Geochemistry of Street Dust in Tyumen, Russia: Influence of Traffic Load

Dmitriy Moskovchenko (✉ moskovchenko1965@gmail.com)

Tyumen State University: Tumenskij gosudarstvennyj universitet <https://orcid.org/0000-0001-6338-7669>

Roman Pozhitkov

Tyumenskii gosudarstvennyi universitet: Tumenskij gosudarstvennyj universitet

Dzhamilya Ukarkhanova

Tyumenskii gosudarstvennyi universitet: Tumenskij gosudarstvennyj universitet

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Abstract

The article presents the results of studying the composition of road dust in Tyumen, a large city in Western Siberia. On roads with different traffic intensities, 50 dust samples were taken, in which the particle size distribution, pH, and organic carbon content were determined. The content of 62 major and trace elements was studied using atomic emission spectrometry (AES-MS) и inductively coupled plasma spectrometry (ICP-MS). It was revealed that the dust has an alkaline reaction $\text{pH} = 7.4 - 10.2$, low organic carbon content (0.07 - 2.9%). The particle size distribution is dominated by particles $\text{PM} = 100-250$. The content of small particles ($\text{PM} < 2$ and PM_{2-10}) representing the greatest environmental hazard is minimal on roads with an average traffic intensity. Studies have shown that the main road dust pollutants in Tyumen are Ni, Sb, Cr, Zn, Co. The average geoaccumulation index (I_{geo}) values are ranked as $\text{Ni} (2.2) > \text{Sb} (1.5) > \text{Cr} (1.3) > \text{Zn} (0.4) > \text{Co} (0.4) > \text{Cu} (0.2)$. The contamination evaluation through enrichment factor (EF) calculation showed that road dust is highly enriched in Ni and significantly in Cr and Sb. More than 80% of Zn, Co, Cu and more than 90% of Ni, Sb, Cr come from anthropogenic sources. By the content of Ni and Cr, Tyumen surpasses all cities of the Earth where similar studies were carried out. The high Ni content is associated with the composition of local soils and roadways, increased content in vehicle exhaust gases, and abrasion of metal parts. Calculations of the total pollution indices (IPI and Z_c) showed that the level of road dust pollution in most of Tyumen's territory is average, moderately hazardous.

Introduction

Road dust is currently one of the main objects in the study of the ecological state of urbanized areas. Its composition depends on the intensity of erosion of roadside soils, emissions from mobile sources, abrasion of road surfaces and markings, abrasion of tires and brake pads of vehicles, corrosion of their metal parts, traffic patterns and the use of deicing agents (Murakami et al. 2007; Irvine et al. 2009; Nazzari et al. 2013). Dust particles accumulate toxic metals and metalloids (TMM), organic compounds, including polycyclic aromatic hydrocarbons (Chow et al. 1996; Varrica et al. 2003; Denier van de Gon 2013; Amato et al. 2014; Kosheleva et al. 2018). The high concentration of toxic substances in the dust makes it hazardous to human health. Dust microparticles increase the risk of respiratory, cardiovascular and oncological diseases (Tager 2005). Small particles with a diameter $< 10 \mu\text{m}$ are most dangerous because they are in the air for a long time, enter the lungs and settle in them (Kjelgaard et al. 2004).

In many cities of Russia, a hazardous environmental situation caused by atmospheric air pollution has been repeatedly noted (Kasimov et al. 2014; Krupnova et al. 2020). However, there are few studies of road dust. The composition of road dust was determined in Moscow (Ladonin, Plyaskina 2009; Vlasov 2015; Kasimov et al. 2019b), cities of the Perm region (Kaigorodov et al. 2009); Chelyabinsk (Krupnova et al. 2020), Alushta (Kasimov et al. 2019a). In Tyumen, 20 samples of road dust were analyzed, as a result of which a conclusion was made about the high content of Cr, Ni, and Co (Konstantinova et al. 2020).

We have studied the content of heavy metals and metalloids in 50 samples of Tyumen road dust. The objectives of the presented work are to identify the features of HMM accumulation in road dust, depending on the traffic intensity and the location of other polluting objects, to assess the ratio of natural and

anthropogenic sources, to evaluate the level of pollution. The present study complements and expands the work of Konstantinova et al. (2020) by: (a) collecting a large number of samples from a wider area around the city of Tyumen (50 sampling sites); (b) measuring the concentrations of other major and trace elements (Al, Ba, Cd, Co, Cr, Cu, Fe, Mn, Ni, Pb, Sc, Sr, Ti, U, V and Zn) in the road dust; (c) estimates of the ratio of natural and anthropogenic sources of HMMs in road dust.

Materials And Methods

Study area

The study was carried out in Tyumen, the largest city in the Tyumen oblast (province), known primarily as a region of the oil production. Tyumen is located in the southwestern part of the West Siberian Plain, in the extreme south of the taiga zone. The climate of Tyumen is cold continental, the average annual air temperature is + 0.9 ° C, the amount of precipitation for the year is 480 mm (Reference Book 1998). Southerly and westerly winds prevail. The rapid development of Tyumen began in the second half of the 20th century after the discovery of numerous oil and gas fields in the north of Western Siberia. In the 1960s the population of Tyumen was about 150 thousand people, at the end of the 1990s it was about 500 thousand, and now it has exceeded 800 thousand people. In Tyumen, there are enterprises of mechanical engineering, instrument making and metalworking, oil refining, production of building materials, energy (combined heat and power plants). Transport streams pass through Tyumen to the northern regions, where oil and gas fields are located. The city operates 385 thousand vehicles, the total length of the road network is 1241 km (Konstantinova et al. 2019). Emissions from vehicles account for more than 80% of the gross emissions of pollutants entering the city's atmosphere (Krestyannikova et al. 2015). The traffic load on the roads with the most intensive traffic reaches 8.1 thousand vehicles per hour (Germanova and Kernozhitskaya 2014).

Sampling and laboratory analysis

Road dust sampling was carried out in June 2020 in dry weather. Roads with different traffic intensities were examined: high (>4 thousand vehicles per hour), moderate (1-4 thousand) and low (<1 thousand). The traffic intensity was determined according to the data (Guseinov, 2000; Germanova and Kernozhitskaya 2014). Roads with low traffic volume are common mainly on the outskirts of Tyumen in low-rise residential area, while traffic intensity is highest in the city center. Samples weighing 200-300 g were taken with a plastic brush and a scoop from the surface of the roadway on test plots 1 × 1 m in size, placed in plastic bags and delivered to the laboratory. A total of 50 samples were taken. The location of sampling points is shown in Fig. 1. A detailed description of the sampling sites is presented in Supplementary Materials Table 1.

In the laboratory, the samples were sieved through a sieve with a mesh size of 1 mm to remove impurities (vegetation residues, debris). Then, the samples were dried slowly using an oven. The pH value was measured in the samples using a HydroMaster HM-500 ionometer. The organic carbon (OC) content was determined by calcining to constant weight (GOST 23740-2016). The particle size distribution was

determined by laser diffraction using a Mastersizer 3000 laser particle size analyzer. The content of 54 trace elements (Li, Be, Sc, V, Cr, Co, Ni, Cu, Zn, Ga, As, Se, Rb, Sr, Y, Zr, Nb, Mo, Rh, Pd, Ag, Cd, Sn, Sb, Te, Cs, Ba, La, Ce, Pr, Nd, Sm, Eu, Gd, Tb, Dy, Ho, Er, Tm, Yb, Lu, Hf, Ta, W, Re, Ir, Pt, Au, Hg, Tl, Pb, Bi, Th, U) and 8 major elements (Na_2O , MgO , Al_2O_3 , P_2O_5 , S, K_2O , CaO , Fe_2O_3) were measured in road dust samples by using Inductively Coupled Plasma ICP-MS (X-7 Thermo Elemental, USA) and atomic emission method AES-MS (iCAP-6500, Thermo Scientific, USA).

For analysis, portions of samples weighing 100 mg were used. Sample digestion was carried out in an open system. The samples were placed in Teflon beakers (volume 50 ml), 0.1 ml of a solution containing $8 \mu\text{g dm}^{-3}$ ^{145}Nd , ^{61}Dy , and ^{174}Yb was added (control of the chemical yield during the sample decomposition procedure), and moistened with several drops of deionized water. Then 0.5 ml of HClO_4 (Perchloric acid fuming 70% Supratur, Merck), 3 ml (HF Hydrofluoric acid 40% GR, ISO, Merck), 0.5 ml of HNO_3 (Nitric acid 65%, max. 0.0000005% GR, ISO, Merck) and evaporated until intense white vapors appeared. The beakers were cooled, their walls were washed with water, and the solution was again evaporated to wet salts. Then 2 ml of HCl (Hydrochloric acid fuming 37% OR, ISO, Merck) and 0.2 ml of 0.1M H_3BO_3 solution (analytical grade) were added and evaporated to a volume of 0.5 - 0.7 ml. The resulting solutions were transferred into polyethylene bottles, 0.1 ml of a solution containing 10 mg L^{-1} In (internal standard) was added, diluted with deionized water to 20 ml, and analysis was performed. As control samples in Teflon beakers, the above procedures were performed without samples, and the resulting solutions were used as controls. Along with the analyzed samples, the decomposition of standard samples was carried out. To check the correctness of the analysis of dust samples, we used multi-element standard samples Trapp ST-2a (Russian State Standard GSO 8671-2005) and Basalt BHVO-2 (US Geological Survey). The methods, recoveries and analytical results of certified reference materials are given in Supplementary Materials (Table 2). The detection limit (DL) for measured elements are listed in Table 2.

Data analysis

Statistical processing of the results was carried out using the Excel and Statistica 6.0 packages. Statistical indicators of the composition of road dust (mean, median, standard deviation) were determined for Tyumen as a whole and separately for roads with different traffic intensities. The significance of differences in mean values for roads with different traffic intensities was assessed using the Mann-Whitney test.

To assess the level of pollution, the following indices were calculated: contamination factor (Cf) geoaccumulation index (Igeo), enrichment factor (EF), integrated pollution index (IPI) and total pollution index Z_c

The contamination factor is calculated based on the following equation:

$$Cf = C_i / C_b \quad (1)$$

where C_i is the concentration of the particular element in the dust and C_b is the content of background values. For road dust, due to the absence of a background analogue, the content of elements was

compared with the average content in the continental crust according to (Rudnisk, Gao, 2003).

Hakanson (1980) divided the contamination factor into four categories: $C_f < 1$ = low contamination factor indicating low contamination, $1 \leq C_f < 3$ = moderate contamination, $3 \leq C_f < 6$ = considerable contamination, $C_f \geq 6$ = very high contamination.

Igeo index was calculated according to Müller (1969) using the following equation:

$$I_{geo} = \log_2 \left(\frac{C_i}{1.5C_b} \right) \quad (2)$$

where C_i is the measured concentration of the particular element and C_b is the geochemical background (crustal average). According to (Müller, 1969), the contamination level can be classified based on a scale, where $I_{geo} \leq 0$ classified as uncontaminated, $0 < I_{geo} \leq 1$ = slightly contaminated, $1 < I_{geo} \leq 2$ = moderately contaminated, $2 < I_{geo} \leq 3$ = moderately to heavily contaminated, $3 < I_{geo} \leq 4$ = heavily contaminated, $4 < I_{geo} \leq 5$ = heavily to extremely contaminated $I_{geo} > 5$ = extremely contaminated (Müller 1969; Sutherland 2000).

The enrichment factor (EF) of an element, an important parameter for evaluating the impact degree of human activities on its enrichment, is the normalization of a measured element against a reference element in a studied sample (Al-Awadhi and AlShuaibi 2013). The EF was calculated according to equation:

$$EF = \frac{C_x}{Al} (sample) / \frac{C_x}{Al} (crust) \quad (3)$$

$EF \leq 2$ means deficiency to minimal enrichment, $2 < EF \leq 5$ corresponds to moderate enrichment, $5 < EF \leq 20$ signifies significant enrichment and significant pollution signal, $20 < EF \leq 40$ indicates very high enrichment and very strong pollution signal and $40 \geq EF$ means extremely high enrichment (Sutherland 2000; Yongming et al. 2006; Lu et al. 2009).

Further, based on the EF values, an assessment was made of the ratio between natural and anthropogenic sources that affect the composition of dust. A simple formula (Han et al. 2007; Chen et al. 2012) is used as below: assuming the concentration of element X in resuspended road dust is the sum of the two sources: crustal and pollution, i.e.

$$X_{road\ dust} = X_{crust} + X_{pollution} \quad (4)$$

$$X_{crust} = Al_{road\ dust} (X/Al)_{crust} \quad (5)$$

$$X_{pollution\ \%} = (1 - (Al/X)_{road\ dust} (X/Al)_{crust}) \times 100\% \quad (6)$$

The generalized level of road dust pollution by metals and metalloids was estimated using the total pollution indicator (Kasimov et al. 2012)

$$Z_c = \sum C_f - (n - 1) \quad (7)$$

For $C_f > 1.5$, where n is the number of elements with $C_f > 1.5$

The following values are taken as gradations of the pollution level: $Z_c < 32$ - non-hazardous pollution level, 32–64 - moderately hazardous, 64–128 - hazardous, 128–256 - very hazardous (Kasimov et al. 2012; Vlasov et al. 2015).

Another method used for the determination of trace element contamination is the IPI, which is defined as the average of the contamination factors computed for each trace element (Tume et al. 2018). The IPI is divided into four categories: low pollution level ($IPI < 1$), moderate pollution level ($1 < IPI < 2$), high pollution level ($2 < IPI < 5$) and extremely high pollution index ($IPI > 5$) (Salmanighabeshi et al. 2015).

To make the results more easily interpretable and to determine sources of pollution, the PCA with VARIMAX normalized rotation was also applied. When PCA with VARIMAX normalized rotation was performed, each PC score contains information on all of the metal elements combined into a single number, while the loadings indicate the relative contribution each element makes to that score (Yongming et al. 2006)

Results And Discussion

Physical and chemical properties of road dust

Table 1 shows the physicochemical indicators and particle size distribution of road dust on roads with different traffic intensities.

Table 1. Physicochemical indicators of road dust in Tyumen on roads with different traffic intensities.

Parameter	Traffic intensity					
	Low		Moderate		High	
	Mean	SD	Mean	SD	Mean	SD
Granulometric composition						
Sand (%)	86.6	5.1	76.8	11.7	81.3	7.4
Silt (%)	12.0	4.3	21.2	11.1	16.7	6.9
Clay (%)	1.5	0.8	2.1	0.7	2.0	0.6
Organic carbon (%)	0.68	0.35	1.22	0.57	1.01	0.44
pH	8.10	0.42	7.82	0.34	8.06	0.51

The dust has an alkaline reaction, the pH value varies from 7.4 to 10.2. The alkaline reaction is caused by the entry of microparticles of carbonate construction dust, as well as the redistribution of pollutants emitted by vehicles. Acidifying gaseous compounds, mainly nitrogen oxides, migrate off the roadway, while alkalizing dust particles (construction carbonate dust, deicing mixtures) remain on the road. The differences in the average pH values on roads with different traffic intensities are small (7.82 - 8.10) and,

according to the Mann – Whitney test, are statistically insignificant. The content of organic matter varies from 0.07 to 2.9% (average 1.1%). The minimum content of organic matter on roads with moderate traffic intensity, the maximum on roads with low traffic (Table 1).

Road dust from different cities of the world has a predominantly alkaline reaction of pH 7–10, the content of C_{org} varies significantly - from 1 to 17%, (Al-Khashman 2007; Ladonin and Plyaskina 2009; Acosta et al. 2011; Yisa et al. 2011; Hu et al. 2011; Sutherland et al. 2012; Vlasov et al. 2015; Gabarron et al. 2017). Thus, in terms of physical and chemical properties, the road dust of Tyumen does not differ from other cities on Earth.

The particle size distribution is dominated by the fine sand fraction (PM 100-250 μm), the content of which is 52% on average (varying from 3.8 to 67.4%). The content of very fine sand (PM = 50-100 μm) is much less and averages 11.2%. The content of the silt fraction is 18.1% (4.6-31.2%). The amount of clay particles (PM <2 μm) is very low (0.7 - 3.2%).

It is believed that the predominance of particles with a size of 180 ... 240 μm in road dust is evidence of the influx of soil particles and particles associated with traffic - abrasion of the roadway, tires, metal parts of cars (Christoforidis and Stamatis 2009). Particles with a size of 60 ... 90 μm come with emissions from industrial sources and thermal power plants (Krupnova et al. 2020). According to (Zaytseva et al. 2013), the dust particles less than 10 μm is formed in all technological processes at metallurgical enterprises. A significant predominance of particles with a diameter of 100–250 μm and a very small amount of PM <10 μm particles (Fig. 2) in the Tyumen road dust indicates a weak influence of industrial enterprises on its composition and the predominant effect of transport and soil erosion processes. Measurements of the number of particles PM 2.5 and PM 10 in the air confirm the low dustiness of the surface layer of the atmosphere in Tyumen. During the sampling period of road dust (June 2020), the average concentration of PM10 particles in the air was 0.020 mg / m³, which is significantly less than in most cities on Earth (Pozhitkov 2020).

Comparison of data on the granulometric composition of Tyumen road dust with data for other cities showed that the predominance of the sand fraction with a particle diameter of PM> 50 is typical for road dust and weakly depends on the composition of urban soils, population size and industrial specialization. So, in Moscow, road dust has a predominantly sandy loam composition (sandy loam), the share of PM> 50 averages 69.1%, PM10-50 - 16.3%, PM1-10 - 12.8%, PM <1 - 8% (Vlasov et al. 2015). In Chelyabinsk (Russia), road dust was dominated by particles ranging in size from 30 ... 40 to 250 ... 300 μm , (Krupnova et al. 2020). In Turin (Italy), road dust was significantly dominated by fractions of 50-200 μm and 200-2000 μm , the content of which was 27 and 62%, respectively (Padoan et al. 2017). In Shanghai, China, road dust is dominated by particles ranging in size from 100 to 400 μm (Shi et al. 2011). The predominance of the sandy fraction in road dust is associated with the removal of PM <50 μm particles, which rise during the movement of vehicles as a result of the action of turbulent eddies and can be carried even by weak winds (Vlasov et al. 2015). As the traffic intensity increases, the proportion of large sand particles in the composition of road dust increases (Nikiforova et al. 2011).

The distribution of road dust fractions on roads with different traffic intensities in Tyumen is as follows (Fig. 2). The largest number of small particles (PM <2 and PM2-10), which are most easily carried by the wind and pose the greatest danger to public health, was noted on roads with medium traffic. Least of all small particles on roads with low traffic volume, large roads occupy an intermediate position. Differences in the particle size distribution of dust on small medium and large roads are small. Verification using the Mann-Whitney criterion showed that the difference in the average values of the content of particles of the same aerodynamic diameter, depending on the traffic intensity, was not significant. Probably, natural winds have a decisive influence, under the influence of which PM <50 µm particles are carried away. The average wind speed in Tyumen is 3 m/s, during the sampling period (May) - 3.3 m/s (Reference Book 1998). Even at low wind speeds (2–3 m/s), particles 1–2 µm in size do not settle due to gravity (Charron and Harrison 2005;Viana et al., 2006).

Major elements

The compositions of major elements in road dust of Tyumen are shown in Fig. 3. For comparison, the composition of Quaternary deposits (aeolian sand) in the vicinity of Tyumen is presented in accordance with (Sizov et al. 2020). The composition of road dust is dominated by MgO (on average 12.9%), CaO (7.7%) Al₂O₃ and Fe₂O₃ (5.2%). Then K₂O, Na₂O, P₂O₅ follow in descending order. In the continental crust (Rudnick and Gao 2003), the content of elements decreases in a slightly different sequence MgO (46.7%), Al₂O₃ (15.4%), Fe₂O₃ (5.0%), CaO (3.6%) Na₂O (3.27%) K₂O (2.8%). Thus, in comparison with the Earth's crust, road dust is enriched with calcium, which is caused by the intake of carbonate cement dust during abrasion of curbs, the use of anti-ice reagents. Road dust also contains significantly more CaO than sandy aeolian deposits. More MgO, P₂O₅, Fe₂O₃ in road dust than in Quaternary sediments (see Fig. 3). The intake of phosphorus is associated with soil organic matter. Fe₂O₃ comes from the corrosion of metal parts of vehicles, magnesium is part of deicing agents.

Trace elements content in road dust

Table 2 shows statistics related to the contents of trace elements. A number of elements (Se, Rh, Pd, Te, Re, Ir, Pt, Au) were excluded from the calculations, since their content in more than 50% of the samples was below the detection limit. For other elements, in the event that the content was <DL, half the value of the DL was indicated. Compared to the average content in the earth's crust, the Tyumen road dust is enriched in Ni, Sb, Cr, Zn, Co, Cu, Pb, Cd, W, Ag. The concentrations of Mo, As, Sn, Bi are close to the average content in the earth's crust, the rest of the elements are in deficit. The concentrations of Cu (18–130.4 mg kg⁻¹), Ni (322.4–1042 mg kg⁻¹), Hg (0.01-0.15 mg kg⁻¹), Pb (8.5–120.2 mg kg⁻¹), and Cd (0.02–0.44 mg kg⁻¹) in street dust of Tyumen varied greatly. This indirectly indicates the influence of pollution sources that cause an increase in the TMM content in road dust.

Table 2. Summary statistics of trace elements content in Tyumen road dust (n=50).

Element	Detection limit	Mean	SD	Median	min	max	Earth crust (Rudnik and Gao 2003)	Cf
Li	0.03	6.8	1.4	6.7	4.1	10.2	21.0	0.32
Be	0.03	0.6	0.14	0.6	0.4	1.0	2.1	0.29
Sc	0.09	10.1	1.2	9.9	5.5	13.3	14.0	0.72
V	0.8	66.4	10.5	66.5	27.5	104.9	97.0	0.68
Cr	0.7	507.9	112.7	495.3	282.6	740.6	92.0	5.5
Co	0.08	39.6	8.3	38.8	24.8	59.3	17.3	2.3
Ni	0.7	632.1	164.7	617.2	322	1042	47.0	13.4
Cu	0.8	57.4	21.5	52.2	18.0	130.4	28.0	2.1
Zn	0.5	160.8	44.7	157.8	59.6	271	67.0	2.4
Ga	0.1	5.2	0.8	5.3	3.1	6.6	17.5	0.29
As	0.1	5.7	3.0	5.3	1.7	20.9	4.8	1.19
Rb	0.1	27.1	6.4	28.0	15.6	40.3	84.0	0.32
Sr	0.07	147.3	28.9	145.8	100	244	320	0.46
Y	0.02	7.3	1.6	7.5	3.1	10.7	21.0	0.35
Zr	0.04	60.7	21.2	60.4	17.4	133.5	193.0	0.31
Nb	0.02	6.6	2.4	6.3	1.7	12.4	12.0	0.55
Mo	0.04	1.4	0.7	1.2	0.53	4.6	1.1	1.30
Ag	0.03	0.076	0.038	0.07	0.04	0.31	0.053	1.4
Cd	0.04	0.2	0.08	0.15	0.02	0.44	0.09	1.8
Sn	0.03	2.2	1.0	1.9	0.6	5.8	2.1	1.05
Sb	0.06	3.1	1.9	2.6	1.1	13.2	0.4	7.8
Cs	0.01	0.9	0.20	0.84	0.4	1.4	4.90	0.17
Ba	0.05	317.1	69.7	328.4	177.6	435.8	624	0.51
La	0.009	8.9	2.7	8.9	3.7	15.2	31.0	0.29
Ce	0.008	20.1	5.6	20.3	8.4	31.2	63.0	0.32
Pr	0.005	2.0	0.6	2.1	0.9	3.3	7.1	0.29
Nd	0.009	8.7	2.5	9.0	4.2	14.8	27.0	0.32
Sm	0.004	1.6	0.4	1.6	0.8	2.8	4.7	0.35

Eu	0.006	0.4	0.09	0.42	0.22	0.62	1.0	0.43
Gd	0.007	1.4	0.34	1.5	0.63	2.2	4.0	0.36
Tb	0.004	0.2	0.05	0.22	0.10	0.33	0.7	0.31
Dy	0.007	1.3	0.30	1.32	0.55	2.0	3.9	0.34
Ho	0.005	0.3	0.06	0.27	0.11	0.41	0.8	0.33
Er	0.003	0.8	0.17	0.82	0.36	1.19	2.3	0.35
Tm	0.004	0.1	0.03	0.12	0.04	0.17	0.3	0.39
Yb	0.003	0.8	0.2	0.8	0.29	1.3	2.0	0.41
Lu	0.005	0.1	0.0	0.1	0.04	0.20	0.3	0.40
Hf	0.02	1.9	0.7	1.8	0.5	4.8	5.3	0.35
Ta	0.01	0.5	0.24	0.5	0.15	1.8	0.9	0.61
W	0.03	3.2	1.3	3.1	0.8	7.1	1.9	1.7
Hg	0.01	0.024	0.021	0.019	0.005	0.146	0.1	0.48
Tl	0.005	0.143	0.026	0.146	0.1	0.2	0.9	0.16
Pb	0.06	33.9	20.7	28.7	8.5	120.2	17.0	2.0
Bi	0.01	0.17	0.28	0.12	0.03	2.08	0.2	1.07
Th	0.01	1.9	0.6	1.9	0.7	3.5	10.5	0.18
U	0.01	1.1	0.5	1.0	0.50	4.4	2.7	0.41

The values of the pollution index Cf are maximal for Ni, Sb, Cr, Zn, Co (Table 2). According to the classification of Hakanson (1980), in terms of Cf, the content of Co, Cu, Zn, As, Mo, Ag, Cd, Sn, W, Pb Bi corresponds to the level of "moderate contamination", Cr - "significant contamination" and Ni, Sb - "very high contamination". The pollution of road dust in Tyumen with chromium and nickel was previously noted by Konstantinova et al. (2020), who revealed that the concentration of these elements is 4 and 7 times higher, respectively, compared to urban soils. An increased content of Cr, Ni, Pb, V was also noted in the snow cover (Guseinov et al. 1997), which confirms the atmospheric input of these metals.

Geoaccumulation index and enrichment factor

The calculated results of Igeo of heavy metals in road dust from Tyumen are presented in Fig. 4. In terms of Igeo values, the elements form the following row: Ni_{2.2} Sb_{1.5} Cr_{1.3} Zn_{0.4} Co_{0.4} Cu_{0.2} Cd_{0.1} Pb_{0.1} W_{0.04} (the index denotes the average of the Igeo). For the rest of the elements, Igeo < 0. Thus, according to the classification (Müller 1969), Ni belongs to the category "moderately to heavily contaminated", Sb, Cr - "moderately contaminated", Zn, Co, Cu, Cd, Pb, W – "slightly contaminated". As Cd, Ag, Mo, Sn, Hg, Sb, Pb are characterized by a wide variation in Igeo values (Fig. 3), depending on local sources of pollution. For

example, for Pb, the values of $I_{geo} > 1$, corresponding to the “moderately contaminated” level, were noted near the car battery production plant. Earlier it was revealed that the concentration of Pb in the soils on the territory of the battery plant is extremely high ($>1000 \text{ mg kg}^{-1}$) (Skipin and Beresneva, 2014).

The value of EF values on roads with different traffic intensities is given in Table 3 (only metals with EF values > 1.5 are presented). On roads with low traffic volumes, the EF value of Ni, Co, Cr, Sb is lower than on roads with high and medium traffic volumes. Verification using the Mann-Whitney test showed that small roads significantly differ from medium and large ones in the enrichment of road dust with chromium, nickel, antimony, mercury and bismuth ($p = 0.01$). Differences between the EF of cadmium for small and large roads are also significant. Differences in the content of other elements are statistically insignificant. Thus, the EF calculations confirm the effect of transport on the release of Ni, Cr, Sb into the environment. An assessment of the ratio between natural and anthropogenic sources affecting the composition of dust, carried out in accordance with formulas 4-6, showed that the elements form the following series (the index means the share of anthropogenic input, %): Ni₉₇ Sb₉₅ Cr₉₃ Zn₈₅ Co₈₄ Cu₈₂ Pb₇₉ W₇₆ Cd₇₆ Ag₇₄ Mo₇₁ Sn₆₁ Sc₅₃ Bi₅₂ V₅₀. The rest of the elements come primarily from natural sources.

Table 3. EF values on roads with different traffic volumes.

Elements	Traffic intensity		
	low	moderate	high
Ni	23.7	33.6	33.7
Sb	16.7	21.9	19.8
Cr	10.3	13.9	13.6
Pb	5.6	5.6	3.9
Cd	5.5	5.0	3.6
Zn	4.9	6.1	5.7
Cu	4.7	5.2	4.7
Co	4.4	5.7	5.7
Mo	3.3	3.3	2.9
W	3.5	4.2	4.2
As	3.1	2.8	2.9
Ag	3.2	3.8	3.2
Sn	2.0	2.7	2.5
Bi	1.6	2.2	3.2
Hg	1.9	1.2	1.0
Sc	1.69	1.75	1.73
V	1.74	1.66	1.60

It is characteristic that on small roads the dust is enriched with some chalcophilic elements (Hg, As, Pb, Cd). This is probably due to the influence of burning coal for heating private residential buildings, which prevail in urban areas with low traffic. Combustion of hard coal is the major source of airborne Hg, Mo and a very significant source for As, Cr, Mn, Sb (Nriagu and Pacyna 1988).

To assess the features of pollution in Tyumen, we compared the calculated EF values with the EF values in other cities of the Earth (Table 4). In Tyumen, the composition of elements enriched in road dust differs sharply from other cities. As a rule, in most cities on Earth, the maximum degree of enrichment is characteristic of chalcophilic elements, which are highly mobile. In Changsha City, the mean EFs revealed the following order: $Cd > Zn \approx Pb \approx Cu > Cr$ (Li et al. 2016). Similarly, in Kabul (Afghanistan), the EF values decrease in the sequence $Cd > Pb > Zn > Cu > Ni > Cr$ (Jadoon 2018). In Beijing, bulk atmospheric deposition Cd is the most anthropogenic enrichment metal with an EF of 32.4 proceeding As (11.5), Zn (9.3), Pb (7.7), and Cu (5.4) (Guo et al. 2017). In Tyumen, a different distribution of EF values is observed, which decrease

in the sequence Ni> Cr> Zn Co> Pb> Cd. To understand the reasons for the identified differences, it is necessary to determine the sources of the main road dust pollutants in Tyumen.

Table 4 The average EFs values of heavy metals in road dust collected in different areas

City, country	Cr	Co	Ni	Cu	Zn	As	Cd	Pb	Mn	Reference
Tyumen, this study	<i>13,3</i>	<i>5,5</i>	<i>32,3</i>	4,9	<i>5,8</i>	2,9	4,4	4,8	2,8	This study
Kabul, Afghanistan	<i>1.10</i>	<i>0.82</i>	<i>1.76</i>	2.46	3.92	-	<i>8.75</i>	<i>5.97</i>	<i>0.88</i>	Jadoon 2018
Bayan Obo, China	<i>5.31</i>	3.98	2.53	3.74	<i>8.82</i>	2.7	<u>81.4</u>	<i>19.5</i>	<i>11.0</i>	Li et al. 2015
Changsha City, China	<i>1.71</i>	-	-	2.6	4.0	-	<u>128.0</u>	3.41	-	Li et al. 2016
Tongchin, China	<i>1.74</i>	3.32	<i>0.88</i>	<i>1.55</i>	2.04	<i>0.61</i>	-	3.58	<i>0.67</i>	Zhang et al. 2015
Dehli, India	3.5-16.1	-	2-3	3.3-22.4	3.3-7.3	-	-	5.1-21.9	-	Rajaram et al. 2014

EF ≤ 2 means deficiency to minimal enrichment; normal font - 2 < EF ≤ 5 - moderate enrichment; bold italic 5 < EF ≤ 20 means significant enrichment; bold - 20 < EF ≤ 40 - very high enrichment; bold with underline 40 ≥ EF - extremely high enrichment

Identification of sources of pollution.

Studies have shown that Ni, Sb, Cr, Zn, Co are the main road dust pollutants in Tyumen. The content of these elements is increased throughout the city. Ni, Cr, Co belong to the geochemical group of siderophilic elements (Kabata-Pendias 2011) i.e. elements related to iron. The main source of nickel and chromium pollution is metallurgical plants (Saet et al 1990). Elements related to ironworking activities are Fe, Co, Cr, Mn, Ni (Zibert and Rokavec 2010). In Tyumen, the few and low-power metallurgical and metal-working enterprises (Tyumen electrometallurgical plant, JSC Sibneftemash, Zavod Tyumenremdormash CJSC) do not cause road dust pollution throughout the city, since no increase in Ni and Cr was found in their locations. Only the influence of the Tyumen Battery Plant, in the area of which a high lead content was found, has been reliably confirmed.

Soils are a natural source of Ni input. In the soils of Tyumen, the Ni content is increased, its average concentration is, according to various estimates, from 46.5 mg kg⁻¹ (Konstantinova et al. 2019) to 95 mg kg⁻¹ (Moskovchenko 1998), which is significantly higher than the average world content in the surface horizon of background chernozem soils constituting, according to (Kabata-Pendias 2011) 25 mg kg⁻¹. However, the Ni content in road dust is one mathematical order of magnitude higher than in soil (on average 632 mg kg⁻¹, see Table 2). Similarly, the average Cr content in the soils of Tyumen is estimated

from 107 mg kg⁻¹ (Konstantinova et al. 2019) to 176 mg kg⁻¹ (Moskovchenko 1998), with an average world value for chernozems of 77 mg kg⁻¹ (Kabata-Pendias, 2011). But in Tyumen road dust, the concentration of Cr exceeds 500 mg kg⁻¹. Co content in soils 16 - 20.9 mg kg⁻¹ (Moskovchenko 1998; Konstantinova et al. 2019), in road dust 39.6 mg kg⁻¹. A significant excess of the content of Ni, Cr, Co in road dust compared to soils, as well as an increased content in snow of Tyumen (Guseinov et al. 1997), gives grounds for the conclusion that there are anthropogenic sources of these elements.

According to Yang et al. (2010), Ni, Fe, Cr enter city dust mainly from transport. At the same time, the sources of Ni input are diverse. The most important source of Ni is the combustion of petroleum and petroleum products (Nriagu and Pacyna 1988). Global emission of Ni from oil products combustion it is estimated to range from 10 to > 40 kt yr⁻¹ (Kabata-Pendias 2011). Diesel engines use fuels with Ni additives (Al-Khashman 2007). Nickel in road dust is also found in asphalt bitumen and gabbro rock material (Lindgren (1996). Crushed stone used in road construction in Tyumen consists of rocks, including gabbro. Corrosion of metal parts of motor vehicles is a significant source of Ni (Ferguson and Kim 1991; Akhter and Al-Jowder 1997; Atiemo et al. 2011) Cr, Co and W also come with metal abrasive dusts, including as a result of abrasion of metal parts of vehicles (Saet et al 1990).

In addition road dust pollutants in Tyumen are Sb, Zn, Cu, Cd, Pb, W. Antimony used in the manufacture of car batteries. Abrasion of tires, metal parts of cars and road markings is also a source of Sb (Vlasov et al. 2015; Ozaki et al. 2021). As noted by Hjortenkrans et al. (2006) on motorway sections where traffic is braked (traffic lights, intersections, etc.) in decelerating environments such as roads with traffic lights, roundabouts and intersections, the Sb concentration was more than 8 times the background level. Similarly, Amato et al. (2009), studying the variability of the chemical composition of road dust in Spain, found that Sb, Zn and Mo are concentrated in the areas of vehicle braking, showing a strong relationship with brake pad and tire wear. It was also noted that Sb, As, Sn are supplied by burning coal for heating residential buildings (Landing et al., 2010).

Zinc and copper are also classified as elements which largely come to the environment from transport. Zn may have its origin from automotive sources, such as wear and tear of vulcanized rubber tires, lubricating oils, and corrosion of galvanized vehicular parts (Banerjee 2003; Ellis and Revitt 1982). Zn is added to tire tread as zinc oxide and organozinc compounds to facilitate the vulcanization process (Dall'Osto et al. 2014). The concentration of Zn in car tires is about 1% (Adachi and Tainosho 2004), Principal source of Cu in the atmosphere is mainly fossil fuel burning, traffic emissions, fuel combustion, and industrial combustion (Nriagu 1996).

To clarify the sources of pollution, a PCA analysis of the obtained data set was carried out, including the content of macro- and microelements, physical and chemical properties and particle size distribution. PCA with VARIMAX normalized rotation was applied to assist in the identification of sources of pollutants. The essence of PCA analysis is to "fold" a multicomponent data set to a limited, user-selected number of factors that determine the sample variance. The results obtained made it possible to identify 3 main components (PC), which determine the regularities of the formation of the chemical composition of road dust (Table 5).

Table 5. Varimax rotated principal component (PC) loadings for element concentrations, pH and clay content in road dust samples.

Variables	PC1	PC 2	PC 3
V	-0.04	0.13	0.69
Cr	0.92	0.11	-0.21
Co	0.93	0.16	-0.16
Ni	0.92	0.10	-0.26
Cu	-0.14	0.26	0.61
Zn	0.15	0.77	0.08
As	0.32	0.41	-0.15
Mo	-0.14	0.52	0.33
Ag	-0.25	0.32	0.03
Cd	0.04	0.69	0.13
Sn	0.22	0.62	0.09
Sb	-0.04	0.54	0.14
W	0.47	0.60	0.07
Hg	-0.35	0.24	-0.03
Pb	-0.24	0.51	0.10
Bi	0.17	0.32	-0.18
MgO	0.88	0.21	-0.37
Al ₂ O ₃	-0.65	0.24	0.58
P ₂ O ₅	-0.01	0.65	-0.13
S	0.42	0.54	0.02
K ₂ O	-0.80	-0.07	0.45
CaO	0.34	0.41	0.02
TiO ₂	-0.48	-0.09	0.68
MnO	0.05	0.33	0.83
Fe ₂ O ₃	0.75	0.17	0.51
C _{total.}	0.12	0.53	-0.70
Clay	0.12	0.53	-0.70
pH	-0.17	0.03	0.49

Expl.Var	6.25	4.90	4.50
Variance, %	22	17	16

Factor loads are classified as “strong”, “medium” and “weak” with values > 0.75; 0.75-0.50 and 0.5-0.3, respectively (Liu et al. 2003). Heavy loads are shown in bold in Table 5. The three PCs together account for 55% of the variance. The first component explains 22% of the total variance and has strong loading of Cr, Co, Ni, MgO, Fe₂O₃. This PC is to be associated with the traffic sources. The second factor has strong loading of Zn and moderate loading of Mo, Cd, Sn, Sb, W, P, P₂O₅, S, C_{total}, clay content. The most important source of Cd and Zn is the combustion of solid fuels and household waste (Saet et al 1990; Nriagu and Pacyna 1988), which gives grounds to associate this PC with the influence of household activities of the population. The third factor account for 16% of the total variance. It is composed of MnO (strong correlated), V, TiO₂, Cu, Al₂O₃, C_{total}, and clay content (moderate correlated). This factor can be interpreted as a natural factor of soil erosion. A similar distribution of factor loads was established in the soils of Tyumen. According to Konstantinova et al. (2020) V – Cr – Co – Ni – Cu association had the most significant strong positive value (47.5% of total variance).

Environmental assessment

The value of the total pollution index IPI, which takes into account the contents of all elements, varies in a small range, from 0.66 to 1.95. Low pollution level (IPI <1) is noted at 2 points. All others belong to the moderate pollution level (1 <IPI <2). Calculations of the total pollution index Zc, which takes into account only those elements whose content is 1.5 higher than the background level and excludes random variations, showed that in 82% of cases the pollution level is moderate (Zc = 32-64). A hazardous level of pollution was detected at 1 sampling point, low, non-hazardous - in 16%. The maximum value of Zc was noted at the sampling point located in the industrial zone of the city, near the thermal power plant. At this observation point, an abnormal concentration of antimony (13.2 mg kg⁻¹) and zinc (258 mg kg⁻¹) was detected. Thus, various indices showed that the level of road dust pollution in Tyumen is generally moderate.

To assess the ecological situation in Tyumen, we made a comparison with the data obtained in various cities of the Earth (Table 6). The concentration of trace elements in the road dust of different cities is very different, which is caused by differences in the intensity of the traffic load, the use of fuel of different composition, the characteristics of the road surface, the soils prevailing in the city, etc. The comparison shows that Tyumen has significant pollution with nickel, cobalt and chromium. Both our data and the data obtained earlier by Konstantinova et al. (2020) indicate that Tyumen is far superior to other cities in the concentration of these elements. The average concentration of Ni and Cr in the road dust of Tyumen is one order of magnitude higher than in other cities of Russia and the Earth as a whole. The content of Cu, Zn, As, Sb relative to other cities is at an average level, and Pb, Cd is rather low.

Table 6. Literature data on published metal median concentrations (mg kg⁻¹) in street dust from cities around the world.

City, country	Cr	Co	Ni	Cu	Zn	As	Cd	Pb	Sb	Reference
Tyumen, this study	508	39,6	632	57,4	161	5,7	0,2	33,9	3,1	This study
Tyumen, Russia	415,4	25,6	324	51,3	105	8,8	0,19	20,1	1,83	Konstantinova et al. 2020
Moscow, Russia	50	8,0	26	93	252	2,8	0,61	53	4,6	Vlasov et al. 2015
Chelyabinsk, Russia	48,5	6,3	21,9	55,9	154	3,8	0,4	14,4	1,3	Krupnova et al. 2020
Alushta, Crimea	31	7.4	33	44	127	8.0	0.3	37	1,5	Kasimov et al. 2019a
Ottawa, Canada	43.3	8.3	15.2	65.8	112	1.3	0.6	39	0.89	Rasmussen et al. 2001
Kabul, Afganistan	38.4	8.52	66.4	43.6	122.5	-	1.16	28.7	-	Jadoon 2018
Hangzhou, China	51	20	26	116	321	-	1.59	202	-	Zhang and Wang (2009)
Guangzhou, China	36-176	2.8-8.8	9.4-48.3	69-376	196-1150	-		40.5-406	-	Bi et al. 2013
Urumqi, China	54.3	10.9	43.3	94.5	294	-	1.17	53.5	-	Wei et al. 2010
Tongchuan, China	106.5	31.7	25.3	32.4	142	6.7	-	75.2	-	Zhang et al. 2015
Seul, Korea	151	-	-	396	795	-	-	144		Kim et al. 2007
Dhanbad and Bokaro region, India	57	10	24	26	78	-	-	48	-	Singh 2011
Luanda, Angola	26	2.9	10	42	317	5.0	1.1	351	3.4	Ferreira-Baptista and De Miguel 2005
Oslo, Norway	-	19	41	123	412	-	1.4	180	6.0	De Miguel et al.1997
Xi'an, China	145	30.9	30,8	54.7	268.6	-	-	124.5	-	Pan et al., 2017
Manchester, UK	-	-	-	113	282	-	-	653	-	Robertson et al. 2003

The high level of Ni Cr Co content in the Tyumen road dust is caused, in our opinion, by several reasons. Firstly, the increased content of these elements in soils, caused by natural lithological features, affects. The

enrichment of soils and parent rocks of Cr and Ni in the vicinity of Tyumen was noted by Konstantinova et al. (2019). Secondly, there is a large number of cars in the city and a high traffic load, which leads to a significant intake of these microelements due to corrosion and abrasion of metal parts. In our opinion, the abrasion of the studs used in the "winter" anti-icing tires contributes to the pollution. The body of the studs is made of either aluminum alloy or steel, the core is made of tungsten carbide. In the cheaper tires, which are widely used in Tyumen, the studs have a steel shell. Considering that, winter tires in Tyumen are used for almost half a year, the abrasion of steel studs is a significant source of Fe, Ni, Cr, Co - those elements, the content of which is increased in Tyumen compared to other cities.

Conclusion

In the presented work, the composition of road dust in Tyumen, an intensively developing industrial city in Siberia, has been studied. The physicochemical parameters, particle size distribution, the content of heavy metals and metalloids on roads with different traffic intensities have been determined. Road dust in Tyumen, like in most other cities on Earth, has an alkaline reaction, which is caused by the entry of carbonate microparticles. The total content of organic matter varies from 0.07 to 2.9%, the largest amount of organic matter is noted on roads with low traffic intensity, which indicates the influence of soils. The particle size distribution is dominated by the PM fraction of 100-250 μm . The content of finer particles (PM <50 μm), which largely come from industrial plants, is low. Thus, the composition of road dust is formed mainly under the influence of traffic-related and soil erosion sources. The largest number of small particles (PM <2 and PM₂₋₁₀) was observed on roads with medium traffic intensity. On roads with the most intensive traffic, blowing out small particles leads to a decrease in their proportion in the particle size distribution. The increased content of small particles on roads with medium traffic volumes leads to the fact that there is the highest concentration of pollutants.

Compared with the average content in the Earth's crust, Tyumen's road dust is enriched in Ni, Sb, Cr, Zn, Co, Cu, Pb, Cd, W. More than 90% of Ni, Sb, Cr comes from anthropogenic sources. Dust collected on low-traffic roads contains less Cr, Ni, Sb, Bi, Hg than on medium-to-high traffic roads. The Mann-Whitney test showed a high degree of significance of differences ($p = 0.01$), which indicates a transport source of these elements.

Comparison of the obtained results with the data on the trace elements content in the road dust of other cities of the Earth showed that in Tyumen the road dust contains a lot of Ni, Cr, Co. The average concentration of Ni and Cr in the road dust of Tyumen is one order of magnitude higher than in other cities of the Earth. The high content of these metals is associated with the composition of local soils and roadways, abrasion of metal parts of machines. However, other trace elements that are considered indicators of anthropogenic impact (Cu, Cd, Pb) are contained in Tyumen's road dust in moderate amounts. Therefore, the value of the total pollution indicators (IPI, Zc) at most observation points corresponds to the average level.

Declarations

Ethics approval and consent to participate

Not applicable (this paper does not contain studies involving human participants, human data, or human tissue).

Consent for publication

Not applicable (this paper does not contain any individual person's data in any form).

Availability of data and materials

The datasets generated during and/or analyzed during the current study are available from the corresponding author on reasonable request.

Competing interests

The authors declare that they have no competing interests.

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Authors' contributions

Conceptualization and methodology, D.M; Field sample acquisition, R.P and D.U.; Writing— re-view & editing, D.M., A.Z.; Visualization, R.P and D.U. All authors have read and agreed to the published version of the manuscript.

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Corresponding author

Correspondence to Dmitriy Moskovchenko

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Figures

Figure 1

Sampling sites and land-use areas within the city of Tyumen, Russia: 1 – sampling sites; 2–power plants; 3 – main federal roads; 4 – trans-siberian railway; 5 – main city roads; 6 – high-rise residential area; 7 – low-rise residential area; 8 – modern business zones; 9 – historical center; 10 – industrial zones; 11 – recreation and unbuilt areas

Figure 2

The proportion of particle size fractions in road dust. traffic intensity: 1 - low, 2 - moderate, 3 - high

Figure 3

Major elements mean concentrations (in weight percent oxide) in Tyumen road dust (this study, n=50) and Quaternary aeolian deposits (Sizov et al. 2020) 1 - road dust 2 - aeolian deposits

Figure 4

Boxplots of the geoaccumulation index (I_{geo}) for the 14 heavy metals and metalloids in road dusts of Tyumen. The horizontal line within the box represents the median value. The box represent the 25th and 75th quartiles. The middle line represent the 95% confidence interval. The data point not in the range of whiskers is considered as outliers. EF calculations showed that Tyumen's road dust is very high enriched in Ni and significantly enriched in Cr and Sb. 70% of the samples are significantly enriched in Zn, 62% in Co, 42% in Cu, 34% in Pb, and 26% in Cd. Moderate enrichment of road dust was noted for Mo, As, W, Sn, Bi, W. The remaining 30 elements are classified as "deficiency to minimal enrichment" ($EF < 2$).

Supplementary Files

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