Li, X.; Zeng, S.; Wang, X.; Wang, D. Understanding heavy metal accumulation in roadside soils along major roads in the Tibet Plateau. *Sci. Total Environ.* **2022**, *802*, 149865. [CrossRef] [PubMed]
- 3. Suvetha, M.; Charles, P.E.; Vinothkannan, A.; Rajaram, R.; Paray, B.A.; Ali, S. Are we at risk because of road dust? An ecological and health risk assessment of heavy metals in a rapid growing city in South India. *Environ. Adv.* 2022, 7, 100165. [CrossRef]
- Mmolawa, K.B.; Likuku, A.S.; Gaboutloeloe, G.K. Assessment of heavy metal pollution in soils along major roadside areas in Botswana. *Afr. J. Environ. Sci. Technol.* 2011, 5, 186–196. Available online: https://www.ajol.info/index.php/ajest/article/view/ 71926 (accessed on 16 January 2024).
- 5. Binner, H.; Sullivan, T.; Jansen, M.; McNamara, M. Metals in urban soils of Europe: A systematic review. *Sci. Total Environ.* 2023, 854, 158734. [CrossRef]
- Shi, D.; Lu, X. Accumulation degree and source apportionment of trace metals in smaller than 63 μm road dust from the areas with different land uses: A case study of Xi'an, China. *Sci. Total Environ.* 2018, 636, 1211–1218. [CrossRef]
- 7. Choi, J.Y.; Jeong, H.; Choi, K.-Y.; Hong, G.H.; Yang, D.B.; Kim, K.; Ra, K. Source identification and implications of heavy metals in urban roads for the coastal pollution in a beach town, Busan, Korea. *Mar. Pollut. Bull.* **2020**, *161*, 111724. [CrossRef]
- Huang, C.; Zhang, L.; Meng, J.; Yu, Y.; Qi, J.; Shen, P.; Li, X.; Ding, P.; Chen, M.; Hu, G. Characteristics, source apportionment and health risk assessment of heavy metals in urban road dust of the Pearl River Delta, South China. *Ecotoxicol. Environ. Saf.* 2022, 236, 113490. [CrossRef]
- 9. De Miguel, E.; Llamas, J.F.; Chacón, E.; Berg, T.; Larssen, S.; Røyset, O.; Vadset, M. Origin and patterns of distribution of trace elements in street dust: Unleaded petrol and urban lead. *Atmos. Environ.* **1997**, *31*, 2733–2740. [CrossRef]
- Nakayama, S.M.; Ikenaka, Y.; Hamada, K.; Muzandu, K.; Choongo, K.; Teraoka, H.; Mizuno, N.; Ishizuka, M. Metal and metalloid contamination in roadside soil and wild rats around a Pb–Zn mine in Kabwe, Zambia. *Environ. Pollut.* 2011, 159, 175–181. [CrossRef]
- Heidari, M.; Darijani, T.; Alipour, V. Heavy metal pollution of road dust in a city and its highly polluted suburb; quantitative source apportionment and source-specific ecological and health risk assessment. *Chemosphere* 2021, 273, 129656. [CrossRef] [PubMed]
- 12. Li, R.; Yuan, Y.; Li, C.; Sun, W.; Yang, M.; Wang, X. Environmental health and ecological risk assessment of soil heavy metal pollution in the coastal cities of Estuarine Bay—A case study of Hangzhou Bay, China. *Toxics* **2020**, *8*, 75. [CrossRef]
- Vasile, G.; Dinu, C.; Kim, L.; Tenea, A.G.; Simion, M.; Ene, C.; Spinu, C.; Ungureanu, E.-M.; Manolache, D. Platinum group elements in road dust and vegetation from some European and national roads with intensive car traffic in Romania. *Rev. Chim.* 2019, 70, 286–292. [CrossRef]
- Kim, L.; Vasile, G.; Stanescu, B.; Calinescu, S.; Batrinescu, G. Distribution and bioavailability of mobile arsenic in sediments from a mining catchment area. J. Environ. Prot. Ecol. 2015, 16, 1227–1236. Available online: http://dspace.incdecoind.ro/bitstream/1234 56789/528/1/Distribution%20and%20bioavailability%20of%20mobile%20arsenic_JEPE_2015.pdf (accessed on 16 January 2024).
- 15. Chojnacka, K.; Chojnacki, A.; Górecka, H.; Górecki, H. Bioavailability of heavy metals from polluted soils to plants. *Sci. Total Environ.* **2005**, *337*, 175–182. [CrossRef]
- 16. Stanescu, B.; Cuciureanu, A.; Cernica, G.; Catrina, G.A. The current state of the quality of urban soils in Bucharest. *Rom. J. Ecol. Environ. Chem.* **2020**, *2*, 202–209. [CrossRef]
- Long, Z.; Zhu, H.; Bing, H.; Tian, X.; Wang, Z.; Wang, X.; Wu, Y. Contamination, sources and health risk of heavy metals in soil and dust from different functional areas in an industrial city of Panzhihua City, Southwest China. *J. Hazard. Mater.* 2021, 420, 126638. [CrossRef]
- Nduka, J.K.; Umeh, T.C.; Kelle, H.I.; Mgbemena, M.N.; Nnamani, R.A.; Okafor, P.C. Ecological and health risk assessment of heavy metals in roadside soil, dust and water of three economic zone in Enugu, Nigeria. Urban Clim. 2023, 51, 101627. [CrossRef]
- Enyoh, C.E.; Wang, Q.; Eze, V.C.; Rabin, M.H.; Rakib, M.R.J.; Verla, A.W.; Ibe, F.C.; Duru, C.E.; Verla, E.N. Assessment of potentially toxic metals adsorbed on small macroplastics in urban roadside soils in southeastern Nigeria. *J. Hazard. Mater.* 2022, 7, 100122. [CrossRef]
- 20. Yesilkanat, C.M.; Kobya, Y. Spatial characteristics of ecological and health risks of toxic heavy metal pollution from road dust in the Black Sea coast of Turkey. *Geoderma Reg.* 2021, 25, e00388. [CrossRef]
- 21. Xu, C.; Pu, J.; Wen, B.; Xia, M. Potential ecological risks of heavy metals in agricultural soil alongside highways and their relationship with landscape. *Agriculture* **2021**, *11*, 800. [CrossRef]
- 22. Hakanson, L. An ecological risk index for aquatic pollution control: A sedimentological approach. *Water Res.* **1980**, *14*, 975–1001. [CrossRef]
- Dinu, C.; Gheorghe, S.; Tenea, A.G.; Stoica, C.; Vasile, G.-G.; Popescu, R.L.; Serban, E.A.; Pascu, L.F. Toxic Metals (As, Cd, Ni, Pb) impact in the most common medicinal plant (*Mentha piperita*). *Int. J. Environ. Res. Public Health* 2021, *18*, 3904. [CrossRef] [PubMed]
- 24. Dinu, C.; Vasile, G.; Tenea, A.G.; Stoica, C.; Gheorghe, S.; Serban, E.A. The influence of toxic metals As, Cd, Ni and Pb on nutrients accumulation in *Mentha piperita*. *Rom. J. Ecol. Environ. Chem.* **2021**, *3*, 141–152. [CrossRef]

- 25. Li, C.; Du, D.; Gan, Y.; Ji, S.; Wang, L.; Chang, M.; Liu, J. Foliar dust as a reliable environmental monitor of heavy metal pollution in comparison to plant leaves and soil in urban areas. *Chemosphere* **2022**, *287*, 132341. [CrossRef] [PubMed]
- 26. Khan, S.; Khan, M.; Rehman, S. Lead and cadmium contamination of different roadside soils and plants in Peshawar City, Pakistan. *Pedosphere* **2011**, *21*, 351–357. [CrossRef]
- 27. Commision Regulation 2011/574/EU Amending Annex I to Directive 2002/32/EC of the European Parliament and of the Council as Regards Maximum Levels for Nitrite, Melamine, *Ambrosia* spp. and Carry-Over of Certain Coccidiostats and Histomonostats and Consolidating Annexes I and II Thereto. Available online: https://eur-lex.europa.eu/legal-content/EN/TXT/PDF/?uri=CELEX:32011R0574 (accessed on 21 November 2023).
- 28. Tome, F.V.; Rodríguez, M.B.; Lozano, J. Soil-to-plant transfer factors for natural radionuclides and stable elements in a Mediterranean area. *J. Environ. Radioact.* 2003, *65*, 161–175. [CrossRef]
- 29. A1 Motorway. Available online: https://en.wikipedia.org/wiki/A1_motorway_(Romania) (accessed on 26 November 2023).
- 30. Car Census 2015. Available online: https://www.cestrin.ro/assets/pdf/recensamant%202015.pdf (accessed on 7 January 2024).
- 31. Car Census 2022. Available online: https://www.cestrin.ro/assets/pdf/recensamant%202022.pdf (accessed on 7 January 2024).
- 32. Li, H.; Shi, A.; Zhang, X. Particle size distribution and characteristics of heavy metals in road-deposited sediments from Beijing Olympic Park. *J. Environ. Sci.* 2015, *32*, 228–237. [CrossRef]
- 33. SIST ISO 11466:1996. Soil Quality–Extraction of Trace Elements Soluble in Aqua Regia. Available online: https://standards.iteh. ai/catalog/standards/sist/81e301a6-0954-4959-805b-6488f9a94126/sist-iso-11466-1996 (accessed on 16 January 2024).
- 34. Order No. 756 of 3 November 1997 for the Approval of the Regulation on Environmental Pollution Assessment. Eminent: Ministry of Waters, Forests and Environmental Protection. (Published in: Official Gazette No 303 bis of 6 November 1997). Available online: http://legislatie.just.ro/Public/DetaliiDocumentAfis/151788 (accessed on 26 November 2023). (In Romanian).
- Taylor, S.R.; McLennan, S.M. *The Continental Crust: Its Composition and Evolution*; Blackwell Scientific Publications: Hoboken, NJ, USA, 1985. Available online: https://www.osti.gov/biblio/6582885 (accessed on 16 January 2024).
- 36. Müller, G. Index of geoaccumulation in sediments of the Rhine River. Geojournal 1969, 2, 108–118.
- Tomlinson, D.; Wilson, J.; Harris, C.; Jeffrey, D. Problems in the assessment of heavy-metal levels in estuaries and the formation of a pollution index. *Helgoländer Meeresunters* 1980, 33, 566–575. Available online: https://link.springer.com/article/10.1007/BF024 14780 (accessed on 16 January 2024). [CrossRef]
- 38. Dinelli, E.; Lombini, A. Metal distributions in plants growing on copper mine spoils in Northern Apennines, Italy: The evaluation of seasonal variations. *Appl. Geochem.* **1996**, *11*, 375–385. [CrossRef]
- 39. Reimann, C.; Filzmoser, P.; Garrett, R.G. Background and threshold: Critical comparison of methods of determination. *Sci. Total Environ.* **2005**, *346*, 1–16. [CrossRef] [PubMed]

Disclaimer/Publisher's Note: The statements, opinions and data contained in all publications are solely those of the individual author(s) and contributor(s) and not of MDPI and/or the editor(s). MDPI and/or the editor(s) disclaim responsibility for any injury to people or property resulting from any ideas, methods, instructions or products referred to in the content.