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Chemical composition, morphology, and distribution of particulate matter produced by road pavement abrasion using different types of aggregates and asphalt binder

Dusan Jandacka¹*, Dasa Kovalova², Daniela Durcanska¹ and Martin Decky¹

Abstract: This study provides an assessment of the impact of asphalt mixture composition on the particulate matter (PM) emission from the pavement surface into the atmosphere. Pavement surface abrasion was also analyzed and potential differences among asphalt mixtures were determined by the chemical composition of captured PM. Various types of asphalt mixtures and the influence of their compositions on PM concentrations and chemical composition were studied and compared. The laboratory test setup included 12 different pavement samples. On average, the asphalt mixtures with a combination of melaphyre and dolomite as aggregates exhibited the highest values of PM. Measurements of PM₂.₅ and PM₁ in the wheel tracking machine were used to study the impact of asphalt mixture properties on pavement surface abrasion. The dependence between the aggregate used in the tested mixtures and the abraded particulates has been confirmed. The Ca content in aggregates was strongly correlated with the abraded particulates, which was not observed for the Si content. The multidimensional statistical methods used showed the division of markers of the asphalt mixture

ABOUT THE AUTHOR

The authors work at the University of Zilina (UNIZA), Slovakia. Their research interests are focused on air pollution from road transport, road design and environmental impact assessment, holistic approach to sustainable construction and management of roads, especially their pavements, development of innovative climate-adaptive materials and pavement constructions. The authors collaborated on the international project “Air Tritia – Uniform Approach to the Air Pollution Management System for Functional Urban Areas in Tritia Region”, and other. During the project was created an air quality system “AQMS” for functional urban areas of the TRITIA region – Poland, the Czech Republic and Slovakia, and a strategy of sustainable transport of territorial units. The research reported in this paper represents an important area of the particulate matter air pollution, which is a part of the authors’ research on the air pollution sources identification.

PUBLIC INTEREST STATEMENT

Particulate matter (PM) air pollution is a persistent problem in an increasingly large area. In particular, cities are affected, where many sources contribute to this pollution. However, do we know which ones? Major sources of PM air pollution in urban areas include exhaust emissions, road dust resuspension (traffic-related abrasions and pavement surface abrasions), and wood and coal household heating. This study reveals the potential for particulate matter production from road surface abrasion. During the research, several asphalt mixtures used for the road surface were studied. The chemical composition of the asphalt mixtures components (aggregate and asphalt binder) and the particulate matter produced during the road surface abrasion was identified. Finally, the sensitivity of asphalt mixtures to abrasion and chemical fingerprint of used aggregate and asphalt binder was revealed. This knowledge can be used to identify sources of particulate matter air pollution in the environment.
materials into various factors (sources). The morphology of the solids was examined by electron microscopy with X-ray analysis, indicating the origin of the particles (aggregate, asphalt binder).

Subjects: Environmental Change & Pollution; Multivariate Statistics; Transportation Engineering; Pavement Engineering; Pollution

Keywords: particulate matter; abrasion; asphalt mixture materials; chemical elements; pavement surface

1. Introduction

Air pollution is a global threat leading to large impacts on health and ecosystems. Emissions and their concentrations have increased in several areas over the world. In Europe, air quality remains unsatisfactory in many areas, despite reductions in emissions and ambient concentrations (EEA, 2017; EEA, 2018). A total of 13% and 6% of the European Union (EU)-28 urban population was exposed to PM_{10} (inhalable particles, with diameters that are generally 10 µm and smaller) and PM_{2.5} (inhalable particles, with diameters that are generally 2.5 µm and smaller) levels above the corresponding daily limits in 2016, respectively. These percentages were the lowest since 2000 for PM_{10} and 2006 for PM_{2.5} (EEA, 2018).

Air pollution continues to have significant impacts on the health of the European population, particularly in urban areas. Considerable economic impacts, reduced life expectancy, increased medical costs, and reduced work productivity are also consequences of air pollution. Europe’s most dangerous pollutants in terms of harm to human health are particulate matter (PM), nitrogen dioxide (NO_{2}), and ground-level ozone (O_{3}) (EEA, 2017; EEA, 2018). Estimates of the health impacts attributable to exposure to air pollution indicate that PM_{2.5} concentrations in 2015 were responsible for approximately 422 000 premature deaths originating from long-term exposure in Europe (over 41 countries), from which approximately 391 000 were in the EU-28 (EEA, 2018).

Road transport is one of the main sectors that contributes to emissions of air pollutants in Europe. The issue of emissions from road transport (passenger and freight transport) is continuously aggravated as the number of vehicles and driven kilometers annually increase. The consequences are particularly severe in cities with a dense urban road network and related high traffic volume (Amato et al., 2014; Drliciak et al., 2020; Jandacka et al., 2017; Thorpe & Harrison, 2008). Vehicle motions cause mechanical wearing of the pavement surfaces by vehicle tires, which leads to gradual fragmentation and abrasion of the aggregates (Florkova & Jambor, 2017).

Considering several scientific papers, studies, and researches on PM production from road transport (Durcanska, 2010; Gehrig et al., 2010; Gustafsson et al., 2009; Haider et al., 2009; Kwak et al., 2013; Pio et al., 2013; Tervahattu et al., 2006), the present study focused on non-exhaust emissions, specifically PM from abrasion of pavement surfaces. Non-exhaust emissions of PM from road transport may originate from abrasion of vehicle parts (tires, brake linings, clutch, and bodywork), pavement surface abrasion, as well as resuspension of PM from surfaces (Pirjola et al., 2009). Non-exhaust particles are mainly abrasion products originating from the interaction processes between road surfaces and tires. Minor possible processes are brake and clutch abrasion as well as corrosion.

Identifying pavement surface abrasion as a PM source, separating it from other abrasions, and quantifying the corresponding amount of produced PM from field measurements at traffic sites is a very difficult process (Amato et al., 2014; Gehrig et al., 2010; Gustafsson et al., 2009; Haider et al., 2009; Pirjola et al., 2009). Specific differentiation between emissions due to abrasion and resuspension from road pavement is particularly difficult because of their similar elemental composition (Gehrig et al., 2010).
Foreign researches (Haider et al., 2009; Hussein et al., 2008; Kupiainen & Pirjola, 2011; Pirjola et al., 2009) have shown the influence of road pavement type, tire type (Gustafsson et al., 2008; Wik & Goran, 2009), and speed on the production of PM from road surface abrasion. This issue has been particularly addressed in countries such as Norway, Finland, and Sweden (Gustafsson et al., 2009; Hussein et al., 2008; Pirjola et al., 2009) because of the use of studded tires during winter. In some Norwegian cities, adverse health effects have led to the introduction of fees for drivers who use studded tires (Gustafsson et al., 2009). Road surface conditions and properties, such as pavement type (Haider et al., 2009) and year of construction, and meteorological factors including road surface moisture also affect emissions (Omstedt et al., 2005).

Thus, this study aimed to compare the various asphalt road pavements in terms of PM production. PM measurements were performed in laboratory conditions, in which captured particulates are not contaminated with exhaust emissions, abraded particles from vehicle parts, resuspension of road dust, and are not subjected to climatic effects (Haider et al., 2009). In addition, chemical composition and size distribution analyses were performed to obtain a better understanding of PM origin and behavior.

The study also aimed to confirm the abrasion of pavement surface as a source of PM and to verify the impact of the composition of asphalt mixtures on the PM production. Thus, a possible dependence between the composition of asphalt mixtures and measured PM mass concentrations was evaluated. Asphalt pavements are the most extensively used type of pavement. The findings from chemical analyses of basic materials and intercepted PM were used to verify the impact of asphalt mixture composition on PM production (Y. Yang et al., 2010). Each of the tested trial samples had a specific composition (type and amount of bituminous binder, type of aggregate, and different lines of aggregate granularity) (Amato et al., 2013).

2. Methodology of laboratory tests
Vehicle motions cause mechanical wearing of pavement surfaces by vehicle tires, which leads to gradual fragmentation, abrasion, and polishing of aggregates (Florkova & Jambor, 2017). The abrasion reduces aggregate angularity on the asphalt mixture surface and thus creates the condition for gradual polishing of used aggregate (Florkova & Pepucha, 2017). Asphalt pavements are the most extensively used type of road pavement. The most commonly used asphalt mixtures for wearing courses are asphalt concrete (AC) and stone mastic asphalt (SMA). The individual asphalt mixtures differ from each other by the relative proportion of the individual components, which affects the final mixture properties, appearance, and surface texture.

The measurements were performed in the laboratory at the Department of Highway Engineering of the University of Žilina. The used asphalt mixture samples (plates of size 320 × 260 mm and thicknesses of 40 mm) were tested in a wheel tracking machine DYNA-TRACK (Figure 1).

2.1. Elemental analysis of asphalt mixture materials
Before the asphalt mixture rutting, chemical composition tests of asphalt mixture materials (aggregates, bituminous binder) were performed by an X-ray fluorescence spectroscopy (XRF) analyzer. The machine SPEKTRO iQ II (AMATEK, Germany) was used for the experiments. From the analysis of individual materials, the percentage of the monitored elements was determined. The tests were performed in the laboratory at the Faculty of Civil Engineering of the Technical University of Košice. The composition of the rubber wheel used to drive on asphalt pavements was also determined.

2.2. Asphalt mixture rutting
The wheel tracking apparatus consists of a loaded wheel over a sample held on a moving table (Wheel tracking machine, 2000). The mixtures were rutted by a typified wheel for 12 h under steady working conditions. During one rutting of asphalt mixture samples, 20 000 cycles were performed (i.e., 40 000 running gears). The use of the wheel tracking machine at the laboratory
enables to sample abraded particles without contamination from surrounding sources and no influence from exhaust emissions (Haider et al., 2009). As temperature is an important factor influencing road pavement, the same initial temperatures were used to maintain the same conditions for each measurement.

Air sampling from the inside of the wheel tracking machine during the rutting of samples was performed using an aerodynamic particle sizer (APS) and a scanning mobility particle sizer (SMPS), which intercepted and distributed particles ranging from 0.012 to 20 µm along with three pieces of low-volume samplers Leckel LVS3 (Figure 1). The air around the sample was circulated (it was swirled by two fans, which were part of the device) to maintain stable temperature. During the asphalt mixture sample rutting, two fractions of PM2.5 and PM1 (inhalable particles, with diameters that are generally 1.0 µm and smaller) were captured on the cellulose nitrate (CN) membrane.
filters using Leckel LVS3 for the subsequent evaluation of mass concentrations and chemical analyses.

2.3. Elemental analysis of abraded PM
PM was captured on CN membrane filters for element determination. The filters were digested in Teflon vessels with HNO₃ at a temperature of 230°C using a high-temperature and high-pressure microwave digestion system SW-4 (Berghof, Germany). Samples were transferred and appropriately diluted on a triple quadrupole inductively coupled plasma mass spectrometer (ICP-MS—Agilent 8800 Triple Quadrupole ICP-MS, Japan) before element determination. The tests were performed in the laboratory of the Transport Research Centre in Brno, Czech Republic (Jandacka & Durcanska, 2019).

2.4. PM morphology
PM shapes and sizes were monitored using a VEGA TS 5136 LSU electron microscope (Tescan, Czech Republic). Polycarbonate membrane filters were used for PM sampling (Willis et al., 2002). The particles trapped on the filters were covered with a 20-nm layer of gold for better monitoring of shape and size (Licbinsky et al., 2010; Weinbruch & Ebert, 2004). PM imaging was performed in the laboratory of the Transport Research Centre in Brno, Czech Republic.

3. Results of experimental measurements
The laboratory test setup included 12 different pavement samples. Two plates of each asphalt mixture were available. Each was tested twice under the same conditions for 12 h. Thus, a total of

| No. | Asphalt mixture | Aggregate | Percentage share | Quarry | Asphalt binder | Indication of mixture |
|-----|-----------------|-----------|------------------|--------|----------------|----------------------|
| 1   | AC 11 O 50/70,II| siliceous limestone | 89.6 % | Tunežice | CA 50/70 | D—5 |
| 2   | AC 11 O 50/70,II| dolomite | 90.6 % | Biely Potok | CA 50/70 | R—9 |
| 3   | AC 11 O 50/70,II| melaphyre dolomite | 61.0 % | Solašnica Trstin | CA 50/70 | S—9 |
| 4   | AC 11 O PMB 45/80-75, I | melaphyre dolomite | 64.0 % | Malužín | Biely Potok | PMB 45/80-75 | R—8 |
| 5   | AC 11 O PMB 45/80-75, I | melaphyre dolomite | 46.0 % | Solašnica Trstin | PMB 45/80-75 | S—2 |
| 6   | AC 11 O 50/70,II | andesite dolomite | 46.6 % | Badín Biely Potok | CA 50/70 | K—4 |
| 7   | AC 11 O PMB 45/80-75, I | andesite dolomite limestone | – | Vtáčník Varín | PMB 45/80-75 | STR—1 |
| 8   | AC 11 O PMB 45/80-75, I | diorite porphyrite dolomite limestone | 46.0 % | Maglovec Olcnava | PMB 45/80-75 | C—5 |
| 9   | SMA 11 CRMB | spilite | 83.8 % | Litice | CRMB | GU—1 |
| 10  | SMA 11 PMB 45/80-75 | melaphyre dolomite | 71.0 % | Solašnica Trstin | PMB 45/80-75 | S—4 |
| 11  | SMA 11 PMB 45/80-75 | basaltic andesite limestone | 69.0 % | Večec Hostovce | PMB 45/80-75 | SKA—1 |
| 12  | SMA 11 PMB 45/80-75 | diorite porphyrite dolomite limestone | 62.5 % | Maglovec Olcnava | PMB 45/80-75 | C—4 |
48 measurements were performed (576 h of measurement). From the rutted asphalt mixtures, eight types of AC and four types of SMA were used. The main difference between the pavements was the percentage used stone material (Table 1).

4. Results of asphalt mixture elemental analysis

The chemical composition of asphalt mixture materials were determined by XRF. The tests were performed for all types of aggregate and asphalt binder used in the asphalt mixture samples. Aggregates from 15 quarries and two asphalt binders were used. The results are shown in Tables 2 and Tables 3.

Si and Ca represent the largest portions in the aggregates. In Table 2, the aggregates are divided into two groups, namely aggregates with Si predominance (mellaphyre, andesite, basaltic andesite, diorite porphyrite, and spilite) and aggregates with Ca predominance (dolomite, limestone, dolomitic limestone, and siliceous limestone). Elements such as Mg, Al, and Fe also represent a high share of identified elements. Dolomites and dolomitic limestones present high Mg concentrations, and high concentrations of Al and Fe are observed in aggregates with Si predominance. Bituminous binders contain approximately 4.8% to 10% of inorganic elements depending on the asphalt binder type. S represents the highest share of inorganic elements. Unmodified (CA 50/70) and polymer modified bituminous binders (PMB—45/80-75) contain 4.19% and 10.08% of S, respectively (Table 3).

5. Mass distribution and concentration of PM during asphalt mixture rutting

PM mass distribution and concentrations were determined using APS and SMPS devices and the low-volume sampler Leckel. The mass concentrations of two fractions of PM$_{2.5}$ and PM$_{1}$ were determined for each measurement by a reference gravimetric method.

To determine the mass distribution of PM by SMPS and APS, 180 air samplings were performed. The air sampling was performed every 4 min during 12 h of measurements. During each of the four measurements (for each asphalt mixture), 180 4-min air samplings were realized. Consequently, each mixture had 720 air samplings. All the acquired air samplings for the three selected rutted asphalt mixture samples are presented in Figures 2 and 3.

Figures 2 and Figures 3 represent the measured PM mass concentrations according to aerodynamic particle diameters for all air samplings. Therefore, they show a time course of PM mass concentrations for four measurements per asphalt mixture. Aerodynamic diameters with the highest mass concentration ranged from 0.15 to 0.5 µm (Figures 2 and Figures 3). The tendency of PM mass concentrations also indicates that no aerodynamic diameter higher than 2.13 µm was observed. Particulates of diameters lower than 2.5 µm were in majority during the air sampling. For this reason, two fractions of PM, PM$_{1}$ and PM$_{2.5}$, were measured during the rutting using low-volume samplers for subsequent chemical analysis and PM imaging.

To determine mass concentrations of PM$_{1}$ and PM$_{2.5}$ by the gravimetric method (Leckels), air samplings were performed for 12 h. For each tested mixture, the mass concentrations inside the wheel tracking machine were determined as the average value from 4 measurements for 2 plates. The results of the average PM mass concentrations of the two fractions for each tested asphalt mixture are shown in Table 4. On average, PM$_{1}$ fraction represented 65.70% of the PM$_{2.5}$ fraction.

The highest and lowest average PM$_{2.5}$ mass concentrations were measured for AC11 (S-9) and SMA11 (GU-1) mixtures, respectively. The mixtures differed in the use of aggregate and asphalt binder, and in their surface texture. In the mixture with the highest PM$_{2.5}$ concentration, mellaphyre (Si predominance) and dolomite (Ca predominance) were used, whereas in the mixture with the lowest PM$_{2.5}$ concentration, spilite was used (Si predominance).

In Table 4, the mixtures and their abrasion are divided into two groups, namely AC11 and SMA11. The highest and lowest average mass concentrations of abraded PM were measured for
Table 2. Chemical composition of aggregates used (significant values are in bold)

| Chemical composition of aggregates [mg/g] |
|------------------------------------------|
| Na | Mg | Al | Si | P | S | Cl | K | Ca | Ti | Mn | Fe | V | Cr | Cu | Zn | Sr | Ta |
|-----------------|-----|----|----|----|----|----|----|----|----|----|----|----|----|----|----|----|----|----|
| Melaphyre (Maluziná) | 0.00 | 36.20 | 75.98 | 238.10 | 0.86 | 0.15 | 0.27 | 10.34 | 72.26 | 8.48 | 1.97 | 57.47 | 0.30 | 0.05 | 0.05 | 0.11 | 0.14 | 0.37 |
| Melaphyre (Solašnica) | 0.00 | 62.52 | 78.74 | 236.50 | 1.51 | 0.13 | 0.35 | 9.23 | 71.30 | 8.90 | 2.05 | 74.66 | 0.32 | 0.12 | 0.04 | 0.23 | 0.19 | 0.44 |
| Melaphyre (Lošonec) | 0.00 | 70.27 | 63.59 | 189.70 | 1.48 | 0.11 | 0.31 | 6.90 | 88.02 | 7.11 | 1.81 | 59.44 | 0.32 | 0.14 | 0.00 | 0.15 | 0.18 | 0.00 |
| Andesite (Badín) | 0.00 | 18.83 | 103.10 | 281.20 | 0.77 | 0.09 | 0.28 | 10.20 | 63.52 | 4.76 | 0.98 | 40.68 | 0.23 | 0.00 | 0.03 | 0.04 | 0.23 | 0.00 |
| Andesite (Vtáčnik) | 37.30 | 9.37 | 118.00 | 280.60 | 0.77 | 0.23 | 0.00 | 12.50 | 63.70 | 2.37 | 0.36 | 26.90 | 0.08 | 0.00 | 0.00 | 0.00 | 0.30 | 0.00 |
| Basaltic Andesite (Vechec) | 7.35 | 12.21 | 57.10 | 199.20 | 0.40 | 0.07 | 0.30 | 9.89 | 46.67 | 3.73 | 0.95 | 36.25 | 0.14 | 0.06 | 0.01 | 0.03 | 0.23 | 0.00 |
| Diorite Porphyrite (Maglovec) | 7.45 | 24.45 | 85.33 | 264.50 | 0.67 | 0.06 | 0.08 | 13.99 | 44.33 | 4.17 | 0.92 | 48.27 | 0.00 | 0.00 | 0.00 | 0.00 | 0.00 | 0.00 |
| Spilite (Litice) | 0.00 | 73.53 | 79.91 | 186.70 | 1.93 | 1.30 | 0.07 | 5.32 | 136.70 | 8.99 | 2.27 | 71.13 | 0.49 | 0.45 | 0.08 | 0.00 | 0.26 | 0.18 |
| Dolomite (Trstín) | 0.00 | 160.80 | 3.00 | 7.63 | 0.10 | 0.19 | 0.42 | 0.00 | 259.10 | 0.19 | 0.32 | 1.89 | 0.09 | 0.29 | 0.00 | 0.00 | 0.09 | 0.00 |
| Dolomite (Biely Potok) | 0.00 | 175.60 | 9.52 | 204.1 | 0.23 | 0.10 | 0.52 | 0.00 | 304.20 | 0.47 | 0.40 | 3.53 | 0.11 | 0.39 | 0.00 | 0.00 | 0.10 | 0.00 |
| Limestone (Hostovce) | 0.00 | 2.65 | 0.41 | 1.14 | 0.00 | 0.03 | 0.00 | 0.00 | 447.90 | 0.01 | 0.30 | 0.33 | 0.00 | 0.00 | 0.00 | 0.00 | 0.00 | 0.00 |
| Limestone (Žírany) | 0.00 | 15.09 | 2.37 | 5.10 | 0.04 | 0.07 | 0.22 | 0.00 | 513.10 | 0.11 | 0.00 | 1.40 | 0.08 | 0.21 | 0.00 | 0.00 | 0.15 | 0.00 |

(Continued)
Table 2. (Continued)

| Chemical composition of aggregates [mg/g] |
|------------------------------------------|
| Dolomitic Limestone (Olcnava)            |
| 0.00 55.18 1.32 3.45 0.00 0.06 0.06 0.00 358.89 0.05 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 |
| Dolomitic Limestone (Varín)              |
| 26.50 143.40 36.00 21.80 0.00 2.17 0.45 0.89 316.70 0.21 0.00 2.55 0.00 0.00 0.00 0.00 0.46 0.00 |
| Siliceous Limestone (Tuženice)           |
| 0.00 46.85 5.45 26.22 0.25 0.18 0.38 1.12 482.90 0.29 0.35 3.48 0.12 0.36 0.00 0.00 0.70 0.00 |

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AC11 (S-9) (13.51 μg/m³) and SMA11 (GU-1) (5.49 μg/m³), respectively. The measured mass concentration of abraded particulates for the sample AC11 (S-9) was higher by 8.02 μg/m³ compared to the sample SMA11 (GU-1). This might be due to the asphalt mixture surface morphology and configuration of aggregates on the macro-texture level, which influences the contact area and the duration of the wheel-pavement interaction (Kotek and Kováč, 2015).

### 6. Results of PM elemental analysis

A total of 152 filters were analyzed by ICP-MS. The quantity of chemical elements (Ca, Si, Mg, Al, Fe, P, S, Cl, K, V, Cr, Mn, and Na) in the PM$_{2.5}$ was determined. The elements Ca and S were the most significant.

Si, characteristic of aggregates melaphyre, andesite, and porphyrite, was not significantly represented in the particulates, which is probably due to its properties, such as high hardness, which provides high resistance to abrasion (Table 5).

Ca represented the largest portion in the sample AC11 (D-5), with a concentration of 263.76 mg/g in the PM$_{2.5}$. In this asphalt mixture sample, aggregate siliceous limestone (Ca predominance) and unmodified bituminous binder CA 50/70 were used. Higher concentrations of Ca were observed for the sample AC11 (STR-1), with andesite and dolomitic limestone as aggregates and PMB 45/80–75, and also for the sample AC11 (C-5) with aggregates diorite porphyrite and dolomitic limestone and PMB 45/80–75. S showed the second largest portion of the analyzed PM, with concentrations ranging from 57.64 to 160.97 mg/g in the PM$_{2.5}$. With regard to S, the highest concentration was measured in the sample SMA11 (C-4), with a value of 160.97 mg/g in the PM$_{2.5}$. In this sample, diorite porphyrite and dolomitic limestone as aggregates and PMB 45/80-75 were used (Table 5).

Regarding the selected elements for analyses and subsequent confirmation of asphalt pavement abrasion, it is clear that all the elements occur in the captured PM. The chemical analyses indicated that the PM released from pavement surface abrasion during the rutting had a similar composition as that of the basic materials of tested asphalt mixtures (aggregates, bituminous binders). The representation of the analyzed elements was in the range from 28% to 60% of the total particulates mass. The highest chemical element share was analyzed for the sample AC11 (STR-1), representing 59.94% of the total particulate mass.

Table 5 shows that the highest mass concentrations in the abrasion from all the tested asphalt mixtures were observed for Ca. Ca concentration values ranged from 110.01 to 263.76 mg/g in the PM$_{2.5}$. The highest Ca concentration was obtained for the sample AC11 (D-5), in which siliceous limestone was used as the aggregate, which is more prone to abrasion. Si concentration values ranged from 6.47 to 24.92 mg/g in PM$_{2.5}$. Quartz-rich samples are more resistant to abrasion (Table 5).

### Table 3. Chemical composition of bituminous binders used (significant values are in bold)

| Chemical composition of asphalt binders [mg/g] |
|----------------------------------------------|
| Al | Si | P | S | Cl | K | Ca | V | Fe | Ni | Ag | Br |
|-----|----|---|---|----|---|----|---|----|----|----|----|
| CA 50/70 |    |    | 0.10 | 2.10 | 41.90 | 1.70 | 0.80 | 0.60 | 0.20 | 0.10 | 0.10 | 0.00 |
| PMB 45/80–75 | 0.10 | 2.40 | 0.00 | 100.80 | 0.70 | 0.30 | 0.80 | 0.20 | 0.10 | 0.10 | 0.00 |
|     | 0.10 |    |    |    |    |    |    |    |    |    |    |    |
Figure 2. PM mass distribution during sampling by SMPS (12 – 580 nm).
Figure 3. PM mass distribution during sampling by APS (0.5 – 20 μm).
### Table 4. Concentrations of measured PM during tests

| No. | Asphalt mixture | PM concentrations [μg/m³] |
|-----|-----------------|---------------------------|
|     |                 | PM$_{2.5}$     | PM$_{1}$     |
| 1   | AC 11 (D-5) siliceous limestone—CA 50/70 | 7.21          | 4.46          |
| 2   | AC 11 (R-9) dolomite—CA 50/70            | 9.04          | 7.55          |
| 3   | AC 11 (S-9) melaphyre, dolomite—CA 50/70 | 13.51         | 9.23          |
| 4   | AC 11 (R-8) melaphyre, dolomite—PMB 45/80-75 | 11.83         | 8.50          |
| 5   | AC 11 (S-2) melaphyre, dolomite—PMB 45/80-75 | 9.39          | 6.08          |
| 6   | AC 11 (K-4) andesite, dolomite—CA 50/70  | 7.99          | 4.22          |
| 7   | AC 11 (STR-1) andesite, dolomitic limestone—PMB 45/80-75 | 8.38          | 6.98          |
| 8   | AC 11 (C-5) diorite porphyrite, dolomitic limestone—PMB 45/80-75 | 8.16          | 6.31          |
| 9   | SMA 11 (GU-1) spilite—CRMB              | 5.49          | 4.73          |
| 10  | SMA 11 (S-4) melaphyre, dolomite—PMB 45/80-75 | 7.11          | 6.77          |
| 11  | SMA 11 (SKA-1) basaltic andesite, limestone—PMB 45/80-75 | 12.42         | 8.87          |
| 12  | SMA 11 (C-4) diorite porphyrite, dolomitic limestone—PMB 45/80-75 | 7.27          | 4.80          |

7. Dependence between the investigated elements of abrasion and tested asphalt mixtures

As a part of the research on abrasion of asphalt mixtures, the dependence between average mass concentrations of the examined 13 elements (Ca, Si, Mg, Al, Fe, P, S, Cl, K, V, Cr, Mn, Na) determined by chemical analyses in aggregates and asphalt binders and in the PM, representing surface abrasion of these asphalt mixtures was investigated. The dependence between element concentrations was expressed by a linear function.

The dependence was determined for the abrasion elements and elements in the individual materials of the tested mixtures. It means that the dependence was determined between aggregates and abrasion, bituminous binders and abrasion, and between the elements determined as a sum of aggregates and bituminous binders and abrasion. For the tested mixtures composed of two types of aggregates, the dependence between each aggregate and the abrasion was determined separately. The purpose of investigating these dependencies was to determine which materials of asphalt mixtures, whether aggregate or asphalt binder, produce abraded PM. The results of these comparisons are shown in Table 6 by means of the determination coefficient,
### Table 5. Chemical composition of PM$_{2.5}$

| Chemical composition of PM$_{2.5}$ [mg/g] | Ca  | Si  | Mg  | S    | Cl  | Fe  | Na  | K    | Al  | P   | V   | Cr  | Mn  |
|-----------------------------------------|-----|-----|-----|------|-----|-----|-----|------|-----|-----|-----|-----|-----|
| AC 11 (D-5)—siliceous limestone—CA 50/71 | 263.76 | 9.79 | 7.31 | 126.15 | 36.06 | 37.63 | 10.46 | 19.39 | 7.93 | 2.06 | 0.04 | 1.55 | 1.94 |
| AC 11 (R-9)—dolomite—CA 50/71          | 1324 | 6.75 | 4.01 | 6097  | 31.03 | 22.02 | 8.35 | 10.59 | 4.51 | 2.16 | 0.01 | 0.67 | 1.76 |
| AC 11 (S-9)—mélaphyre, dolomite—CA 50/71 | 124.66 | 6.47 | 3.39 | 104.84 | 36.54 | 11.04 | 3.07 | 10.34 | 0.76 | 0.73 | 0.03 | 0.67 | 0.3  |
| AC 11 (R-8)—mélaphyre, dolomite—PMB 45/80-76 | 126.3 | 14.01 | 3.43 | 57.64  | 12.28 | 12.77 | 3.39 | 0.46 | 0.02 | 0.49 | 0.71 |     |
| AC 11 (S-2)—mélaphyre, dolomite—PMB 45/80-76 | 141.64 | 16.66 | 4.59 | 155.62 | 21.85 | 17.59 | 8.55 | 9.29  | 0.57 | 0.03 | 0.09 | 0.65 |     |
| AC 11 (K-4)—andesite, dolomite—CA 50/71 | 135.03 | 7.69 | 6.08 | 93.6  | 21.53 | 12.47 | 6.93 | 17.75 | 1.41 | 1.73 | 0.01 | 1.22 | 0.56 |
| AC 11 (STR-1)—andesite, dolomitic limestone—PMB 45/80-76 | 254.41 | 20.96 | 11.64 | 133.11 | 46.66 | 80.83 | 10.16 | 12.02 | 18.71 | 3.94 | 0.05 | 0.22 | 6.66 |
| AC 11 (C-5)—diorite porphyrite, dolomitic limestone—PMB 45/80-76 | 215.18 | 24.92 | 5.89 | 98.82 | 27.37 | 28.21 | 16.35 | 12.37 | 7.44 | 0.72 | 0.04 | 0.14 | 1.08 |
| SMA 11 (GU-1)—spilite—CRMB               | 167.18 | 9.27 | 2.92 | 113.05 | 52.34 | 18.33 | 5.94 | 5.81  | 2.61 | 22.19 | 0.08 | 0.1  | 0.22 |
| SMA 11 (S-4)—mélaphyre, dolomite—PMB 45/80-76 | 110.01 | 17.67 | 6.77 | 113.5  | 27.91 | 26.89 | 14.35 | 17.65 | 7.38 | 1.78 | 0.14 | 1.7  | 2.09 |
| SMA 11 (SKA-1)—basaltic andesite, limestone—PMB 45/80-76 | 125.57 | 22.69 | 4.92 | 102.3  | 25.6  | 38.78 | 13.12 | 17.06 | 7.8  | 0.68 | 0.02 | 0.25 | 5.36 |
| SMA 11 (C-4)—diorite porphyrite, dolomitic limestone—PMB 45/80-76 | 161.69 | 17.57 | 5.95 | 160.97 | 31.61 | 29.62 | 18.43 | 25.82 | 8.74 | 3.39 | 0.05 | 0.24 | 1.62 |
Table 6. Mutual correlation between average mass concentrations of abrasion elements and materials of tested asphalt mixtures, numerically indicated by the correlation coefficient \((r)\) and coefficient of determination \((R^2)\)

| Asphalt mixture | Coefficient | Aggregate 1/ Abrasion | Aggregate 2/ Abrasion | Asphalt binder/ Abrasion | Aggregate +Asphalt binder/ Abrasion |
|-----------------|-------------|------------------------|------------------------|--------------------------|-----------------------------------|
| AC 11 (D-5)     | \(r\)       | 0.9792                 | -                      | 0.0566                   | 0.9884                            |
| - Ag. 1: siliceous limestone - CA 50/70 | \(R^2\) | 0.9589                 | -                      | 0.0032                   | 0.9771                            |
| AC 11 (R-9)     | \(r\)       | 0.8063                 | -                      | 0.1855                   | 0.8362                            |
| - Ag. 1: dolomite - CA 50/70 | \(R^2\) | 0.6501                 | -                      | 0.0344                   | 0.6993                            |
| AC 11 (S-9)     | \(r\)       | 0.1187                 | 0.8060                 | 0.0707                   | 0.6522                            |
| - Ag. 1: melaphyre - Ag. 2: dolomite - CA 50/70 | \(R^2\) | 0.0141                 | 0.6497                 | 0.0050                   | 0.4253                            |
| AC 11 (R-8)     | \(r\)       | 0.1421                 | 0.7443                 | 0.2114                   | 0.6879                            |
| - Ag. 1: melaphyre - Ag. 2: dolomite - PMB 45/80-75 | \(R^2\) | 0.0202                 | 0.5540                 | 0.0447                   | 0.4732                            |
| AC 11 (S-2)     | \(r\)       | 0.0927                 | 0.6517                 | 0.4949                   | 0.6424                            |
| - Ag. 1: melaphyre - Ag. 2: dolomite - PMB 45/80-75 | \(R^2\) | 0.0086                 | 0.4247                 | 0.2449                   | 0.4127                            |
| AC 11 (C-5)     | \(r\)       | 0.1039                 | 0.9576                 | 0.1034                   | 0.8528                            |
| - Ag. 1: dorate porphyrite - Ag. 2: dolomitic limestone - PMB 45/80-75 | \(R^2\) | 0.0108                 | 0.9170                 | 0.0107                   | 0.7273                            |
| AC 11 (K-4)     | \(r\)       | 0.1091                 | 0.8660                 | 0.0458                   | 0.6996                            |
| - Ag. 1: andesite - Ag. 2: dolomite - CA 50/70 | \(R^2\) | 0.0119                 | 0.7500                 | 0.0021                   | 0.4894                            |
### Table 6. (Continued)

| Asphalt mixture                        | Coefficient | Aggregate 1/ Abrasion | Aggregate 2/ Abrasion | Asphalt binder/ Abrasion | Aggregate +Asphalt binder/ Abrasion |
|----------------------------------------|-------------|------------------------|------------------------|---------------------------|-------------------------------------|
| **AC 11 (STR-1)**                      | r           | 0.0866                 | 0.7836                 | 0.2484                    | 0.5938                              |
| - Ag. 1: andesite                      | R²          | 0.0075                 | 0.6141                 | 0.0617                    | 0.3526                              |
| - Ag. 2: dolomitic limestone           |             |                        |                        |                           |                                     |
| - PMB 45/80-75                         |             |                        |                        |                           |                                     |
| **SMA 11 (S-6)**                       | r           | 0.2261                 | 0.7815                 | 0.1749                    | 0.7357                              |
| - Ag. 1: melaphyre                     | R²          | 0.0511                 | 0.6107                 | 0.0306                    | 0.5412                              |
| - Ag. 2: dolomite                      |             |                        |                        |                           |                                     |
| - PMB 45/80-75                         |             |                        |                        |                           |                                     |
| **SMA 11 (C-6)**                       | r           | 0.0100                 | 0.8193                 | 0.4332                    | 0.7487                              |
| - Ag. 1: diorite porphyrite            | R²          | 0.0001                 | 0.6712                 | 0.1877                    | 0.5606                              |
| - Ag. 2: dolomitic limestone           |             |                        |                        |                           |                                     |
| - PMB 45/80-75                         |             |                        |                        |                           |                                     |
| **SMA 11 (SKA-1)**                     | r           | 0.1277                 | 0.8103                 | 0.4581                    | 0.8594                              |
| - Ag. 1: basaltic andesite             | R²          | 0.0163                 | 0.6566                 | 0.2099                    | 0.7385                              |
| - Ag. 2: limestone                     |             |                        |                        |                           |                                     |
| - PMB 45/80-75                         |             |                        |                        |                           |                                     |
| **SMA 11 (GU-1)**                      | r           | 0.3947                 | -                      | -                         | 0.3947                              |
| - Ag. 1: spilite                       | R²          | 0.1558                 | -                      | -                         | 0.1558                              |
| - CRMB                                  |             |                        |                        |                           |                                     |

Note: Ag. 1—aggregates no. 1 in the tested asphalt mixture  
Ag. 2—aggregates no. 2 in the tested asphalt mixture
which indicates the degree of variability of a given dependency. It was assumed that during the rutting of tested mixtures, the asphalt layer and the aggregates were abraded and gradually polished, and concurrently particulates with their chemical composition were released into the environment.

By comparing the mass concentrations of the selected elements between the materials used in the asphalt mixtures and the captured abraded particulates, significant facts were found. The dependence between used aggregates and abraded particulates was confirmed. Aggregates with Si predominance (containing harder minerals) do not correlate with particulates determined as resulting from abrasion. The average correlation coefficient and coefficient of determination were $r = 0.14$ and $R^2 = 0.03$, respectively. This means that, on average, only 3% of the variability of abrasion element concentrations is explained by the element concentration of this type of aggregate. In contrast, for aggregates with Ca predominance (containing softer minerals), a high dependence between Ca concentration and abrasion has been confirmed. The average correlation coefficient was $r = 0.82$ and the average coefficient of determination was $R^2 = 0.68$. This means that, on average, 68% of the variability of abrasion element concentrations is explained by the concentration of this type of aggregates. The correlation coefficient values ranged from 0.65 to 0.98. Relevant dependence was also observed between the concentrations of elements represented as the sum of aggregates and asphalt binder in the mixture and the abrasion. The average correlation coefficient was $r = 0.72$, and the average coefficient of determination was $R^2 = 0.55$.

8. Origin of PM$_{2.5}$ in the components of the asphalt mixture

Multivariate statistical analyses using principal component analysis (PCA) and factor analysis (FA) were used for statistical assessment (Guo et al., 2004; Kachigan, 1991; Lu et al., 2010; Manly, 2004; Song et al., 2006; Spencer, 2013; Z. Yang et al., 2011).

FA is a multidimensional technique to investigate internal connections, relationships, and correlations and reveal the basic structure of the source data matrix. The basic task of FA is to determine the matrix of factor loads $A$ and the values of communities $h_i^2$, $i = 1, \ldots, m$. There are several methods to determine these values, from which the initial factor extraction was applied. After quantifying the first factor estimates, the factor matrix is rotated and the corresponding factors are quantified to improve the explanation of the original features. Many FA methods require the number of common factors $p$ to be known in advance. Therefore, it is appropriate to determine the number of factors $p$ using PCA, for example, using the graph of the base of eigenvalues. The combination of these two methods (PCA and FA) enables to achieve the minimum number of latent variables with the maximum original information, use the minimum number of latent variables for FA (factors), and insert the original characters (chemical elements).

The main goal of using FA was to reveal the potential distribution of PM sources (aggregates, asphalt). The source matrix contained the concentrations of the analyzed chemical elements found in the PM$_{2.5}$. These PMs were sampled when driving different asphalt mixtures (see above). The chemical elements were variables (13), and the measurements were objects (24) in the source matrix.

The variables (elements) with factor loadings $<0.7$ are highlighted. Thus, we obtained three factors with the dominant elements for the two types of aggregates (melaphyre, andesite—marker Si and dolomite, siliceous limestone—marker Ca) and asphalt (bituminous binder—marker S) (Table 7).

9. Morphology of abraded PM

Scanning electron microscope (SEM) can be used to determine the possible PM sources based on the size and shape of the particles. In addition to the physical properties of the particle (shape and size), it is appropriate to know their chemical composition. Combining these two information enables to predict the source of this particle. Sharp-edged particles were observed on contaminated filters in
numerous representations (Figure 4). These particles may be characterized as fragments of geological materials (aggregates) (Licbinsky et al., 2010; Weinbruch & Ebert, 2004; Willis et al., 2002; Zongping et al., 2003).

On the X-ray spectrum of the investigated particle, it is possible to see that the elements characterizing the materials of the asphalt mixture are Ca, Si, and S. The highest concentrations were observed for Ca and Si, which showed that soft and hard minerals are abraded, but soft ones are abraded on a larger scale. Higher concentrations of S indicate that the particles are likely to present asphalt binder residues even after grinding. From Figure 5a, 5b, 5c, 5d, different shapes of particles are observed, mostly sharp edges produced by mechanical abrasion.

### 10. Conclusions
The present study investigated and analyzed the formation of PM by asphalt mixture surface abrasion of various compositions. A methodology was developed to identify the origin of PM from various components of the asphalt mixture (aggregate, asphalt binder). This methodology

![Table 7. Factor loads of chemical elements to observed factors (significant values are in bold)](chart.png)
Figure 5a. Shape and analysis of trapped particle “A” during grinding of asphalt mixture STR-1.

Figure 5b. Shape and analysis of trapped particle “B” during grinding of asphalt mixture STR-1.
Figure 5c. Shape and analysis of trapped particle “C” during grinding of asphalt mixture STR-1.

Figure 5d. Shape and analysis of trapped particle “D” during grinding of asphalt mixture STR-1.
also proved to be a useful tool to determine the mass concentrations of PM from various asphalt mixtures and road surfaces. Thus, the resistance to abrasion of various asphalt mixtures was identified.

Various chemical elements were determined in asphalt mixtures. The most important were Ca, Si, and S. Ca and Si come from different types of aggregates, and S from the asphalt binder. These elements were also detected by ICP-MS in the sampled PM$_{2.5}$. The morphology of PM$_{2.5}$ was examined and the chemical compositions were analyzed using SEM. The investigated PM exhibited sharp edges and a chemical composition corresponding to the elemental composition of the individual components of the asphalt mixture (Ca, Si, S).

The concentrations of PM and chemical elements from the abrasion of the tested asphalt mixtures were determined, demonstrating the potential difference between the abrasion of the asphalt mixtures. The measurements showed the difference of produced PM between two types of asphalt mixtures, AC11 and SMA11. In particular, the difference between the produced PM and chemical elements depending on the aggregate used was determined. On average, asphalt mixtures with a combination of melaphyre and dolomite as aggregate showed the highest abrasion values. The relationship between the aggregate used in the test asphalt mixtures and the abraded particles was confirmed. Aggregates with a predominance of Ca (containing more soft minerals) exhibited a high correlation with the abraded particles. In contrast, aggregates with predominance of Si did not exhibit correlation with the abraded particles.

Multivariate statistical analysis revealed the differentiation of chemical elements according to their dominant representation in individual tested materials of the asphalt mixture. The origin of PM$_{2.5}$ was identified in these materials (aggregates, bituminous binder).

The choice of materials can influence the design of the road structure and the subsequent abrasion of the road surface by vehicle movements. By choosing a suitable type of asphalt mixture, we can reduce the abrasion of the road surface and the formation of PM containing hazardous chemical elements. Furthermore, the typical chemical fingerprint of road surfaces abrasion demonstrated by Ca, Si, and S was identified and can be used to identify sources of PM in the environment.

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