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SOURCES OF PM_{10} AIR POLLUTION IN RURAL AREA IN THE VICINITY OF A HIGHWAY IN ŽILINA SELFGOVERNING REGION, SLOVAKIA

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Abstract

Particulate matter results as an aftermath of numerous distinctive processes in the atmosphere and they become a part of everyday life. Their harmful effect and impact on the ambient environment is determined predominantly by the presence of various chemical substances and elements. 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. Particulate Matter (PM) may originate in various natural resources or anthropogenic sources. Among the natural sources sea salt is to be counted on, dust of the earth crust, pollen and volcanic ashes. Anthropogenic sources do include, predominantly, burning fossil fuels in the fossil-fuel power plants, local heating of households, burning liquefied fossil fuels in the combustion engines of vehicles, non-combustion related emissions as a result of vehicular traffic, resuspension of the road-traffic-related dust.

Keywords:

Particulate matter;
Sources of air pollution;
Chemical element;
Heavy metals;
Vehicular traffic.

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 /PM/ within the urban or rural (non-urban) environment. 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 [3].

Particulates 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 [16, 18]. 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. 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 [1, 2]. 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 [4, 8, 11, 14, 15, 17, 19 and 20]. 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 [2, 10, 21]. For the application of methodology suitable software may prove appropriate. Appropriate software 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 [5].

2. Monitoring and chemical analysis of particular matter

Monitoring station was located in the non-urban area in the vicinity of a highway D1 where the measuring was realized during the years 2013 - 2014. There were realized 3 measuring cycles during May 2013, November 2013 and January 2014 on monitoring station “Non-urban area”. There were performed a total of 36 measurements 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) (Fig. 1). Surroundings of monitoring station “Non-urban area” consisted of an open area with agro land and water areas.

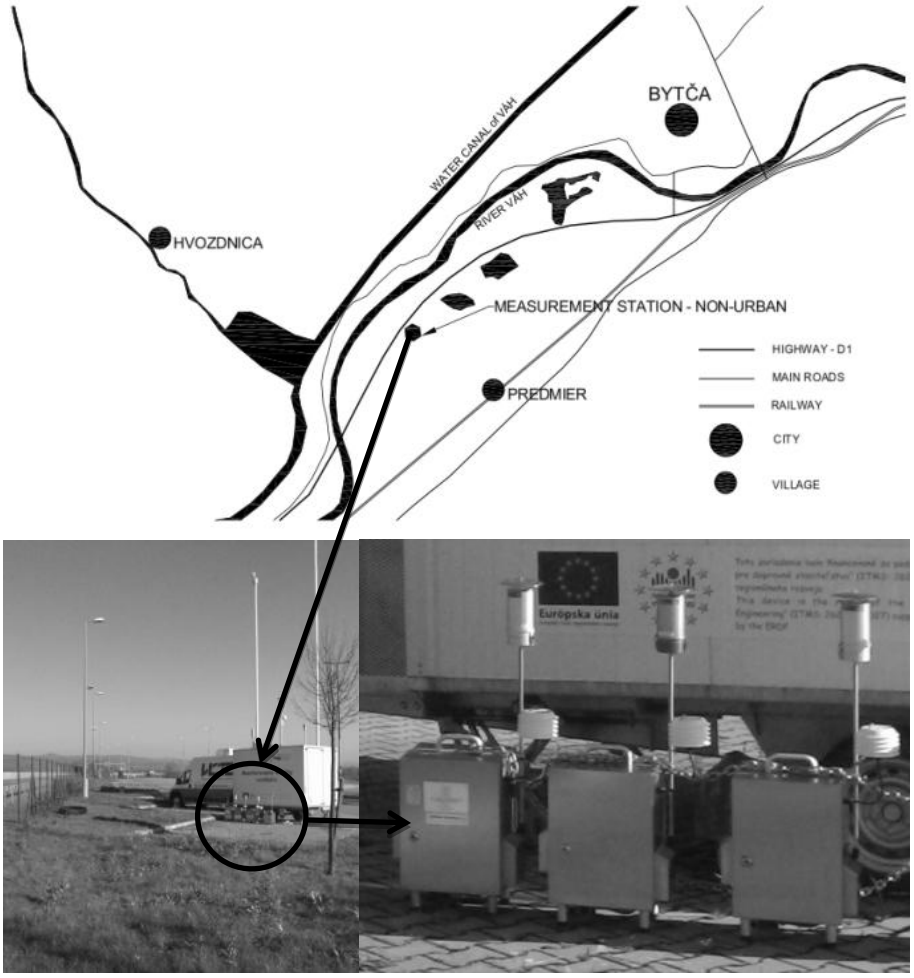

Fig. 1: Non-urban Monitoring Station – highway D1 (SSUD) [7].

These measurements are a key part of an experiment which is aimed at the area where vehicular traffic and road transport 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 (Fig. 2). Particulate matter /PM/ was trapped into nitrocellulose filters, the mesh of which was 47 mm during the time of 24 hours and consequently subjected to gravimetric analysis.

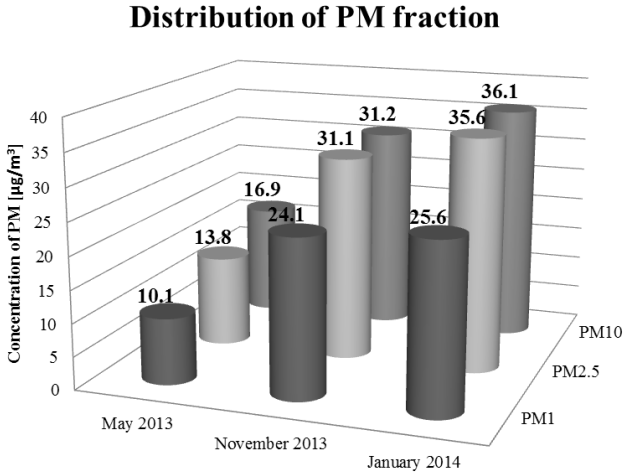

Fig. 2: The average concentrations of PM during different seasons – "Non-urban area".

Concentrations of particulate matter were different in view of the measuring season [6]. The average concentration of PM_{10} was 16.9 µg.m^{-3} during May, 2013 and 36.1 µg.m^{-3} during January, 2014 (Fig. 2).

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, relative 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 PM_{10}. Each of these metals may come from a specific source (Table 1). Based on a sufficiently comprehensive database of data it deems 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 [4, 8, 11, 14, 15, 17, 19, 20].

Source			**Associated elements**
Transportation	***road surface***		Al, Si, Ca, Mg, C, Na, K, V, Ni
	car-body components		Cu, Sn, Cr, Pb, Cd, As, Sb, Fe, Al
	brake callipers, pads and rotors		Cu, Sb, Ba, Cr, Fe, Ni, Pb, Zn
	tyres		Zn, Cd, Pb, Cu, Ni, Fe, Mn, Cr, Co
	fuel and lubricating oil	*diesel*	Al, Ca, Mg, Mn, Cu, Fe, Mo, V, Zn
		gasoline	Sr, Cu, Mn
		oil	Fe, Ca, P, Zn, Mg
	catalytic converter		Pt, Pa, Rh (Platinum metals)
	road dust		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 amount of chemical elements 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 PM_{10} 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.

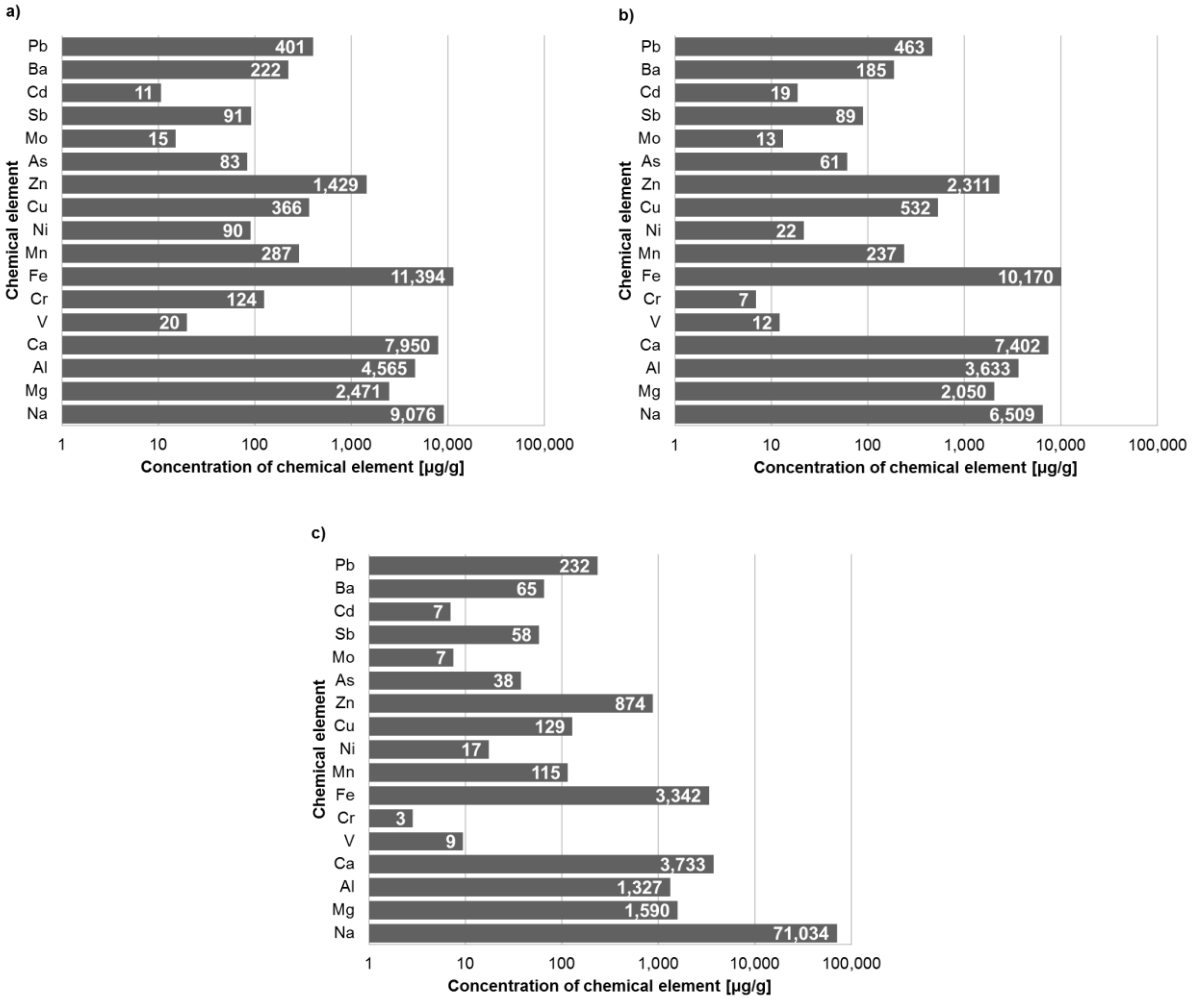

Fig. 3: The average concentration of chemical elements in the non-urban area during different measuring seasons - a) May 2013, b) November 2013 and c) January 2014 (volume of chemical element $\mu g.g^{-1}$ PM_{10}).

Representation of chemical elements in 1 g of particulate matter PM_{10} fraction has a different pattern considering measurement seasons. The concentration of most heavy metals shows higher values in May 2013 than in November 2013 and January 2014. Higher concentrations of PM_{10} were measured in January 2014, what could affect the decline in the share of metals on the PM_{10} fraction. Of the other part, this indicates no great change quantity of selected metals in the air. This suggests not much differentiation potential pollution sources producing these heavy metals during the year. There has been a significant increase of one metal (sodium Na) in January 2014 (the value of 71.034 $\mu g.g^{-1}$) compared to May 2013 (value of 9.076 $\mu g.g^{-1}$) (Fig. 3).

3. Statistical analysis of measured results

Multivariate statistical analyses of PCA – Principal component analysis and FA – Factor analysis were used for statistical assessment.

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 $\mu g.m^{-3}$ resulting from 3 measurement/reading 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.

3.1 Principal component analysis – PCA

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 y_j. These latent variables possess more appropriate and comprehensive properties: their presence is less significant, they capture and represent almost the entire variability of the original characteristics, properties and they are mutually uncorrelated. Latent variables are known as the main components and they represent linear combinations of former variables: the first principal component (PC) 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 variability, spread not-contained within y_1 etc.. Mathematically speaking, the first PC is viewed as a linear combination of input characters which covers the greatest variability, spread among the other linear combinations. In the matrix form we get the PCA model of the following string [2, 5, 10, 12, 13, 21]:

$$X = TP^T + E, \tag{1}$$

where: X - source matrix,
T - matrix of component score,
P^T - transposed matrix of component loadings,
E - matrix of residues.

Whereas at the factor/factorial 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 main components which to a sufficient measure specify the variance of dissipation, spread of characters. Pursuant to the rate of eigenvalues (1 – 8.74, 2 – 2.01, 3 – 1.84) there were 3 main components selected (Fig. 4) (selection criteria eigenvalue > 1.0). The three main components define 78.71 % of the total dissipation, spread of the former characters.

From the analysis of data “Non-urban area” can be seen visible distribution of some elements. Some of elements create clear 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 (Fig. 5).

3.2 Factor analysis – FA

Within the process of factor analysis (FA), so called factor loadings are estimated for particular variables (pollutants) for a generated factor. Factor loadings are the expression of correlations between the particular variables and acquired factors. Based on the values of factor loadings, 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 loadings, the identified factor is appended with an extent of impact on each of particular variables. The variables with the highest factor loadings for a generated factor are 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.

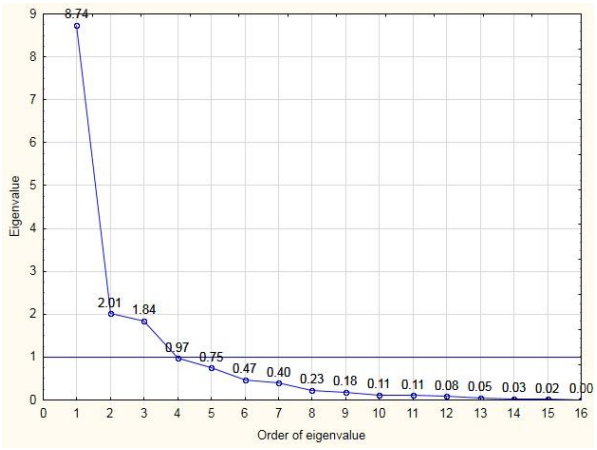

Fig. 4: Graph of "foothills" eigenvalues and Eigenvalues of Correlation Matrix Chart – PCA – "Non-urban area".

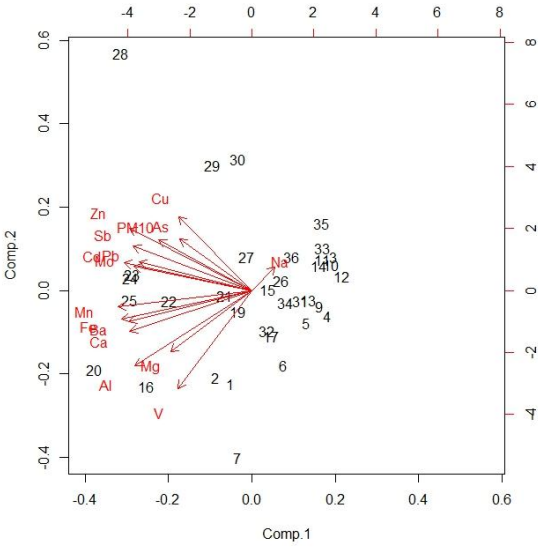

Fig. 5: Principal Components Biplot (PM_{10}) – "Non-urban area".

The basic principle of factor analysis lies in the fact that each of monitored values X_j *(j = 1, …,p)* may be expressed as a sum of a linear combination of a lesser amount *m* non-observed (hypothetical) random values $F_1, ..., F_m$ – so called common factors and the further source of variability E_j *(j = 1, …,p)* – so called specific (residual) elements. In the matrix form we get the FA model of the following string:

$$X = FA^T + E, \tag{2}$$

where: X - source matrix,
F - matrix of factor score,
A^T - transposed matrix of factor loadings,
E - matrix of residues.

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 [12, 13].

There were 3 factors selected for the factor/factorial analysis. The used model was the rotation of Varimax factors. This rotation allowed the detailed distribution factors. In the figures (Figs 6 - 8), 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.

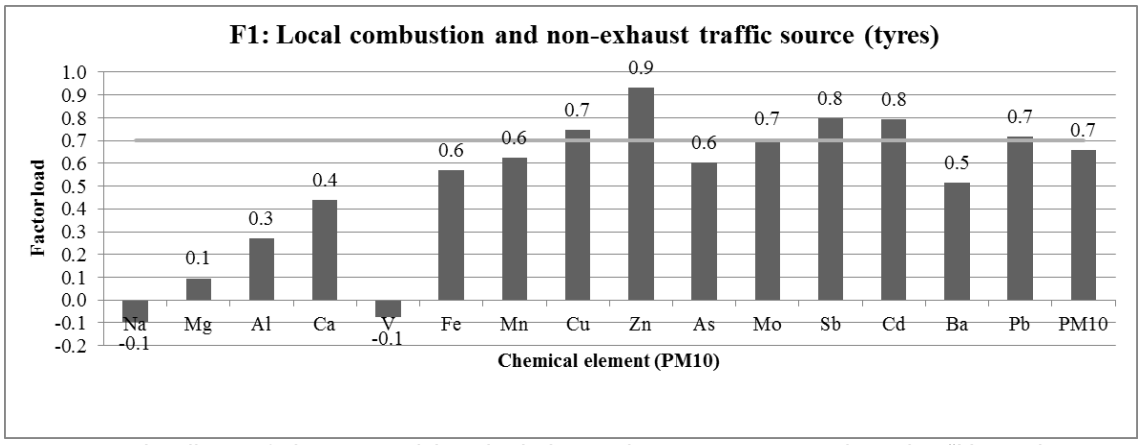

Fig. 6: Factor loadings of characters (chemical element) to Factor 1 – F1 (PM_{10}) – "Non-urban area".

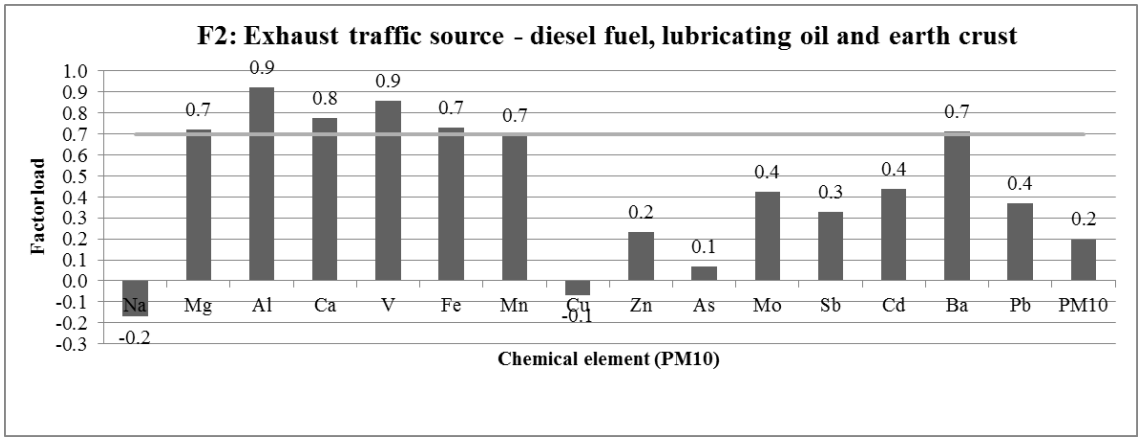

Fig. 7: Factor loadings of characters (chemical element) to Factor 2 – F2 (PM_{10}) – "Non-urban area".

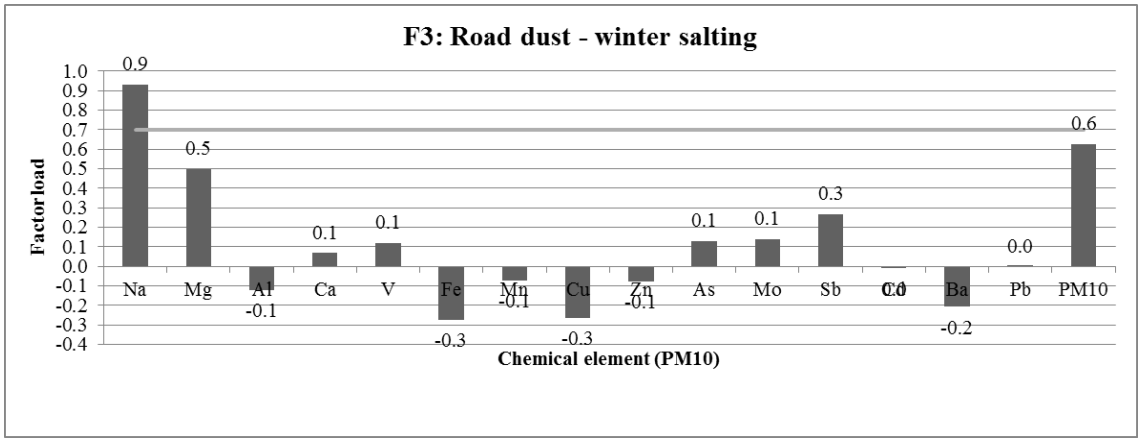

Fig. 8: Factor loadings of characters (chemical element) to Factor 3 – F3 (PM_{10}) – "Non-urban area.

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 (Figs 6 - 8).

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.

3.3 Absolute Principal Component Stores – APCS and Multiple Regression Analysis – MRA

There was used APCS and quantified the contribution of individual factors - pollution sources after defining factors using factor analysis and their interpretation.

First of all the values of factor scores are estimated. In our case, the weighted least squares method of Bartlett was used, which is based on the minimization of the mathematical expression:

$$(X - \Lambda \times F) \times \Psi^{-1} \times (X - \Lambda \times F) \quad (3)$$

where:

$$\Psi = \mathrm{var}(E), X = \begin{pmatrix} X_1 \\ . \\ . \\ . \\ X_P \end{pmatrix}, \Lambda = \begin{pmatrix} \lambda_{11} & \dots & \lambda_{1m} \\ \dots & \dots & \dots \\ \lambda_{p1} & \dots & \lambda_{pm} \end{pmatrix}, F = \begin{pmatrix} F_1 \\ . \\ . \\ . \\ F_m \end{pmatrix}, E = \begin{pmatrix} E_1 \\ . \\ . \\ . \\ E_P \end{pmatrix}, \quad (4)$$

Taking into account the fact of an unknown *F* factor, bearing in mind that instead of unknown loadings and specific variations shall be used their estimations. Henceforth, by means the weighted least squares method of Bartlett we achieve the estimate of the *i-th* line *F* as:

$$F_i = \left(\hat{\Lambda}' \times \hat{\Psi}^{-1} \times \hat{\Lambda}\right)^{-1} \times \hat{\Lambda}' \times \hat{\Psi}^{-1} \times X_i' . \quad (5)$$

As result of the fact that the *F* estimates follow the rotated solution, the *i-th* line estimate of the factor component score acquired as a result of multiplication $U.F^{-1}$ and the U matrix of the *m* order, meeting the condition of $U.U' = I$. In order to acquire to factor scores with the physics-oriented meaning, the APCS matrix is calculated, in the way that the reference value of F_0 is calculated:

$$F_0 = \left(\hat{\Lambda}' \times \hat{\Psi}^{-1} \times \hat{\Lambda}\right)^{-1} \times \hat{\Lambda}' \times \hat{\Psi}^{-1} \times X_0 , \quad (6)$$

where:

$$\left(X_0\right)_j = -\frac{\overline{x}_j}{s_j} , \quad (7)$$

$\overline{X}_j$ - sample average of i-value,
s_j - sample standard deviation of j-value,

which is subtracted from the calculated F matrix following the relation:

$$\mathbf{APCS = F - F_0.} \quad (8)$$

By means of utilization of Multiple Regression Analysis, where PM is a dependent variable and APCS are independent variables, one can ascertain the contributions of separately-identified sources. These are added up, while being summarized, as of the determined regression coefficients by means of their multiplication using the mean value of APCS for each common factor as an absolute contribution, which is out of the total concentration of PM re-calculated and transferred into the percentile contribution of given sources [5].

Results of APCS in conjunction with the MRA are summarized in the following table (Table 2).

Table 2: Results as of the multilayer regression analysis (PM_{10} is a dependant variable, independent variables absolute score for specific factors) for daily and nightly sampling.

Variable	Parameter	Std. Error	t-value	p	Significance of the variable
APCSFactor1	6.82	0.26	26.07	$<2e^{-16}$	***
APCSFactor2	3.73	0.17	22.05	$<2e^{-16}$	***
APCSFactor3	9.35	0.25	38.13	$<2e^{-16}$	***

All the variables (factors) are within the model to be understood as significant ones (***) and to a greater extent contribute to PM_{10} concentrations (Table 2).

Determination coefficient describes a share of the overall distribution, which may be clarified by means of a linear relation. In this case, it bears the value of R^2 = 0.99, which means that the linear relation clarifies a sufficient part of the overall variability.

The significance level is very small P < $2.2e^{-16}$, the adoption of a given model thence sufficiently significant.

Table 3: Absolute contributions of factors towards the mean concentration of PM_{10} [$\mu g.m^{-3}$].

	Factor1	Factor2	Factor3
APCS	6.03	8.44	11.30

There are absolute contributions and percentile contributions of factors towards the mean concentration of PM_{10} in the Table 3 and the Fig. 9. Average absolute contributions and average percentile contributions of factors relate to average concentration of PM_{10} 25.79 $\mu g.m^{-3}$ from all analysed data.

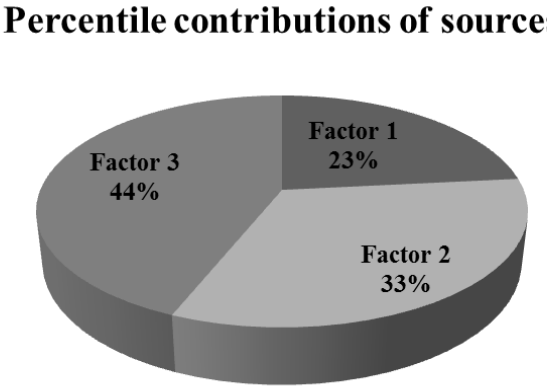

Fig. 9: Percentile contributions of factors towards the mean concentration of PM_{10} [%].

4. Conclusions

Measuring station “Non-urban area” is an open area near highway D1. Measurements of particulate matter PM10 were realized during three measurement cycles in May 2013, November 2013 and January 2014. A total of 37 24-hour particulate sampling were conducted. Samples of particulate matter PM10 were subjected to chemical analysis to determine the amount of 16 chemical elements - heavy metals: Na, Mg, Al, Ca, Cu, Sb, Ba, Pb, Cd, As, Mo, V, Mn, Fe, Zn, Cr, Ni. Concentrations of chemical elements – heavy metals were used for identify sources of PM_{10} by multivariate statistical analysis. Multivariate statistical analysis showed that 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, V, Al, Fe, Mn, Ba, Factor 3 - Na and PM_{10} fraction.

Identified sources of pollution contribute to the formation of PM_{10} differently according to the analysis APCS. Local combustion and non-exhaust traffic source (tyres) contributes to the PM_{10} 23%, Exhaust traffic source – diesel fuel, lubricating oil and earth crust 33% and Road dust - winter salting 44%. Road dust - winter salting contributes most to the formation of PM_{10} on this measuring station. Highways are only salted during winter and return resuspension of spreading materials contributes significantly to air pollution particulate matter.

The aim of paper was to clarify the variation of different sources of PM_{10} particulate matter air pollution in the vicinity of highway D1 in Žilina selfgoverning region, Slovakia.

Road transport manifests as a major source of particulate matter on this station. 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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