The road surface as a source of particulate matter

Daša Fullová¹, Dušan Jandačka¹, Daniela Žurčanská¹, Adriana Eštoková² and Jitka Hegrová³

¹University of Žilina, Faculty of Civil Engineering, Department of Highway Engineering, Univerzitná 8215/1, 010 26 Žilina, Slovakia
²Technical University of Kosice, Faculty of Civil Engineering, Institute of Environmental Engineering, Vysokoškolská 4, 042 00 Košice, Slovakia
³Transport Research Centre, Líšeňská 33a, 636 00 Brno, Czech Republic

E-mail: dasa.fullova@fstav.uniza.sk

Abstract. Traffic volume is still increasing and has unpleasant impact on the environment and longevity of the pavements. It is also associated with increase in emissions of particulate matter. Slovakia has built up the major part of roads with asphalt surface. Therefore, the research presented in this contribution deals with abrasion of bituminous wearing courses of pavements. The asphalt mixtures of wearing courses are compared in terms of released particulate matter. The samples of asphalt mixtures are abraded in wheel tracking machine DYNA-TRACK. The particulate matter measurements were performed in the laboratory conditions. The experimental laboratory measurements make it possible to sample particulates without contamination from exhaust emissions, abraded particles from vehicles, resuspension of road dust and climate affects. The contribution presents results of measured mass concentrations, chemical analyses of asphalt mixture materials and chemical analyses of particulate matter. The rutted asphalt samples are compared in terms of particulate matter mass concentrations and chemical composition. The contribution presents results of measurements on six trial samples of asphalt mixtures with different composition. The concentrations of metals are subjected to the multivariate statistical analysis (factor analysis) for the identification of sources of particulate matter (bituminous binder, aggregates).

1. Introduction and methodology of experimental measurements

Vehicle motions cause mechanical wearing of the asphalt pavement surface - wearing course by vehicle tires. That leads to gradual fragmentation and abrasion of aggregate. Traffic volume is still increasing and has unpleasant impact on longevity of the pavements and the environment. On the basis of several scientific papers, studies and researches about PM production from road traffic [1], [2], [3], [4] our research is focused on non-combustion emissions - particulate matter from abrasion of the pavement surface. It is very difficult process to identify pavement surface abrasion as a source of particulate matter and separate it from other abrasions and quantify its share of the amount of produced particulate matter in situ. Therefore, PM measurements are performed in laboratory conditions and captured particulates supposed to be without contamination from exhaust emissions, abraded particles from vehicles, resuspension of road dust and climate affects [5].

The aim of the research is to confirm the abrasion of pavement surface as a source of particulate matter and to verify the impact of the composition of asphalt mixture on the PM production. An
important phase of the research is to determine chemical elements in the collected particulate matter and to confront it with the representation of these elements in the chemical composition of the asphalt mixture. The research is aimed at finding dependence between the composition of asphalt mixture and collected PM.

The measurements are performed in the laboratory at the Department of Highway Engineering. The asphalt mixture samples of wearing courses are used for laboratory measurements. The used asphalt mixture samples – plates of size 320 x 260 mm and thicknesses of 40 mm are rutted in wheel tracking machine DYNA-TRACK which is used to assess the resistance to rutting of asphalt materials under conditions, which simulate the effect of traffic (Figure 1).

![Figure 1. The used machine technology of laboratory tests and rutted asphalt mixture sample.](image)

Air sampling from the inside of wheel tracking machine during the rutting of samples is performed by means of equipment APS 3321 (Aerodynamic Particle Sizer) and SMPS 3080 (Scanning Mobility Particle Sizer) which intercept and distribute particle range from 0.012 to 20 μm and 3 pieces of Leckel LVS3 which capture particulate matter PM$_{2.5}$ and PM$_{1}$. Chemical composition tests of materials (aggregates, bituminous binder) of samples were carried out by XRF analyser (X-ray fluorescence spectroscopy) before the rutting. For the experiments the machine SPEKTRO iQ II (AMATEK, Germany) was used. Particulate matter was captured on Nitrate membrane filters for subsequent element determination. The filters were digested in Teflon vessels with HNO$_3$ 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 before element determinations on a triple quadrupole ICPMS (ICPQQQMS, Agilent Technologies, Germany).

2. Results of particulate matter chemical composition

Measurements of asphalt mixture abrasion were carried out on 6 samples. Descriptions of rutted samples and mass concentrations of abraded particulate matter are shown in the following Table 1. The highest average PM mass concentration (13.51 μg/m$^3$) was measured for the mixture AC 11 with unmodified bitumen CA 50/70 (number 4) and the lowest average PM mass concentration (7.21 μg/m$^3$) was measured for the mixture AC 11 with unmodified bitumen CA 50/70 (number 1). The mixtures differ in use of aggregate. In the mixture with the highest concentrations the aggregates melaphyre and dolomite were used and in the mixture with the lowest concentrations the siliceous limestone was used.
The sampling of laboratory environment were carried out by means of Leckels before the rutting of samples and after these measurements in order to confirm the difference between the laboratory environment and the environment inside of wheel tracking machine. The difference between PM mass concentrations of laboratory environment and the lowest PM mass concentrations of asphalt mixture is determined at 3.23 µg/m$^3$ for particulates of fraction PM$_{2.5}$ and 1.42 µg/m$^3$ for particulates of fraction PM$_{1}$. 

Table 1. Description of rutted asphalt mixtures and average PM mass concentrations.

| Number | Asphalt mixture          | Bitumen   | Aggregate            | PM$_{1}$ [µg/m$^3$] | PM$_{2.5}$ [µg/m$^3$] |
|--------|--------------------------|-----------|----------------------|---------------------|-----------------------|
| 1.     | AC 11 O 50/70, II        | CA 50/70  | siliceous limestone  | 4.46                | 7.21                  |
| 2.     | AC 11 O 50/70, II        | CA 50/70  | dolomite             | 7.55                | 9.04                  |
| 3.     | AC 11 O 50/70, II        | CA 50/70  | dolomite, andesite   | 4.22                | 7.99                  |
| 4.     | AC 11 O 50/70, II        | CA 50/70  | dolomite, melaphyre  | 9.23                | 13.51                 |
| 5.     | AC 11 O PMB 45/80-75, I  | PMB 45/80-75 | dolomite, melaphyre | 8.50                | 11.83                 |
| 6.     | SMA 11 PMB 45/80-75      | PMB 45/80-75 | dolomite, melaphyre | 4.77                | 7.11                  |
|        | Laboratory environment   |           |                      | 3.04                | 3.98                  |

For identification of particulate matter originating from the asphalt mixture abrasion the chemical composition of basic materials of mixtures were carried out by XRF analyser. Unmodified bituminous binder CA 50/70 contains approximately 4.8 % of inorganic elements. Sulfur S has the largest representation, it represents 4.19 % (Figure 3). The chemical composition tests were also carried out for all kinds of aggregates used in tested samples. The results have shown that elements silicon Si and calcium Ca have the largest percentage in aggregates. The silicon Si predominates in aggregate melaphyre and andesite and the calcium Ca predominates in aggregate dolomite and siliceous limestone. The elements such as Mg, Al, Fe represent a higher share of identified elements (Figure 2). On the basis of analyses of aggregates and bituminous binder 13 chemical elements (Ca, Si, Mg, Al, Fe, P, S, Cl, K, V, Cr, Mn, and Na) were selected for confirmation of abrasion of asphalt mixture surface.

Figure 2. Percentage share of chemical elements of used aggregates in tested samples by XRF analyser.

Figure 3. Percentage share of elements of bituminous binder.

Subsequently, exposed filters (51 filters) were analysed for presence of selected elements by means of mass spectrometry with inductively coupled plasma. The Table 2 presents average mass
concentrations of selected 13 chemical elements in captured particulate matter PM$_{2.5}$ for each tested asphalt mixture. The elements Ca, S, Cl have the highest representation in the analysed filters. The highest concentrations of selected elements were analysed in the sample AC 11 (number 1), elements represent together 833.34 mg/g of PM$_{2.5}$. Calcium Ca has the highest representation in this sample and its concentration is 546.22 mg/g of PM$_{2.5}$. In this sample of asphalt mixture aggregate siliceous limestone is used. Sulphur with mass concentration 126.15 mg/g of PM$_{2.5}$ has also the highest concentration in this sample. With regard to the chlorine the highest concentration (81.47 mg/g of PM$_{2.5}$) was measured in the sample SMA 11 (number 6).

Table 2. Average mass concentrations of analysed elements in captured particulates PM$_{2.5}$

| Mass concentrations of analysed elements [mg/g] | Number of mixture/ | 1. AC 11 | 2. AC 11 | 3. AC 11 | 4. AC 11 | 5. AC 11 | 6. SMA 11 |
|-------------------------------------------------|-------------------|---------|---------|---------|---------|---------|---------|
| Ca                                               | 546.22            | 117.44  | 92.53   | 52.16   | 68.80   | 110.01  |
| Si                                               | 9.79              | 6.75    | 7.69    | 6.47    | 14.01   | 17.47   |
| Mg                                               | 7.31              | 4.01    | 6.08    | 3.39    | 3.43    | 6.77    |
| Al                                               | 7.93              | 4.51    | 1.41    | 0.76    | 3.39    | 7.23    |
| Fe                                               | 37.63             | 22.02   | 12.47   | 11.04   | 12.28   | 26.89   |
| P                                                | 2.06              | 2.16    | 1.73    | 0.73    | 0.46    | 1.78    |
| S                                                | 126.15            | 60.97   | 93.60   | 104.84  | 57.64   | 113.50  |
| Cl                                               | 62.87             | 43.94   | 68.02   | 34.84   | 30.86   | 81.47   |
| K                                                | 19.39             | 10.59   | 17.75   | 10.34   | 12.77   | 17.65   |
| V                                                | 0.04              | 0.01    | 0.01    | 0.03    | 0.02    | 0.14    |
| Cr                                               | 1.55              | 0.67    | 1.22    | 0.67    | 0.49    | 1.70    |
| Mn                                               | 1.94              | 1.76    | 0.56    | 0.30    | 0.71    | 2.09    |
| Na                                               | 10.46             | 8.35    | 6.93    | 3.07    | 6.91    | 14.35   |

On the other hand, the lowest concentrations were measured for the element vanadium V (in range from 10 to 140 µg/g of PM$_{2.5}$) and for elements such as Cr, Mn and P (Table 2).

3. Statistical analysis of measured chemical elements concentrations

Multivariate statistical analyses using Principal component analysis - PCA and Factor analysis – FA were used for statistical assessment [5], [6], [7], [8], [9], [10], [11], [12], [13], [14], [15], [16].

The main objective of using multivariate statistical analysis was to uncover the internal interconnection and the variability of the investigated variables (chemical elements - metals). Identified metals group may characterize some of the partial sources (materials) of asphalt mixtures.

The primary goal of PCA is the transformation of the original characters of $x_j$, $j=1, ..., m$, into a smaller amount of latent variables of $y_j$. These latent variables possess more appropriate properties: their presence is less significant, they capture and represent almost the entire variability of the original characters, properties and they are mutually not correlated – mutually uncorrelated. Latent variables are known as the main components and they represent linear combinations of former variables: the first main component $y_1$ describes the greatest part of variability, hence the dissipation, spread of the original data, the second main component $y_2$ on the other hand the greatest part of dissipation, spread not-contained within $y_1$ etc..

For the first main component the following relation prevails:

$$y_1 = \sum_{j=1}^{m} v_{1j} x_j$$ (1)
where: $x_j$ former character, input variable, $j = 1, \ldots, m$,
$v_{ij}$ coefficients of own vectors.

Within the process of FA so called factorial loads are estimated for particular variables (metals) for a generated factor. Factor loads, so to say the expression of correlations between the particular variables and acquired factors. Based on the values of factor loads it is possible to specify a group of variables for each factor, those ones which correlate with it in the closest-possible manner. And vice versa, by means of factor loads, the identified factor is appended with an extent of impact on each of particular variables. The variables with the highest factor loads for a generated factor are to be considered as decisive even when interpreting such a factor. A data matrix serves the purpose of input for calculations, whose lines correlate with particular measurements (objects) and bars of variables, i.e. measured pollutant (character). The variables to be used are those pollutants which are able to specify anticipated sources of pollution.

The fundamental principle of FA lies in the fact that each and every of monitored values $Y_j (j = 1, \ldots, p)$ may be expressed as a sum of a linear combination of a lesser amount $m$ non-observed (hypothetical) random values $F_1, \ldots, F_m$ – so called common factors and the further source of variability $E_j (j = 1, \ldots, p)$ – so called specific (residual) elements. Let us suppose that the following model prevails:

$$Y_j = \sum_{k=1}^{m} \lambda_{jk} F_k + E_j, j = 1, \ldots, p,$$

where: $\lambda_{jk}$ factor load of the $k$ common factor relevant to the $j$ value and of $k$ factor, $k = 1, \ldots, m$,
$F_k$ the $k$ common factor,
$E_j$ is a random deviation of the exact model, relevant to the one of $k$ value,
$j=1, \ldots, p$.

Values $Y_1, \ldots, Y_p$ are standardized, i.e. they have a zero median value and unit variance. In the FA method it is recommended to have at least 5 samples, while the optimum number of samples could reach 20 per each variable [8], [9].

The input data for the purposes of identification of possible PM sources consisted of the concentrations of particular chemical elements, specifically metals ($\mu g/g$). The data matrix consisted of 13 variables and 24 objects. Multivariate statistical analyses and source identification were processed for the fraction PM$_{2.5}$.

When it is deemed to be necessary to specify the number of factors before the FA calculation can be run, the PCA was performed beforehand. As a result, we could determine the number of principal components which, to a sufficient degree, account for the variability in the data. As is seen in Figure 4, we are able to fix the number of principal components at 4 (Eigenvalue $> 1$). The biplot shown in Figure 5 simplifies our decision-making process over the course of selecting the number of factors for use in FA, where close relationships among several variables can be observed. In this case, there are four collections of variables separated by a significant disparity. That is, there are 4 principal components which may be consecutively used in factor analysis.
Four factors were selected for factor analysis. These four principal components account for 81.9\% of the total variability in the original data.

The fourth factor was eliminated, which resulted from the consecutive FA using the four factors. The fourth factor was made up primarily of one element, chlorine “Cl”, and that is why it represented a specific factor which is at the same time difficult to define and interpret. Three factors account for 76.4\% of the total variability in the original data.

The Varimax model was used for factor rotation. The factor loadings associated with variables may be explained as the correlations between the factors and the variables. They represent the most important unit of information that the interpretation of the factors is based on. Each factor contains information about contributions of selected elements (metals). As the most decisive loading factors,
values close to or greater than 0.7 were selected. Based on the representation and presence of elements in particular factors, the following factors may be named (Figures 6, 7, 8). The factors for PM$_{2.5}$ (Figures 6, 7, 8) were identified based on the origins of metals (Figures 2, 3).

Moreover, the variables (elements) with factor loadings of <0.7 do contribute to a given factor by virtue of their weight and may facilitate its naming. Factors obtained via factor analysis of PM$_{2.5}$ were interpreted as Factor 1, representing aggregate (melaphyre, andesite) with marker silicon “Si” (Figure 6), Factor 2, representing aggregate (dolomite and siliceous limestone) with marker calcium “Ca” (Figure 7), and Factor 3, representing bituminous binder with marker sulphur “S” (Figure 8).

**4. Conclusion**

The highest average mass concentrations of PM$_{2.5}$ were measured for the mixture AC 11 with classic bituminous binder (with mark number 4) and the lowest average mass concentrations were measured for the sample AC 11 with classic bituminous binder (with mark number 1). The mixtures differ in use of aggregate. In the mixture with the highest concentrations the aggregates melaphyre and dolomite were used and in the mixture with the lowest concentrations the siliceous limestone was used.

From chemical analyses of filters the average mass concentrations of selected elements were determined. Calcium Ca has the highest representation in the sample AC 11 with mark number 1 and in this sample the aggregate siliceous limestone is used. The highest concentrations of selected elements were analysed in this sample. Despite of the fact that the sample has the lowest mass concentrations of particulate matter. As resulting, the released particulates from abrasion of mixture surface have similar composition as the basic materials of asphalt mixtures (aggregate, bituminous binder). The first results indicate that it has been found differences in the quality of asphalt mixtures in terms of PM production during the rutting of asphalt pavement surfaces.

Multivariate statistical analysis revealed three meaningful factors (sources of particulate matter) which are characterized by the chemical elements.

The identification of sources based on the presence of metals in the PM$_{2.5}$ fraction, were associated with particular elements: Factor 1, representing aggregate (melaphyre, andesite) (Si, V and Na), Factor
2, representing aggregate (dolomite and siliceous limestone) (Ca, Fe, P and Mn) and Factor 3, representing bituminous binder (S and Mg).

Acknowledgements
The paper originated as being supported by means of a grant KEGA 045ŽU-4/2017 Environmental education of engineers for application in public administration

References
[1] Kwak J, Kim H, Lee J and Lee S 2013 Characterization of non-exhaust coarse and fine particles from on-road driving and laboratory measurements Science of the Total Environment 458-460 pp 273-82
[2] Pio C, Mirante F, Oliveira C, Matos M, Caseiro A, Oliveira C, Querol X, Alves C, Martins N, Cerqueira M, Camoes F, Silva H and Plana F 2013 Size-segregated chemical composition of aerosol emissions in an urban road tunnel in Portugal Atmospheric Environment 71 pp 15-25
[3] Pirjola L, Kuptainen K J, Perhoniemi P, Tervahattu H and Vesala H 2009 Non-exhaust emission measurement system of the mobile laboratory SNIFFER Atmospheric Environment 43(31) pp 4703-13
[4] Ličbinský R, Frýbort A, Huzlík J, Adamec V, Effenberger K, Mikuška P, Vojtěšek M and Krůmal K 2010 Usage of scanning electron microscopy for particulate matter sources identification Transaction on Transport Sciences 3 pp 137-44
[5] Haider M, Folkeson L, Conter, Ramsak M, Cholava, Adamec V, Liebinsky R, Durcanska D and Decky M 2009 Guidelines for the environmental assessment of various pavement types including recommendations to road authorities in NMS, SPENS: Sustainable Pavements for European New Member States (European commission DG research, Sixth framework programme)
[6] Kachigan S K 1991 Multivariate Statistical Analysis: A Conceptual Introduction, 2nd Edition
[7] Manly B F J 2004 Multivariate Statistical Methods: A Primer, Third Edition (Boca Raton, USA: CRC press)
[8] Meloun M and Militký J 2006 Compendium of statistical data processing (in Czech) (Prague: Academia)
[9] Meloun M, Militký J and Hill M 2012 Statistical Analysis of Multidimensional Data in the Examples (in Czech) (Prague: Academia)
[10] Spencer N H 2013 Essentials of Multivariate Data Analysis (Boca Raton, USA: CRC press)
[11] Varmuza K and Filzmoser P 2009 Introduction to Multivariate Statistical Analysis in Chemometrics (Boca Raton, USA: CRC press)
[12] Lu X, Wang L, Li L Y, Lei K, Huang L and Kang D 2010 Multivariate statistical analysis of heavy metals in street dust of Baoji, NW China Journal of Hazardous Materials 173 pp 744-49
[13] Yang Z, Lu W, Long Y, Bao X and Yang Q 2011 Assessment of heavy metals contamination in urban topsoil from Changchun City, China Journal of Geochemical Exploration 108 pp 27-38
[14] Manta D S, Angelone M, Bellanca A, Neri R and Sprovieri M 2002 Heavy metals in urban soils: a case study from the city of Palermo (Sicily), Italy The Science of Total Environment 300 pp 229-43
[15] Guo H, Wang T and Louie P K K 2004 Source apportionment of ambient non-methane hydrocarbons in Hong Kong: Application of a principal component analysis/absolute principal component scores (PCA/APCS) receptor model Environmental Pollution 129 pp 489-98
[16] Song Y, Xie S, Zhang Y, Zeng L, Salmon L G and Zheng M 2006 Source apportionment of PM2.5 in Beijing using principal component analysis/absolute principal component scores and UNMIX Science of the Total Environment 372 pp 278-86