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**Road transport as the primary source of particulate
matter in the ambient air in urban and non-urban areas of
northern Slovakia**

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ABSTRACT

Emissions of particulate matter from different sources create a very complex mixture in the air both in qualitative and quantitative terms. Their composition is the result of distribution of all the sources in space and time, their size and characteristics of the pollutants on one side, and meteorological and climatic conditions on the other. Department of Highway Engineering of the Faculty of Civil Engineering at the University of Zilina deals with monitoring of particulate matter formation initiated by road transport. Long-term research is focused on the measurement of various fractions of particulate matter and determining quantity of heavy metals in the various fraction of particulate matter in the urban and non-urban areas in Zilina Selfgoverning region, Slovakia. The aim is to identify sources of particulate matter by means of statistical methods through representation of selected heavy metals in the particulate matter. The selected heavy metals can come from different sources. There are used heavy metals that are a part of road traffic and road surface during this research. The problem solving assumes the knowledge of multivariate statistical data analysis methods as for instance principal components analysis (PCA), factor analysis (FA) and multivariate regression and vector algebra.

Keywords: particulate matter, road traffic, air pollution, heavy metals, statistical methods.

1 INTRODUCTION

The pattern of particulate matter /PM/ behavior in the air is an interesting process, which is not entirely impacted on by its very sources. One of the primary sources of particulate matter to be considered is vehicular traffic which creates particulate matter within the urban or non-urban environment (Li et al., 2013, Pateraki et al., 2013). Vehicular traffic is also source of noise pollution, this is significant mainly for urban area and for homes close to roads with high traffic volume especially with a large proportion of lorries (Decky et al., 2012).

PM to be found in the atmosphere are exposed to various influences which may diminish their concentration or vice versa. They are in particular meteorological conditions which significantly impact on the concentration of particular matter in the atmosphere (Tecer, 2013, Tiwari et al., 2014).

Their harmful effect and impact on the ambient environment is determined predominantly by the presence of various chemical substances and elements and aerodynamic diameter of PM (Balachandran et al., 2000, Chen et al., 2010, Sysalová et al., 2012). The chemical composition of these particles (organic and elemental carbon, mineral dust, sea aerosols, secondary particles, especially sulphates and nitrates, heavy metals and further elements) is mainly impacted on by their origin, whereas the primary source of the particulate matter is determined and specified by the profile of chemical elements and substances. Heavy metals belong among the most basic groups of contaminants which are monitored in the various parts of the environment. We focused on the presence of heavy metals in particulate matter (Na,

Mg, Al, Ca, Cu, Sb, Ba, Pb, Cd, Cr, As, Mo, V, Mn, Fe, Ni, Zn) in this article. The selected heavy metals can come from different sources (Gatari et al., 2006, Kukutschova et al., 2011, McCullum and Kindzierski, 2001, Pant and Harrison, 2013, Sanderson et al., 2014, Thorpe and Harrison, 2008, Vojtesek et al., 2009, Weinbruch and Ebert, 2004)

Profile of heavy metals in particulate matter PM₁₀ was used to identify sources in urban and non-urban areas. The links between the different chemical elements (heavy metals) were identified by using multivariate statistical methods (principal components analysis (PCA), factor analysis (FA) and multivariate regression analysis (MRA)) (Chen et al., 2010, Manly, 2004, Manta et al., 2002, Varmuza and Filzmoser, 2009, Yang et al., 2011). Based on the presence of heavy metals in the resulting groups (factors), these groups were named as sources of PM₁₀. For the application of methodology suitable software may prove appropriate. Appropriate software – STATISTICA (StatSoft, Inc., 2011) and the method of PCA and FA are used to quantify the contributions of individual sources of air pollution to solid particles. Data matrix with rows corresponding to individual measurements (samples) and columns corresponding to variables (measured pollutants) serve as an input for calculations. Pollutants characterizing and defining the sources of pollution are selected as variables (Huzlik et al., 2011).

2 MONITORING AND CHEMICAL ANALYSIS OF PARTICULAR MATTER

Particulate matter (PM) monitoring was conducted in the urban area in the vicinity of an urban road in the City of Žilina during the years 2010 - 2012. 8 measuring cycles were realized during October 2010, March 2011, April 2011, July 2011, October 2011, January 2012, April 2012 and Jun 2012 on monitoring station “Urban area”. Total of 56 measurements were performed a on this monitoring station. A monitoring station in the City of Žilina was to be found at Vojtech Spanyol Street near the Regional Public Health Authority (RUVZ) (Figure 1). Measuring station “Urban area” is typically city canyon with residential development along the sides of the road.

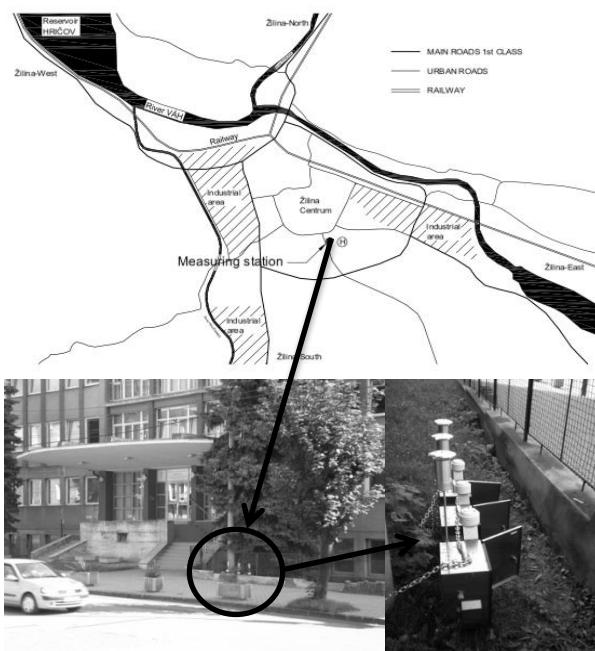


Figure 1: Urban Monitoring Station – urban road in the City of Žilina (RUVZ) (Jandacka and Durcanska, 2014)

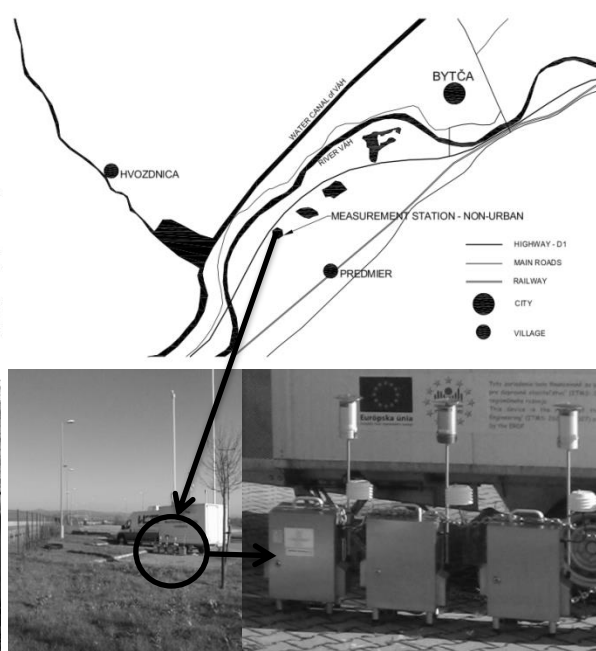


Figure 2: Non-urban Monitoring Station – highway D1 (SSUD) (Jandacka and Durcanska, 2014)

Second monitoring station was located in the non-urban area in the vicinity of a highway D1 where were realized measuring during the years 2013 - 2014. 3 measuring cycles were realized during May 2013, November 2013 and January 2014 on monitoring station “Non-urban area”. Total of 36 measurements were performed a on this monitoring station. Monitoring station “Non-urban“ was situated near the highway D1 in the areal of Centre Management and Maintenance of Highways (SSUD) (Figure 2). Surroundings of monitoring station “Non-urban area” consisted of an open area with agro land and water areas.

These measurements are a key part of an experiment which is aimed at the area where vehicular traffic may seriously impact on the presence and origination of particulate matter.

In order to determine the presence of particulate matter in the atmosphere a reference method pursuant to standards of STN EN 12341 and STN EN 14907 was used. In order to establish readings, low volume flow samplers of LECKEL LVS3 were used, amounting to the total number of 3 pieces. Simultaneously, there were three fractions of particulate matter /PM/ of PM_{10} , $PM_{2.5}$ and PM_1 monitored. Particulate matter /PM/ was trapped into nitrocellulose filters (47 mm in diameter) during the time 24 hours and consequently subjected to gravimetric analysis

The traffic load was monitored in a continuous manner by an automatic radar traffic detector of SIERZEGA SR4 suited for monitoring the traffic intensity. Simultaneously, the impacting meteorological conditions were monitored (temperature, realtive humidity, precipitation, wind speed and wind direction) by means of a weather station.

The particulate matter was to be bound with various elements and compounds. To test all the chemical components of the particulate matter concerned would be ineffective, quite demanding and financially unsustainable. In the first phase we focused on the monitoring of the selected heavy metals found in the fraction of **PM₁₀**. Each of these metals may come from a specific source (Table 1). Based on a sufficiently comprehensive database of data it seems possible by the utilization of multi-layer statistical methods (for instance factorial analysis), to more closely specify the possible source of this particulate matter.

Table 1. Sources of metals contained in the particulate matter - in general (Weinbruch and Ebert, 2004, McCullum and Kindzierski, 2001, Gatari et al., 2006, Vojtesek et al., 2009, Thorpe and Harrison, 2008, Pant and Harrison, 2013, Sanderson et al., 2014, Kukutschova et al., 2011)

Source	Associated elements
Transportation	
<i>road surface</i>	Al, Si, Ca, Mg, C, Na, K, V, Ni
<i>car-body components</i>	Cu, Sn, Cr, Pb, Cd, As, Sb, Fe, Al
<i>brake callipers, pads and rotors</i>	Cu, Sb, Ba, Cr, Fe, Ni, Pb, Zn
<i>tyres</i>	Zn, Cd, Pb, Cu, Ni, Fe, Mn, Cr, Co
<i>fuel and lubricating oil</i>	
	<i>diesel</i>
	Al, Ca, Mg, Mn, Cu, Fe, Mo, V, Zn
	<i>gasoline</i>
	Sr, Cu, Mn
	<i>oil</i>
	Fe, Ca, P, Zn, Mg
<i>catalytic converter</i>	Pt, Pa, Rh (Platinum metals)
<i>road dust</i>	Zn, Al, K, Fe, Na, Mn
Burning coal and wastes	Zn, Sb, Cu, Cd, Hg, Se, As, Cr, Co, Al
Industry	Sb, Ag, V, Ni, As, In, Cu, Mn, Ce, Co, Cr, Pb
Biomass burning	K
Incinerators	Cd, Pb, Sb, Zn

In order to identify, eventually to determine the present chemical form of an observed element in the sample of particulate matter the spectroscopic methods were utilized. The analyses of filters and the determination of metals present in the fraction of **PM₁₀** were performed pursuant to the standard of STN EN 14902.

Prior to specifying inorganic pollutants, the filters were reacted by the mixture of acids (HNO_3 and HF) and oxidizing agents with a resultant specification by means of the mass spectrometry method with inductively coupled plasma mass spectrometer ICP MS (Perkin - Elmer ELAN 6000 - USA) in cooperation with the Institute of Laboratory Research on Geomaterials at the Faculty of Natural Sciences of Comenius University in Bratislava.

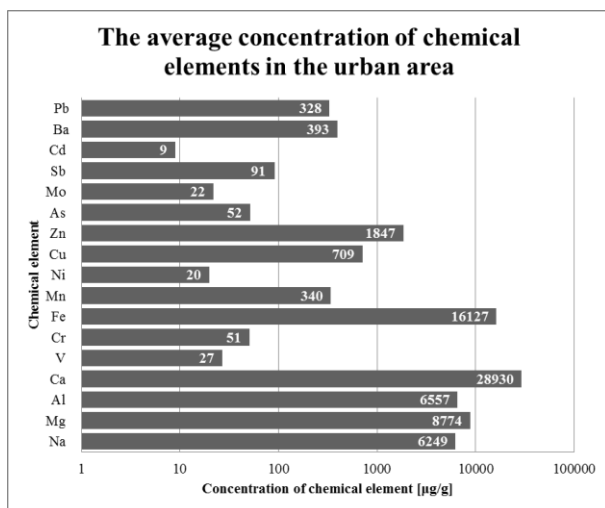


Figure 3: The average concentration of chemical element – urban area (volume of chemical element µg/g PM₁₀)

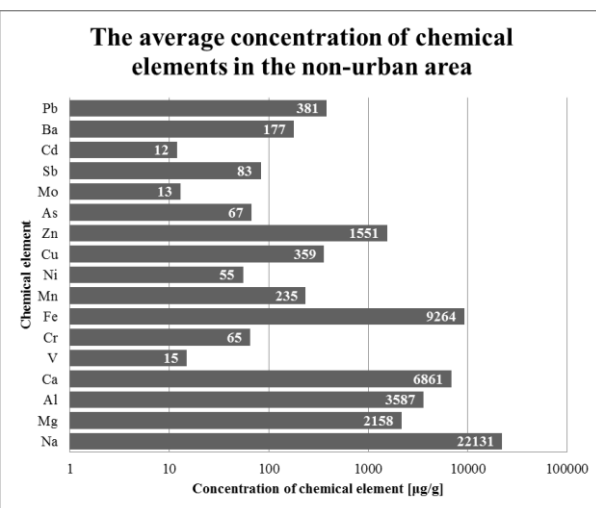


Figure 4: The average concentration of chemical element – non-urban area (volume of chemical element µg/g PM₁₀)

The main elements Fe, Ca, Al, Mg, Na, Ba, Cu, Ni, V shows significant difference between “Urban area” and “Non-urban area” in the number of elements in PM₁₀ (µg/g). Specifically elements - Fe, Ca, Al, Mg, Ba, Cu, Ni, V have substantially higher levels on the measuring station “Urban area” (Figure 3) compared to “Non-urban area”. One element - Na has substantially higher concentration on the measuring station “Non-urban area” (Figure 4) compared to “Urban area”. The average concentrations of PM₁₀ are also different on these two measuring stations. On the measuring station “Urban area” reached an average 54 µg/m³ and on “Non-urban area” 25 µg/m³. Measuring station “Urban area” is in the street canyon, where can be significant accumulation of pollutants.

3 USED STATISTICAL METHODS

Multi-layer statistical analyses of PCA – Principal component analysis and FA – Factor analysis were used for statistical assessment (Kachigan, S. K., 1991, Manly, 2004, Meloun and Militky, 2006, Meloun and Militky, 2012, Spencer, N. H., 2013, Varmuza and Filzmoser, 2009).

The primary goal of PCA is the transformation of the original characters of $x_j, j=1, \dots, m$, 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 principal components and they represent linear combinations of former variables: the first principal component y_1 describes the greatest part of variability, hence the dissipation, spread of the original data, the second principal component y_2 on the other hand the greatest part of dissipation, spread not-contained within y_1 etc. (Meloun and Militky, 2006, Meloun and Militky, 2012).

For the first main components the following relation prevails:

$$y_1 = \sum_{j=1}^m v_{1j}x_j \quad (1)$$

where: x_j former character, input variable, $j = 1, \dots, m$,
 v_{1j} coefficients of own vectors.

Within the process of factor analysis (FA) so called factor loads are estimated for particular variables (pollutants) 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 factor analysis lies in the fact that each and every of monitored values Y_j ($j = 1, \dots, p$) may be expressed as a sum of a linear combination of a lesser amount m non-observed (hypothetical) random values F_1, \dots, F_m – so called common factors and the further source of variability E_j ($j = 1, \dots, p$) – so called specific (residual) elements. Let us suppose that the following model prevails:

$$Y_j = \sum_{k=1}^m \lambda_{jk}F_k + E_j, j = 1, \dots, p, \quad (2)$$

where: λ_{jk} factor/factorial balance (load) of the k common factor relevant to the j value and of k factor, $k = 1, \dots, m$,
 F_k to the k common factor,
 E_j is a random deviation of the exact model, relevant to the one of k value, $j=1, \dots, p$.

Values Y_1, \dots, 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 (Meloun and Militky, 2006, Meloun and Militky, 2012).

4 STATISTICAL ANALYSIS OF MEASURED RESULTS

The basic difference between the FA and the PCA is that the FA explains the correlation between the characters and the PCA explains variability of characters. The method of principal components PCA is used when it is necessary to concentrate the maximum original information to the minimum number of latent variables (principal components) to achieve the best prediction. Factor analysis FA is primarily used to determine the original characters (heavy metals) shared in a common latent variable (factor). The combination of these two methods achieve the minimum number of latent variables with a maximum original information data - PCA and then use the minimum number of latent variables for factor analysis and put into them the original characters (heavy metals) - FA.

4.1 Urban area

Using a data matrix was compiled from the concentrations of selected metals in ng/m^3 (Na, Mg, Al, Ca, Cu, Sb, Ba, Pb, Cd, Cr, As, Mo, V, Mn, Fe, Ni, Zn) and PM_{10} in $\mu\text{g/m}^3$ resulting from 8 measurement cycles between the years of 2010 and 2012. The data matrix contained 18 variables and 56 objects.

Whereas at the factor analysis it is in the very beginning quite necessary to specify the number of factors and it is only then when the calculation may be run, during the first step the analysis of main components was performed. As a result of which we could conclude the possible number of principal components which to a sufficient measure specify the variance of dissipation, spread of characters. Pursuant to the rate of eigenvalues (**1 – 12.42, 2 – 1.94, 3 – 1.07**) there were **3** main components selected (Figure 5) (selection criteria of eigenvalue > 1.0). The three main components define **85.70 %** of the total dissipation, spread of the former characters.

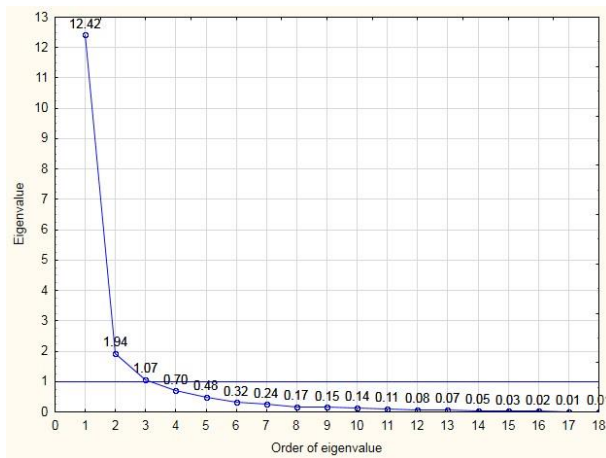


Figure 5: Graph of “foothills” Eigenvalues and Eigenvalues of Correlation Matrix
Chart – PCA – “Urban area”

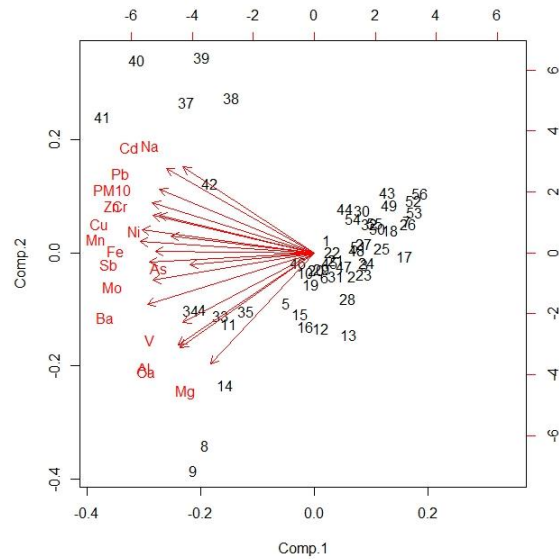


Figure 6: Principal Components Biplot (PM₁₀) – “Urban area”

The first principal component is very significant and characterizes substantial part of the scattering data (Figure 5).

Biplot (Figure 6, 11) combines information about the component weights (distribution of characters in the principal components) and component score (distribution measurements in the principal components). Axis corresponds to the first two principal components “comp. 1” and “comp. 2” in 2D graph (Figure 6, 11). Points indicate the objects and vectors are the projection of characters.

The angle between two vectors of characters x_j and x_k (heavy metals) (Figure 6, 11) is inversely proportional to the size of correlation between these two characters. The smaller angle = the greater correlation. If the angle between two vectors of characters is 90° these two characters are not correlated. Characters Na and Mg are the least correlated and characters Al and Ca are highly correlated (Figure 6). Each vector of character has coordinates on the first and second principal component. The size of each coordinate is proportional to the contribution of the original character x_j to the principal component that is proportional to the component weight. When the object (measurement) is located near the some character x_j in the biplot, it means that the object includes a large proportion of this character and is in the interaction with this character. All elements are highly interconnected.

3 factors were selected for the factor analysis. The used model was the rotation of Varimax factors. This rotation allowed the detailed distribution factors. Most of the elements were merged into the one factor without rotation. In the figures (Figure 7, 8, 9) the factor loads are quoted in relation to particular characters and particular factors. They may be explained as the correlation between the factors and characters. They represent the most important unit of information the interpretation of factors is based on.

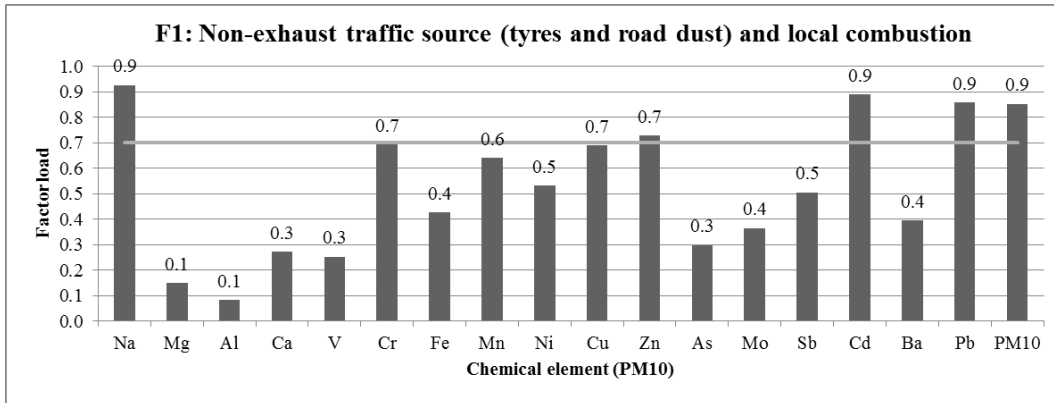


Figure 7: Factor loadings of characters (chemical element) to Factor 1 – F1 (PM₁₀) – “Urban area”

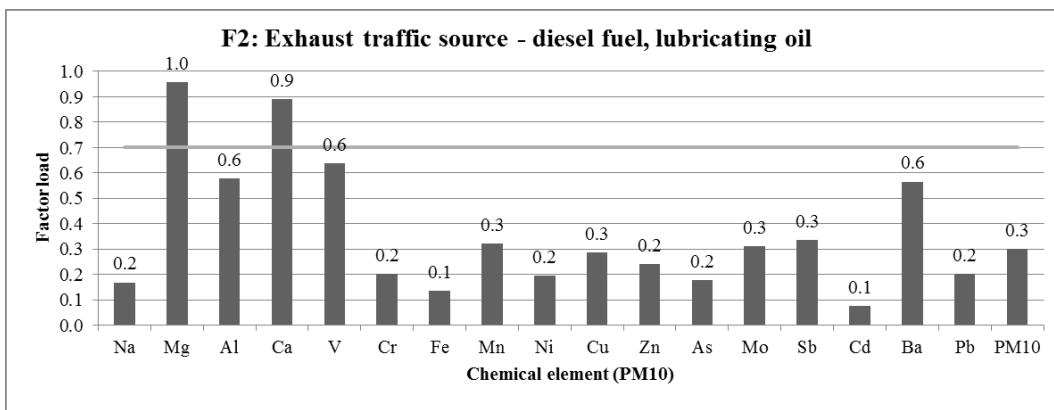


Figure 8: Factor loadings of characters (chemical element) to Factor 2 – F2 (PM₁₀) – “Urban area”

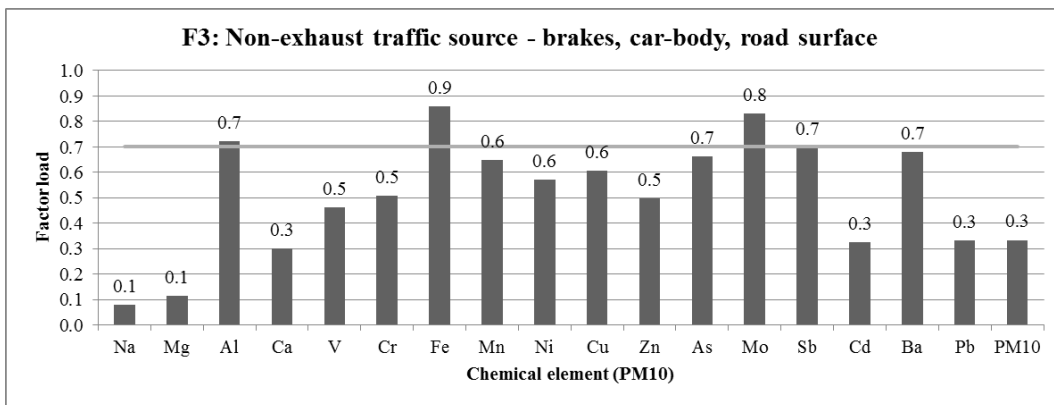


Figure 9: Factor loadings of characters (chemical element) to Factor 3 – F3 (PM₁₀) – “Urban area”

Each factor is contributed by several elements (characters). As the most decisive factor of loads the values close to or greater than 0.7 were selected. Based on the representation of elements in particular factors, the following factors may be named, designated (Figure 7, 8, 9).

4.2 Non-urban area

Using a data matrix was compiled from the concentrations of selected metals in ng/m^3 (Na, Mg, Al, Ca, Cu, Sb, Ba, Pb, Cd, As, Mo, V, Mn, Fe, Zn) and PM_{10} in $\mu\text{g}/\text{m}^3$ resulting from 3 measurement cycles between the years of 2013 and 2014. The data matrix contained 16 variables and 36 objects. The elements Cr and Ni were excluded from the analysis. These elements deformed structure of factors and did not correlate with other elements.

First, PCA was performed again. Pursuant to the rate of eigenvalues (**1 – 8.74, 2 – 2.01, 3 – 1.84**) there were **3** main components selected (Figure 10) (selection criteria eigenvalue > 1.0). The three main components define **78.71 %** of the total dissipation, spread of the former characters.

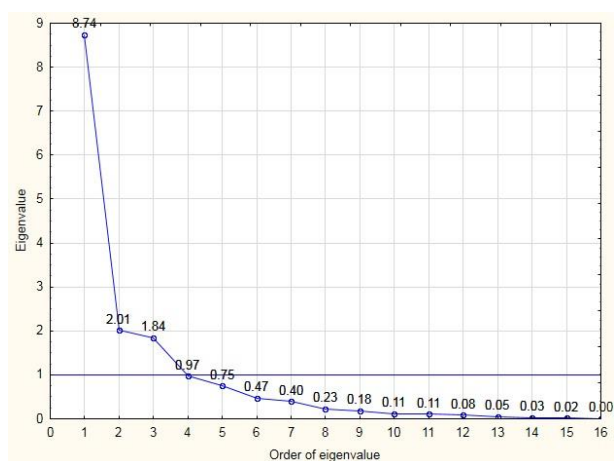


Figure 10: Graph of “foothills” eigenvalues and Eigenvalues of Correlation Matrix Chart – PCA – “Non-urban area”

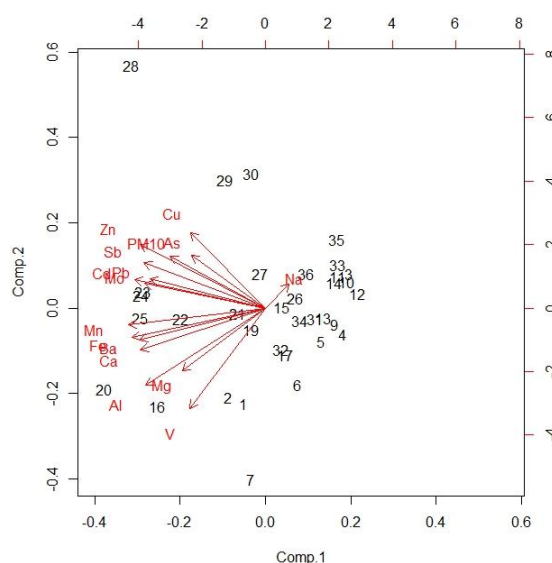


Figure 11: Principal Components Biplot (PM_{10}) – “Non-urban area”

From the analysis of data “Non-urban area” can be seen more visible distribution of some elements. Create clearer clumps. Also, we can see the links between certain elements with specific measurements. In particular element - Na is related mainly with measurements made in winter (measurements number 29 – 36). If the angle between two vectors of characters is 180° these two characters are correlated negative (for example Na and V). Characters Cu and V are the least correlated (Figure 11).

There were 3 factors selected for the factor analysis. The used model was the rotation of Varimax factors.

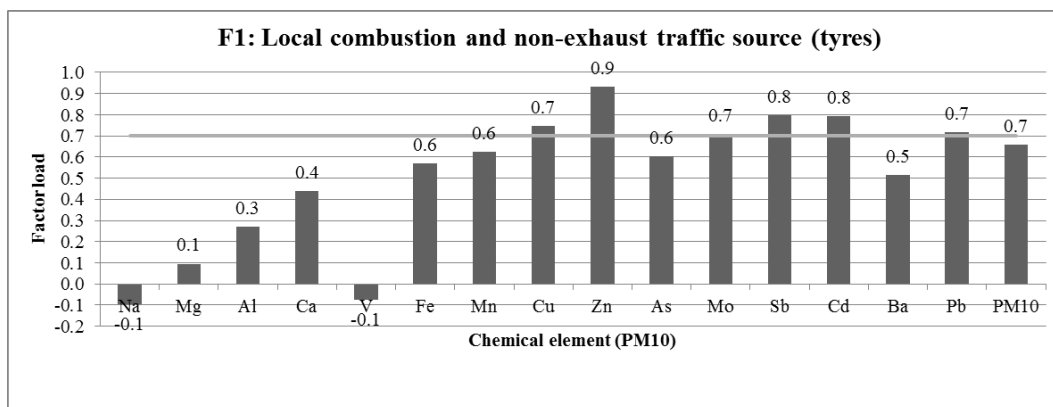


Figure 12: Factor loadings of characters (chemical element) to Factor 1 – F1 (PM₁₀) – “Non-urban area”

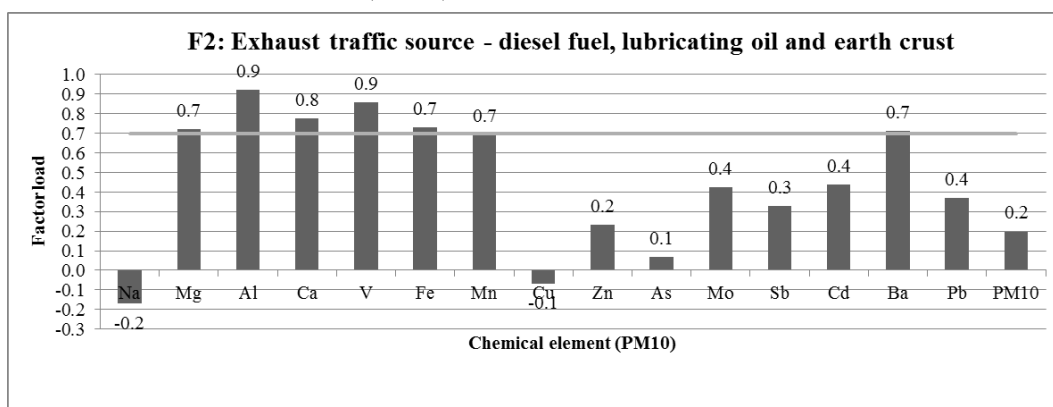


Figure 13: Factor loadings of characters (chemical element) to Factor 2 – F2 (PM₁₀) – “Non-urban area”

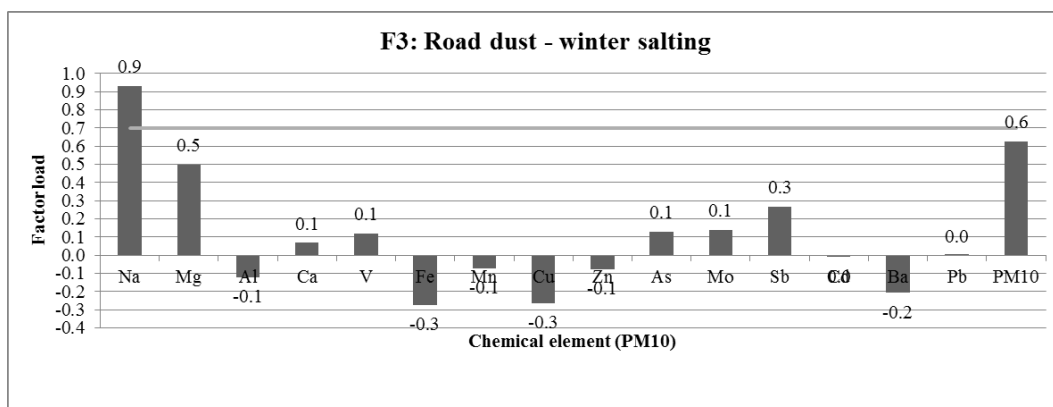


Figure 14: Factor loadings of characters (chemical element) to Factor 3 – F3 (PM₁₀) – “Non-urban area”

Individual elements are broken down into three factors after performing the FA. These factors were named on the basis of the elements involved (Figure 12, 13, 14).

The process of interpretation (named) of each factor is very difficult. Mainly because different sources of PM are mixed in the atmosphere and chemical elements can come from various sources. Interpretations of factors as sources of PM are therefore mainly based on the assessment area and the potential for some sources in this area. Named of each factor was also

confronted with the possibility of the contribution of sources during the year. It was also taken into account the variability of concentrations of chemical elements in different seasons.

5 CONCLUSIONS

Road transport is a significant contributor to air pollution, especially in the major cities. Road traffic has a role also in the air pollution in rural area. This contribution is devoted to the analysis of data about heavy metals in PM_{10} . The main factors were identified with typical chemical elements by representation of selected heavy metals in PM_{10} . Subsequently, sources of pollution were named on the basis of the distribution of chemical elements in each factor and knowledge of the likely sources of pollution in different areas. These were the two measuring stations "Urban area" – in city of Zilina, "Non-urban area" – near the highway D1. Measuring station "Urban area" is a typical urban street canyon. There is high traffic volume on this road in the city. Measuring station "Non-urban area" is an open area near highway D1. Multivariate statistical analysis showed that three factors are involved in the formation of PM_{10} on the measuring station "Urban area". These factors were named: Factor 1 - Non-exhaust traffic source (tires and road dust) and local combustion, Factor 2 - Exhaust traffic source - diesel fuel, lubricating oil, Factor 3 - Non-exhaust traffic source - brakes, car-body, road surface. These factors showed significant linkage with the elements: Factor 1 - Na, Cr, Cu, Zn, Cd, Pb, Mn and PM_{10} fraction, Factor 2 - Mg, Ca, W, Al, Factor 3 - Al, Fe, Mn, Ni, Cu, As, Mo, Sb, Ba. Three factors are involved in the formation of PM_{10} on the measuring station "Non-urban area": Factor 1 – Local combustion and non-exhaust traffic source (tyres), Factor 2 – Exhaust traffic source – diesel fuel, lubricating oil and earth crust, Factor 3 – Road dust – winter salting. These factors showed significant linkage with the elements: Factor 1 - Fe, Mn, Cu, Zn, As, Mo, Sb, Cd, Pb and PM_{10} fraction, Factor 2 - Mg, Ca, W, Al, Fe, Mn, Ba, Factor 3 - Na and PM_{10} fraction.

Road transport manifests as a major source of particulate matter on both stations. However, several sources of particulate matter can be combined in particular factors (Factor 1-3). For example, the winter local heating can contribute to the Factor 1 on monitoring station "Non-urban area", whereas there is the village near the D1 motorway. Therefore, some factors are named extensive and combine contribution of several possible sources.

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7 ACKNOWLEDGEMENTS



This contribution is the result of the project implementation: "Promotion & Enhancement of Transportation Research Centre" (ITMS: 26220220160) supported by the Research & Development Operational Programme funded by the ERDF.