# URBAN AIR POLLUTION CAUSED BY THE EMISSION OF $PM_{10}$ FROM THE SMALL HOUSEHOLD DEVICES AND ABATEMENT MEASURES

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*This paper is focusing on Particulate Matter ($PM_{10}$) as the one of the main pollutants in the urban environments, and the one of the main sources of PM emissions are small household furnaces. This paper shows a part of the results of the research related to measurements of the ambient concentrations of aerosols and definition of the sources of the pollution in one part of Banja Luka city area, with dominant private households. Spatial and seasonal variability of concentrations indicates a significant contribution of biomass and fossil fuel burning as well as specific meteorological conditions. By using positive matrix factorization, (PMF) technique chemical composition data for $PM_{10}$ were analysed in order to identify the possible sources and estimate their contribution to particulate matter mass. A biomass combustion devices were identified as the one of dominant $PM_{10}$ emission sources during the winter season, because highly determined correlation between $PM_{10}$ and potassium content. From the other side, the results of the project "Typology of Residential Buildings in Bosnia and Herzegovina", were used as a tool for prediction and quantification of influence of the household's appliances on the pollution as well as for definition of the possible measures for reduction for this specific location. This approach took in consideration spatial distribution of buildings, but also their type, which can give the opportunity to estimate their present energy needs and predict improvements, which can lead to reduction of the $PM_{10}$ pollution at the end. On that base, some recommendations for the improvements were analysed.*

Key words*: combustion, pollution, emission, $PM_{10}$*

## 1. Introduction

Urbanization, industrialization and economic growth resulted in a profound deterioration of urban air quality [1]. Due to high density of the pollution sources, urban areas are mainly affected by suspended particles, which pose a serious risk to human health, ecosystem damage and degraded visibility [2]. Health effect associated with PM are linked to respiratory, cardiovascular health

problems, and premature mortality [3,4]. PM is composed of a broad class of chemically and physically diverse substances, such as trace metals, ions, BC, PAHs, especially BaP, which is well-known human carcinogen. Some studies presented correlation between the $PM_{10}$ concentrations in the ambient air and the death rate, it was found that long term exposure to fine particulate matter from combustion increasing the risk of cardiopulmonary mortality of 6% and lung cancer mortality around 8% [5].

Trace metal pollution in the atmosphere is mainly related to the inorganic fraction, which is released from various kinds of vehicles, brickfields, constructions, tanneries, navigation, corrosion of metallic parts, soil dust, etc. [6]. Measurements of mass concentration of $PM_{10}$ in urban area of Banja Luka were carried out at four sites, but elemental composition of $PM_{10}$ is still limited. Elemental composition of PM is very important for application of multivariate statistical methods addressed to the identification and quantitative apportionment of air pollutants to their sources. The study was performed at suburban site, Lazarevo, Banja Luka, which is official location site for air quality monitoring network in the Republic of Srpska. This site was selected because covers different sources of pollution including, road traffic, industries, re–suspended dust due to construction, commercial and residential burning of fossil duels and biomass (wood). The important issue is that Lazarevo is suburb area of Banja Luka, with majority of individual family houses, and without centralized system of the heat supply. According to the actual Regulation Plan of Banja Luka City, this is populated area with approximately 10.000 inhabitants, and average population density of 71 inhabitant/ha. Number of the buildings on the location is 1.752, mainly individual houses. Due to that small individual furnaces (heating devices) mainly based on biomass combustion in the households presents high risk for the Lazarevo's population health. The air quality sampling was carried in two seasons, winter and summer, in 2010/2011, and 2013. The aim of this study was to investigate the complexity of urban aerosol and its different pollutant sources my means of $PM_{10}$ chemical characterization concerning ions and trace elements, thus improving local knowledge on the contribution of various species to $PM_{10}$ in the Banja Luka. The objective of this study was to investigate temporal variations in mass concentration of $PM_{10}$, ions and trace metals components in $PM_{10}$ during different seasons collected from the suburban area in Banja Luka. Also, this study aimed to resolve the identities and contributions of different components in $PM_{10}$ using receptor modelling with Positive Matrix Factorization. Thus, this paper provides a framework to determine emission sources, which requires emissions reductions measures introduction, in order to improve air quality in populated area with variety of emissions.

## 2. Sampling and Chemical Analysis

Air quality monitoring was carried out at suburban site Lazarevo, location with different commercial and residential buildings, individual heating, frequent road traffic and industry. The sampler was installed on the green surface, 2 m above the ground; 100 m distance from very frequent road and approximately 1 km from industrial zone with wood processing, smelting and food and drink industry. Fig.1. provides the location of sampling site.

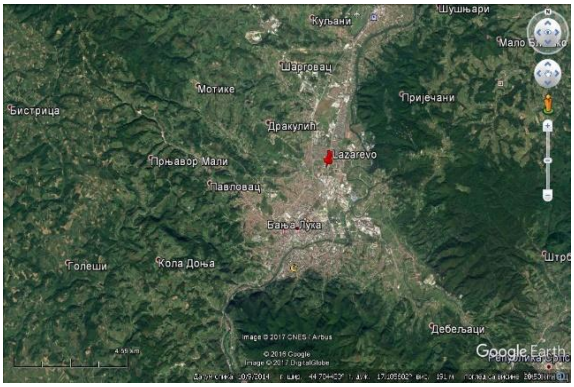



**Figure 1. Location of sampling site**

Sampling was carried continuously for 20 days in summer and winter season, in 2010/2011, and 20 days in summer and winter season in 2013. in order to represent seasonality at site. Ambient concentration data used in the source-receptor modelling include 24h mass concentrations of suspended particles, atmospheric aerosols $PM_{10}$ and their chemical composition (Pb, Cu, Zn, Fe, Mn, Co, Ni, Al, As, Se, Sb, Hg, Ti, V, $Ca^{2+}$, $Mg^{2+}$, $K^{+}$, $Na^{+}$, $NH_4^{+}$, $Cl^{-}$, $SO_4^{2-}$, $NO_3$). Samples were collected in „Summer campaign“ from 6$^{th}$ to 28$^{th}$ Aug 2010 and 10$^{th}$ to 30$^{th}$ July 2013 and „Winter Campaign“ from 2$^{nd}$ to 21$^{st}$ Jan 2011, and 11$^{th}$ to 31$^{th}$ Jan 2013. Ambient air sampling of $PM_{10}$ was carried using MVS6 Sven Leckel air sampler provided with $PM_{10}$ cut off inlet at 2m height according to EN 12341 “Air quality - Determination of the $PM_{10}$ fraction of suspended particulate matter - Reference method and field test procedure to demonstrate reference equivalence of measurement methods”. Samples were collected in preconditioned and pre-weighed Quartz filters (Whatman, 47 mm diameter, 2 μm pore size). Weighing was carried out on a semi-micro balance (OHAUS ADVENTURER AR 2140, max 210 g), with a minimum resolution of 0.01 mg. Loaded and unloaded filters (stored in Petri dishes) were weighed after 48 hours conditioning in a desiccator in the clean room at a relative humidity of (45–55)% and a temperature of (20±2)$^{\circ}$C. Quality assurance was provided by simultaneous measurements of a set of three ‘‘weigh blank’’ filters that were used to correct the sample filter mass changes. Trace elements were estimated from $PM_{10}$ samples collected on Quartz filters using the Atomic Absorption Spectroscopy method (AAS) with Varian Cary 50 Bio. For water-soluble inorganic ion analysis, $PM_{10}$ samples collected on Quartz filters were subjected by ultra–pure water and analysed by Ion Chromatography DIONEX Corporation US. Number of species have very low concentrations, below detection limit of analytical techniques (Al=6.9767 ngcm$^{-2}$,As= 2.3256 ngcm$^{-2}$, Ca=23.2558 ngcm$^{-2}$, Cd=0.2326 ngcm$^{-2}$, Cu=2.3256 ngcm$^{-2}$, Fe=69.7674 ngcm$^{-2}$, Hg=0.4651 ngcm$^{-2}$, K=23.2558 ngcm$^{-2}$, Mg=2.3256 ngcm$^{-2}$, Mn=0.4651 ngcm$^{-2}$, Mo=2.3256 ngcm$^{-2}$, Na=4.6512 ngcm$^{-2}$, Ni=2.3256. ngcm$^{-2}$, Pb=2.3256 ngcm$^{-2}$, Sb=0.9767 ngcm$^{-2}$, Se= 4.6512 ngcm$^{-2}$, Si=4.6512 ngcm$^{-2}$, V=9.3023 ngcm$^{-2}$, Zn=11.6279 ngcm$^{-2}$). The obtained recovery in two runs were within 15% of the certified values for the Cu, Pb, Zn, Mn, V and Fe concentrations and lower for Ni (56%) and Al (72%).

## 3. Model description of EPA PMF 3.0.

Positive Matrix Factorization (PMF) is powerful multivariate technique that constrains the solution to be nonnegative and takes into account uncertainty of the observed data [7]. This method relies on the time invariance of the source profiles and, thus, requires the emission particle size distributions to be stable in the atmosphere between the sources and the receptor site. It is reasonable to expect that particle size distributions will become relatively stable when sampling is carried out at some appropriate distance from the emission sources after initial size distribution changes in the vicinity of the sources due to coagulation and dry deposition [8]. A speciated data set can be viewed as a data matrix *X* of *n* by *m* dimensions, in which *n* is number of samples and *m* is chemical species to be measured. The goal of multivariate receptor modeling is to identify the number of factors *p*, the species profile *F* (*p x m*) of each source, and the amount of mass *G (n x p)* contributed by each factor to each individual sample. The model solves the general equation, Eq. (1),

$$X = G \cdot F + E, \quad (1)$$

where E is the residual matrix (observed estimated).
The most important step in source apportionment analysis using PMF definition of the the number of factors. The model implements nonnegative constraints in order to obtain more physically explainable factors and takes into account errors of the data points, by using them as point-to-point weights.Adjustment of the coresponding error estimates allows it to handle missing and below detection limit data. Data below detection limit (DL) can be retained for use in the PMF model, with the associated uncertainty adjusted so these data points have less influence on the solution than measurements above the detection limit. Data below DL were replaced with value DL/2, and (5/6) ·DL were used as a corresponding uncertainty [9].

## 4. Results and Discussion

### 4.1. $PM_{10}$ levels

Descriptive statistics for $PM_{10}$ concentrations during the summer and winter seasons are summarized in Tab. 1. During the 40 days of measurements in two campaign of measurement average $PM_{10}$ concentration in summer season 2010 is 28.10 μg $m^{-3}$ and 27.38 μg $m^{-3}$ in 2013. During the winter season 2011. average $PM_{10}$ was 132.65 μg $m^{-3}$ and 104.94 μg $m^{-3}$ in 2013 which represent seasonal variability of $PM_{10}$ concentrations.

**Table 1. Descriptive statistics for daily average $PM_{10}$ concentrations (μg $m^{-3}$) at sampling site**

| Banja Luka (suburban) | | | | |
|---|---|---|---|---|
| Time | Summer,2010 | Winter,2011 | Summer,2013 | Winter,2013 |
| N | 20 | 20 | 20 | 20 |
| Mean | 28.10 | 132.65 | 27.38 | 104.94 |
| SD | 8.66 | 61.19 | 5.85 | 48.57 |
| Max | 45.25 | 251.68 | 43.87 | 212.83 |

| | | | | |
|---|---|---|---|---|
| Min | 14.29 | 17.92 | 20.77 | 48.15 |
| Median | 27.39 | 140.85 | 26.38 | 88.80 |
| 98$^{th}$ | 44.34 | 237.12 | 41.01 | 199.72 |
| >LV | 0 | 19 | 0 | 18 |

Average mass concentrations of $PM_{10}$ are found significantly higher in the winter season during the both sampling campaigns than summer (Fig. 2). Concerning the 24h limit value (50 μg $m^{-3}$), 19 exceedances in 20 cases were recorded during the 2011. winter season (95%), and 18 exceedances in 20 cases (90%) during the 2013. winter season. According to the air quality, standard 35 exceedances are allowed throughout a year (9.6%). During the summer season, there were no exceedances 24h limit values. Seasonal variations are obvious due to the increase of heating demand in the winter season, as well as to the fact that prevailing meteorological conditions are less favourable for pollutant dispersion.

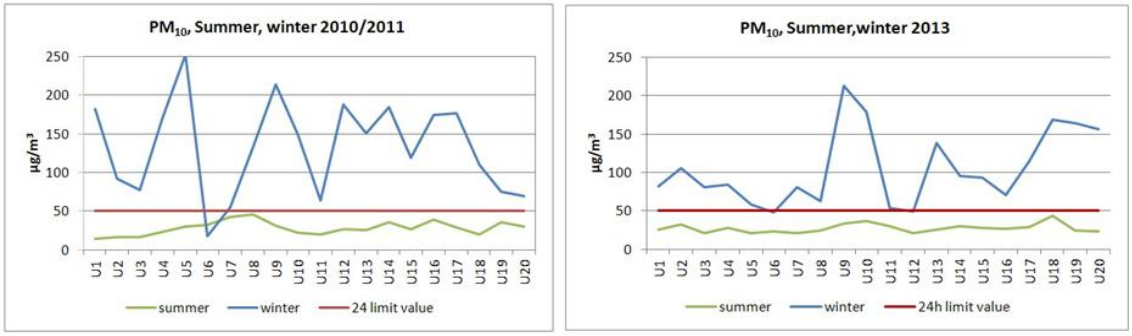



**Figure 2. Average daily mass concentrations of $PM_{10}$ in summer and winter season**

### 4.2. Chemical composition of $PM_{10}$

At the observed site, the mean concentrations of Pb, Zn and Fe in $PM_{10}$ were higher during the winter, season (Fig. 3), as a result of traffic emissions, heating and industrial activities, and K also, which is a significant trace for biomass burning emissions. Some elements from re-suspended dust and crustal materials like Ca, Mg and Na were higher in the summer (Fig. 3), and also secondary aerosols like $SO_4^{2-}$ that can be attributed to long-range transport.

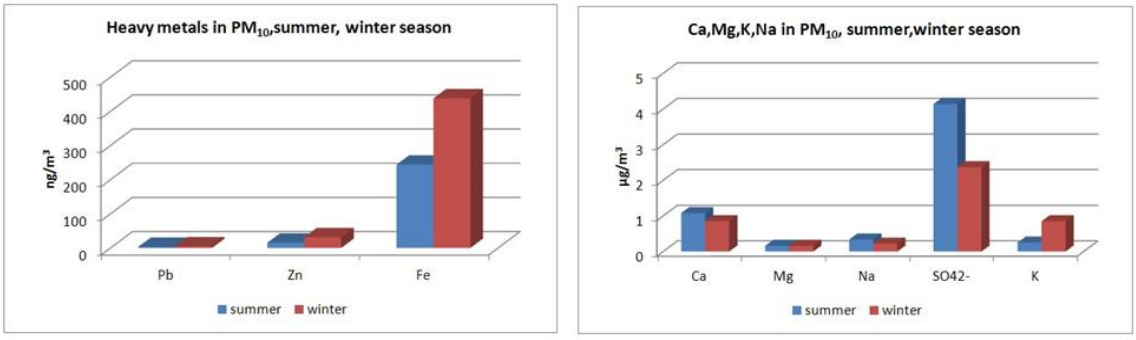



**Figure 3. Daily mass concentrations of heavy metals and cations in $PM_{10}$**

### 4.3. Relations between $PM_{10}$ concentrations and meteorological parameters

In order to estimate the correlation between $PM_{10}$ and concentrations of chemical species in $PM_{10}$, and meteorological parameters, Pearsons correlations coefficient was calculated using SPSS statistical software (Tab. 2).

**Table 2. Pearson correlation coefficients between chemical species and $PM_{10}$ concentrations and meteorological parameters**

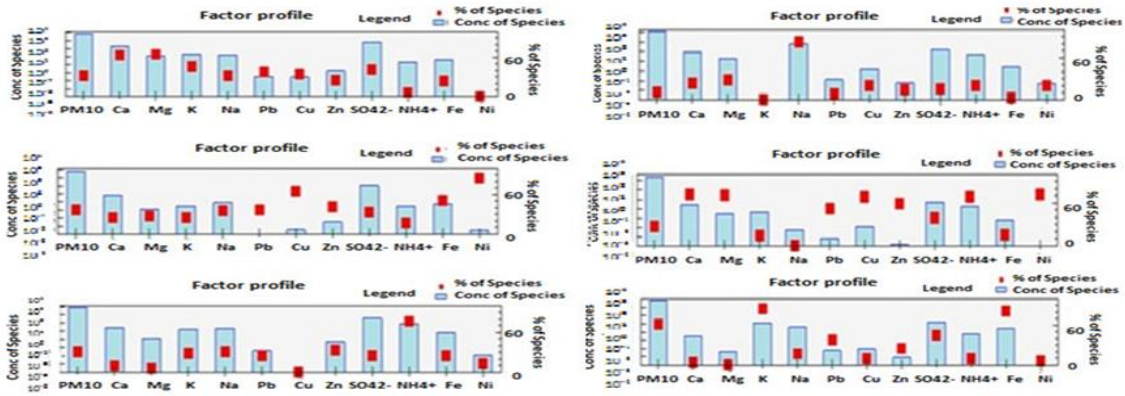
| Location | Season | K | Pb | **Zn** | Fe | $SO_4^{2-}$ | **$NH_4^+$** | Ni | Mn | T°C | **Ws** | Rh |
|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | Summer | .88** | - | - | - | .72** | .74** | - | - | .58** | .12 | -.15 |
| | Winter | .82** | .46* | .49* | .54* | .79** | - | - | - | -.48* | .50* | .09 |

*Correlations significant at the 0.95 confidence level.
**Correlations significant at the 0.99 confidence level.

At the observed site, strong correlations were found between $PM_{10}$ and $SO_4^{2-}$, $NH_4^+$ and daily temperature (T°C) that can be connected with secondary aerosols and long–range transport. High correlation between Ca and Mg, K and T°C and negative correlations with relative humidity can be connected with re-suspended dust in dry days. High correlations between Cu and Fe, Cu and Ni, and significant between Fe and Ni indicate vehicle-related emissions. During the winter season strong correlation between $PM_{10}$ and K, indicates wood burning, as a source of the emission. Negative correlation with T°C and wind velocity is attributed with higher energy demand and better pollutant dispersion. High correlations of $PM_{10}$ and K with Pb, Fe and Zn indicate a traffic contribution.

### 4.4. Positive Matrix Factorization

In order to identify possible sources and estimate their contribution to $PM_{10}$ mass concentration Positive matrix factorization was done by using US EPA - PMF 3.0 software. It is recommended to apply this model for at least 100 samples and 50 species [10,11].




**a)** **b)**

**Figure 4. Factor profile: The level of concentration and proportion of elements in total amount of PM10 in Banja Luka in summer and winter season**

At suburban site in summer season (Fig. 4 a)) Factor 1 is loaded with Ca, Mg, K, Na which could point to the dust re-suspension. Factor 2 is high loaded with Cu, Ni, Pb, Fe which attribute with traffic combustion. Factor 3 is loaded with ammonium ion that is connected with secondary aerosols and long–range transport. In winter season (Fig. 4 b)) Factor 1 is loaded with Na which origin from road dust. Factor 2 is loaded with Zn, Ni, Cu that comes from traffic and Ca, Mg from dust re-suspension. Factor 3 is connected with wood burning due to K as a tracer, because the investigations of the aerosols as a consequence of wood combustion, the elements K, S and Cl dominate the aerosol fraction [12]. Fe in this factor could point to the soil-related. It is important to know that sub-micron flying ash particles (<0.5 μm) are coming from a process of vaporization of easily volatilized ash components (S, Cl, Na, and K), and the heavy metal Zn, but also Pb and P, while coarse particles are coming from residual flying ash particles, which is ejected mechanically from the fuel bed and are carried by the flue gas upwards or intractable ash compounds of Ca, Mg, Si (if present) [5,13].

## 5. Prevention and mitigation measures for PM emission reduction

As previous investigation indicated, one of the major pollution problems is in the relation of the emissions from small household furnaces, mainly using biomass (wood) as fuel. Primary particulate emissions from biomass combustion systems coming from two main sources: from complete combustion, including inorganic material in the flying ash, and from incomplete combustion, which includes soot, condensable organic particles (tar), and char [5]. Unfortunately, the existing technical regulation does not require any systems for treatment of PM in small boiler units. Only units with a capacity 100 $kW_t$ and higher have to have some of the mechanical systems for PM removal. In nowadays there are some development attempts, which are going in way of development of some scrubber devices, predicted as technology for PM removal in residential applications [14]. Some efforts are going in direction of the new combustion technologies application, which are already commercialised, as concept based on a fixed-bed downdraft gasifier directly coupled with a multi-stage gas burner. This concept has proven its applicability for small-scale residential wood chip and pellet boilers. It is important to mention that this technology in case of the utilisation of ash and K-rich biomass assortments, can keep the fine particulate and the TSP emissions on such low levels (<5 mg/MJNCV) that no additional filter as in conventional combustion systems is needed [15]. As mentioned above Lazarevo has 1.752 buildings, and according to the "Typology of the residential buildings in Bosnia and Herzegovina", the most of the single-family houses were built in period 1961-1980, and average percentage on the level of Bosnia and Herzegovina was 31,1% (1961-1970), and 68,9% (1971-1980) [16]. According to Typology for the houses built in 1961-1970, average net surface of the heated space is 55,65 $m^2$ , and specific energy need for intermitted heating is 464,90 kWh $m^{-2}year^{-1}$. For the houses built in 1971-1980, average net surface of the heated space is 67,83 $m^2$ and specific energy need for intermitted heating is 381,59 kWh $m^{-2}year^{-1}$.

Rough calculation based on the above presented data shows that total energy need for Lazarevo area heating is 45.341 MWh $year^{-1}$ and it comes mainly from small biomass furnaces and boilers.

There are two approaches to mitigate PM and other emissions and pollution of the Lazarevo area, the first is the systematic improvements of the single houses, and the second is construction one of more

small district heating systems in this area. In the first case, suggested improvement measures will be insulation of the external wall with thermal insulation layer 10 cm (0,041 $Wm^{-1}K^{-1}$), insulation of the construction between the floors with the insulation layer 10 cm (0,041 $Wm^{-1}K^{-1}$), and the existing window replacement with the new ones $U$=1,6 [$Wm^{-2}K^{-1}$]. Beside of that installation of the central heating system for heating and domestic hot water based on wood or wood pellet boilers and accumulator with high efficiency, can reduce heat requirement for 63% in a case of single houses built in 1961-1970, and 65%, in a case of single houses built in 1971-1980.

This means that with the above suggested improvements total energy need for Lazarevo area heating will be 16.776 MWh $year^{-1}$, which is almost 2,7 times less than initially calculated.

With the more intensive improvement, suggested improvement measures will be insulation of the external wall with thermal insulation layer 20 cm (0,041 $Wm^{-1}K^{-1}$), insulation of the construction between the floors with the insulation layer 20 cm (0,041 $Wm^{-1}K^{-1}$), and the existing window replacement with the new ones ($U$=1,0 $Wm^{-2}K^{-1}$). Beside of that installation of the central heating system for heating and domestic hot water based on wood or wood pellet boilers and accumulator with high efficiency (including installation of thermostatic radiator valves), can reduce heat requirement for 77% in a case of single houses built in 1961-1970, and 79%, in a case of single houses built in 1971-1980. This means that with the above suggested improvements total energy need for Lazarevo area heating will be 5.269 MWh $year^{-1}$, which is 4,6 times less than initially calculated. The second option of small DHS obviously should be carefully planned, taking in to consideration possibilities to improve every single building first, because capacities of the future DHS will strongly depend on the steps taken as a first. The analysis provided in document called "Cost-optimal analysis of Buildings in Bosnia and Herzegovina with Testing of the New Climate Data for Residential and Non-residential Buildings", shows that specific primary energy for the selected single houses form construction periods 1981-1991 and 1992-2014 varied from 335 to 382 kWh/$m^2$a. After implementation of the above-presented measures, estimations shows that specific primary energy for this selected houses will decrease and will be in the range 90 to 114 kWh/$m^2$a. This means that reduction of energy needs for the buildings supply for 2.9 to 4,2 times, causes reduction of needs for the fuel supply which is in direct relation with reduction of the emission from the boilers and stoves used for space heating. Single houses from Lazarevo area are mainly from older period, which means that, expected reductions in energy consumption and emissions will be bigger.

## 6. Conclusion

The study was performed at urban site, Lazarevo, Banja Luka with different sources of pollution, road traffic, industries, re-suspended dust due to construction, and commercial and residential burning of fossil duels and wood also in the area without district heating. The air quality sampling was carried in two seasons, winter and summer, in 2010/2011, and 2013. 24h mass concentrations of suspended particles $PM_{10}$ were measured and their chemical composition (Pb, Cu, Zn, Fe, Mn, Co, Ni, Al, As, Se, Sb, Hg, Ti, V, $Ca^{2+}$, $Mg^{2+}$, $K^+$, $Na^+$, $NH_4^+$, $Cl^-$, $SO_4^{2-}$, $NO_3^-$). Average mass concentrations of $PM_{10}$ are found significantly higher in winter season during the both sampling campaign with exceedances 24h limit values than summer that represent seasonal variability of $PM_{10}$ concentrations. Concentrations of chemical species in $PM_{10}$ (Pb, Cu, Zn, Fe, Mn, Co, Ni, Al, As, Se, Sb, Hg, Ti, V, K, Na, Ca, Mg, $Cl^-$, $SO_4^{2-}$, $NO_3^-$ and $NH_4^+$) were found to be far below the standard limits. Seasonal variability of

concentrations indicates a significant contribution of wood and fossil fuel burning as well as specific meteorological conditions. Pearsons correlations coefficient show high correlations between concentrations of $PM_{10}$ and other pollutants. Strong correlations were found between $PM_{10}$ and K indicate wood burning as a source of emission. Positive Matrix Factorization was applied in order to identify possible sources of emissions and estimate their contribution to particulate matter mass. Three possible sources of emissions are estimated at sampling site. Traffic combustion, industry and other, not identified source, as secondary aerosols were the factors that highly contributed to the pollution of $PM_{10}$ during summer time. A wood combustion was identified as possible source during the winter season7. Due to that, provided analysis shows that improvements of the buildings envelope as well as installation of the high efficient combustion systems can reduce PM emissions significantly due to reduced need for fuel consumption.

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