18th WORLD CLEAN AIR CONGRESS 2019

WCAC'19

23-27 September 2019

ISTANBUL / TURKEY

PROCEEDINGS

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Melik KARA, Yetkin DUMANOĞLU, Gizem TUNA TUYGUN,
Abdurrahman BAYRAM, Tolga ELBİR

Turkish National Committee for Air Pollution Research and Control (TUNCAP)

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organized by

Turkish National Committee for
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and

The International Union of Air Pollution Prevention and Environmental Protection Associations (IUAPPA)

in collaboration with

ITU – Istanbul Technical University
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M. Kara, Y. Dumanoglu, G. Tuna Tuygun,
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PREFACE

18th World Clean Air Congress was held from 23 to 27 September 2019 in Istanbul, Hilton Maslak Hotel. The Congress was organized by the Turkish National Committee for Air Pollution and Control (TUNCAP) and International Union of Air Pollution Prevention Associations (IUAPPA) in collaboration with regional air pollution control organizations from around the world.

The 18th Congress, held in Istanbul, follows earlier congresses which have been held in the worldwide. For example, in 2016 it was held in Busan, Korea, proceeded by Cape Town, South Africa in 2013, and before that Vancouver, Canada, Brisbane, Australia, and London, in the UK, in 2004. Next World Clean Air Congress will be held in Singapore in 2021.

The 18th World Clean Air Congress, as one of the premier events in the field, provided a worldwide platform for scientists, policy makers and industrialists to discuss state-of-the-art scientific knowledge and latest progress and technical solutions in improving air quality. The declaration of the last World Clean Air Congress, in Busan, South Korea in 2016, was focused on identifying the impact of air pollution on health and climate, achieving climate mitigation targets, the adopting two-pronged approaches in to achieve climate targets and the sustainable development goals at the local, regional and global levels. This Congress had extended these efforts, while seeking to develop integrated strategies to improve air quality under changing climate. For these reasons the main theme of the 18th World Clean Air Congress was selected as “One Atmosphere: Integrating Climate and Air Pollution Science and Policy”.

The 5-day conference had about 300 participants from 44 countries in 5 continents. In total 20 plenary talks and 168 oral and 32 poster papers were given. Six major side events were organized by the international institutions during the world conference. We would like to convey my thanks and appreciation to the World Bank, European Federation for Clean Air and Environmental Protection Associations, Clean Air Asia and Malaysian Clean Air Association and World Resources Institute for their contributions by performing the Side Events. Special thanks to International Advisory Committee, Program Committee, reviewers, Session Chairs, and student volunteers who contributed to the success of this Conference. I also thank to the Scientific and Technological Research Council of Turkey, Istanbul Technical University, Dokuz Eylul University and for their support.

Finally, we would thank to all participants and sponsors without them we would not have been able to organize this outstanding conference.

We sincerely hope that this congress has been a place to enhance mutual understanding and deepen friendship, and promote cooperation among all participants and participating countries.

Selahattin Incecik
Congress Chair

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organized by TUNCAP and IUAPPA

Wintertime urban air pollution in Macedonia – composition and source contribution of air particulate matter

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Abstract. High air pollution episodes in most urban areas in Macedonia fill the headlines in recent years, reinforcing public perception that polluted air is by far most important environmental and health problem that urban population face nowadays. Ambient pollutants concentrations often reach dramatic levels triggering warnings and action plans that are mostly based on personal exposure reduction and hopes for changes in weather conditions, thus leaving public disappointed and confused. Recent studies show that traffic, domestic heating, natural dust and industrial activities are the main sources of PM contributing to urban pollution in European cities. However, there are significant differences between sources and the components of urban AP in different cities. While domestic heating (biomass burning) dominates the contributions to PM in Eastern Europe and in many developing countries, sea salt is the most important (natural) source of PM10 in north-western Europe. Therefore, detailed characterization (determination of size, form and chemical composition) of suspended air particulates is of crucial importance for definition of possible adverse health effects, sources allocation and applicable control measures.

During the last two years (2018-2019), AMBICON team has collected and analysed suspended particulate matters from specific urban zones throughout the country. Samples were taken according to standard gravimetric method (EN 12341:2014) using a low volume sampler and 47 mm PTFE filters. Chemical composition was determined using Fluorescent X-ray Spectrometer (Shimadzu EDX-900HS) according to EPA/625/R-96/010a. Seasonal and diurnal variation were obtained from MOEPP Air Quality Portal, as much as from AMBICON independent monitoring network with in house developed ambient particulate monitors.

The results demonstrate clear domination of biomass burning as primary contributor with much smaller contribution of traffic, industrial and crustal matter sources.

Keywords: Particulates, Concertation, Patterns, PMF, Contribution.

1. Introduction

Nine out of ten people are exposed to air pollution every day, establishing air pollution as the greatest environmental risk to health, that kills 7 million people prematurely every year from diseases such as cancer, stroke, heart and lung disease (WHO, 2019). Although the pollution sources significantly differ between the regions, around 90% of these deaths are in low- and middle-income countries, with high volumes of emissions from industry, transport, agriculture, and often most important dirty cookstoves and fuels in homes (WHO, 2019). And small, landlocked Macedonia is well fitted in this grim picture, as the largest urban areas are often high on the various pollution list, while capitol Skopje was pointed as most polluted capitol city in Europe (UNEP, 2018) with PM_{10} annual mean of about 69 µg/m^3 for 2016 (WHO, 2018).

What is even worse, this is not a new problem, and first research efforts, published more than a decade ago indicated high air pollution problem, locating heavy traffic (Dimitrovski and Bojkovska, 2002) and domestic heating and industry (Stafilov et al., 2003) as a main pollution source. As pollution patterns clearly shows some changes through a virtual elimination for some of the pollutants like Pb, and reduction for some SO_2, NO_2 and some metals like Cd, Fe and Mn, reflecting changes in source profiles like introduction of leadless gasoline and low sulfuric fuel, shifts from heavy oil to LNG in city heating plants, as much as upgrade of largest industrial plants with appropriate control equipment, particulate levels keep the trends showing same pattern throughout the last decade (Mirakovski et.al. 2018).

High particulates spikes, especially during the winter season, often reach hazardous levels thought the country urban areas, thus filling the headlines in recent years and reinforcing public perception that polluted air is by far most important environmental and health problem that urban population face nowadays. Confusing and partial data from different sources, slow warnings and inefficient action plans mostly based on personal exposure reduction and hopes for changes in weather conditions, leaving public disappointed and even more confused.

Ambient particulates or suspended particulate matter are common name for heterogenous mixture of solid and liquid particles which composition, biological and physical properties vary from location to location and changes over time, greatly due to temporal-spatial variations in emissions, varying proximity to sources, meteorological variables, and local topographies (Cho et al, 2018). Based on their size, these particles are divided into two major categories. Particles designated as PM2.5 (fine particles) have a diameter of less than or equal to 2.5 micrometres (µm), while particles with a diameter between 2.5 µm and 10 µm are designated as PM10 (coarse) particulates. Depending on the atmospheric conditions those particles can remain airborne for long periods and invade the indoor air environment. Majority of particulate mass is comprised of different forms of carbonaceous substances, sulphates, ammonium, nitrate, and various ions. Manganese, copper, zinc, cadmium, chromium, iron, nickel, potassium, calcium, vanadium, barium, arsenic, selenium and strontium are the most commonly found metals in the pollution sources (Magnani et al 2016). Chemical composition of PM is important determinant in its health outcomes (Kundu and Stone, 2014). Also, chemical composition of particulates at the point of impact or receptor, can be used to evaluate the contamination and pollutant sources contributions through a receptor modelling (some of commonly referred factor analysis methods).

Therefore, detailed characterization (determination of size, form and composition) of air particulates is of crucial importance for definition of possible adverse health effects, sources allocation and applicable control measures.

Traying to shed a light on the problem, during the last two years (2018-2019), we have collected and analysed suspended particulate matters and concentration data from several urban zones throughout the country. Samples were taken in Skopje, according to standard gravimetric method (EN 12341:2014) using a low volume sampler and 47 mm PTFE filters. Chemical composition was determined using Fluorescent X-ray Spectrometer (Shimadzu EDX-900HS) according to EPA/625/R-96/010a method. Seasonal and diurnal variation were obtained with real time monitoring during the sampling campaigns, as much as from UGD AMBICON independent monitoring network in cities out of Skopje. Positive matrix factorisation was used to identify major PM_{10} contribution sources in Skopje.

2. Materials and methods

2.1. Sampling

Ambient air monitoring was performed at two locations in Skopje, one road side at Boulevard "Ilindenska" and one background location at "Aminta the III" (Figure 1). The road side (RS) sampling point was located within the Skopje City Hall courtyard, about 2 meters from the road edge and background sampling (BS) point was located about 600 m to the east within Ministry of Agriculture courtyard.

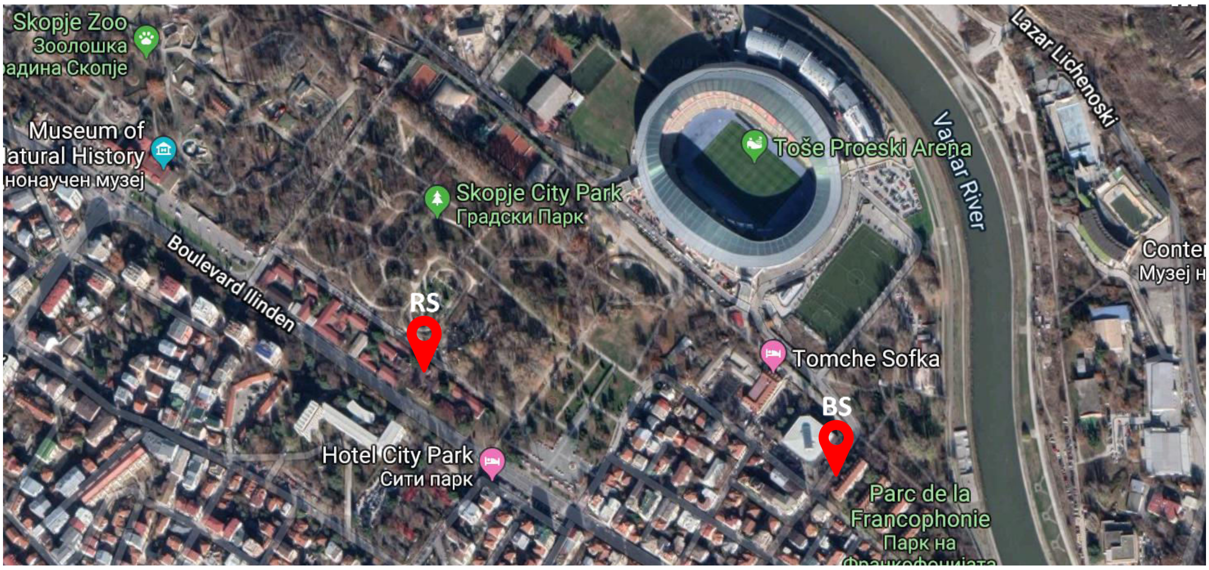

Figure 1. Sampling locations

Both locations were equipped with sequential dust sampling system PNS 16T-3.1 (Comde Derenda, Germany) with 16 filter cassettes for continuous collection of particulate matter and Air Pointers (MLU Recordum, Austria) for real time monitoring of PM_{10}, $PM_{2.5}$, NO_2 and CO using compliance or equivalent methods.

Sampling was performed at 2.2 meters height, continuously during 14 consecutive days in each season starting from November 8 - 21.2018, January 18-31.2019, May 6-27.2019 and July 13-27.2019. Particulate ($PM_{1}0$) samples were collected on 47 mm PTFE filters and handled and measured gravimetrically fully in line with recommendation given in EN 12341:2014 Ambient air - Standard gravimetric measurement method for the determination of the PM_{10} or $PM_{2,5}$ mass concentration of suspended particulate matter.

2.2. Elemental analysis

Elemental composition was measured by the energy dispersive X-ray fluorescence (EDXRF) using Fluorescent X-ray Spectrometer (Shimadzu EDX-900HS, Japan) for determination of Na, Cl, K, Ca, Mn, Fe, Ni, Cu, Zn, As, Cd, Pb and Si fuly in line with EPA/625/R-96/010a, Method IO-3.3 Determination of Metals in Ambient Particulate Matter Using X-Ray Fluorescence (XRF) Spectroscopy. Black Carbon was analysed using OT21 Transmissometer with dual wavelength light source; 880nm providing the quantitative measurement of Black Carbon in PM, and a 370nm for qualitative assessment of certain aromatic organic compounds.

2.3. Source contribution calculation

Source contribution/apportionment of PM_{10} mass by Positive Matrix factorisation was performed using the EPA PMF version 5.0.14.21735 program in accordance with the user's guide (EPA, 2014). Positive

Matrix Factorization (PMF) is a receptor model, developed by Dr. Pentti Paatero (Department of Physics, University of Helsinki) in the middle of the 1990s (Paatero and Tapper, 1994), in order to develop a new method for the analysis of multivariate data that resolved some limitations of the PCA (Camero, 2009). One of the main positive aspects is the use of know experimental uncertainties as input data which allow individual treatment of matrix elements and can accommodate missing or below-detection-limit data that are a common feature of environmental monitoring (Song et al., 2001). PMF results have a quantitative nature and therefore it is possible to obtain the composition of the sources determined by the model (Paatero, 2004). Concentration and uncertainty data matrices were compiled as recommended in PMF 5.0 Fundamentals and User Guide (EPA, 2014). In total 20 base runs were performed, using 4 factors and base random seed with 0 % extra modelling uncertainty. Using the calculated sound to noise (S/N) ratios as recommended, Cl and Cd were categorized as "Bad" and excluded from the analysis. Na and As were included as "Weak" while the K, Ca, Mn, Fe, Ni, Cu, Zn, Pb, Si and EC were categorized as "Strong".

3. Results and discussion

3.1. Particulates concertation

Data collected confirm more or less well-known fact that high pollution episodes occur almost exclusively during the heating seasons, in periods with stable atmospheric conditions, as the deep valleys express strong temperature inversion (normal decrease of air temperature with height is switched to increase) that prevent normal circulation in the atmosphere, creating some times prolonged periods with weak winds and leading to accumulation of emitted substances near the ground surface. And this is common for most of the country urban areas including Skopje, Prilep, Kavadarci, Strumica, Tetovo and so on (Figure 2). Most of the urban areas does manifest PM_{10} yearly average above the TLV of 40 μg/m^3, like Skopje agglomeration (53.6 μg/m^3), Bitola (44 μg/m^3), Kumanovo (52 μg/m^3), Tetovo (49 μg/m^3), Kicevo (43 μg/m^3) and Kavadarci (55 μg/m^3), but values out of the heating seasons are mostly well within the TLV's (MOEPP, 2019).

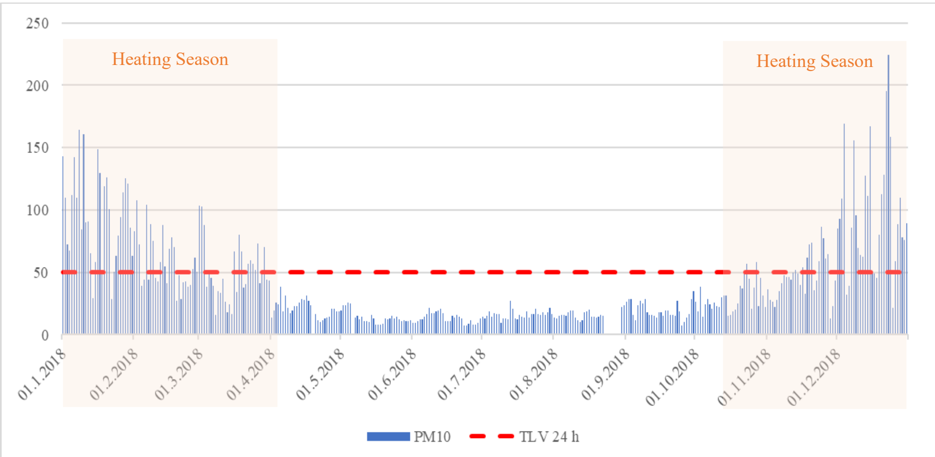

Figure 2. PM_{10} daily averages for Kavadarci Center (UGD.AMBICON Monitoring Network)

Distinct diurnal cycles are also common for most the cities covered with the monitoring systems. During the high pollution episodes, they all exhibit bimodal pattern, with two peaks in morning and late evening (Figure 4). Such patterns are driven with natural changes in boundary layer height, but are also in direct conjuction with patterns of home heating usage, which also peaks in the morning and evening hours. And if the periods of stagnant atmosphere are prolonged for several consecutive days, the pattern persist with the maximums increased each consecutive day (Figure 3).

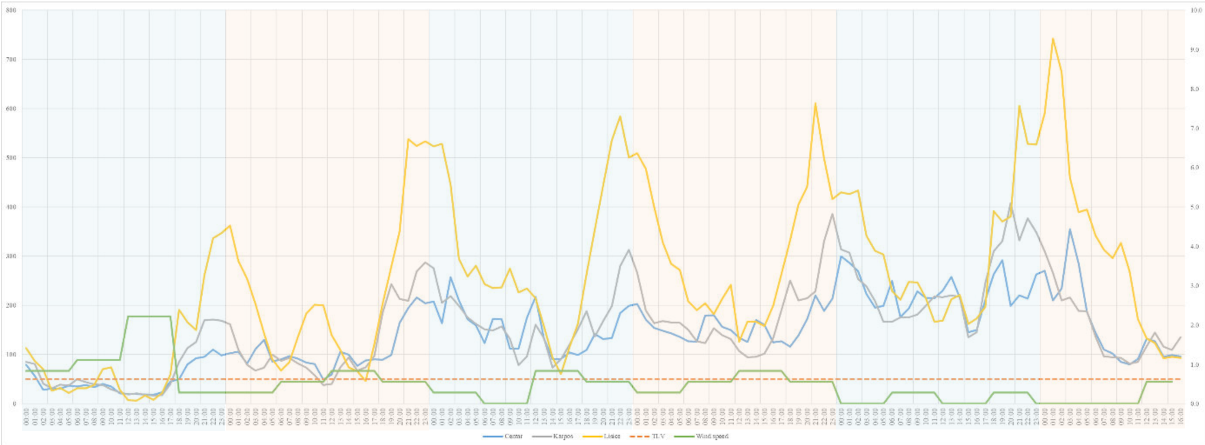

Figure 3. PM_{10} Diurnal patterns for Skopje agglomeration 04-09.01.2018 (Mirakovski, 2018)

Similar diurnal patterns are reported elsewhere, for regions where domestic wood combustion for home heating is known to be a significant contributor to PM10 concentrations during the winter (Ancelet et. al., 2014, Trompetter et. al., 2010).

Figure 4. PM_{10} diurnal patterns in Skopje, Strumica, Kavadarci and Prilep (16.01.2019 - UGD.AMBICON)

In addition, there is a correlation between particulates concentration (PM_{10}) and carbon monoxide (CO), which can be used as tracer for pollutants from combustion related sources (Sahu and Lal, 2013). Data obtained during our measurement campaign, shows excellent correlation during the heating season (autumn/winter) with Pearson coefficient value r = 0.88, and no correlation between the same during the spring/summer season (r = - 0.12). Knowing CO emission inventory for Skopje agglomeration where domestic heating participate with 59%, industry with 16 % and traffic with 25 % (MOEPP, 2019) it could be safely concluded that domestic wood combustion contributes to significant part of winter time air pollution and actually drive pollution patterns explained above. And assuming the similar conditions in other cities mentioned above like Strumica or Prilep, where industry and traffic have even lower contributions, domestic wood combustion could be the single important source of pollution.

3.2. Source contribution

As the composition of the particulate matters at background and road side locations monitored in Skopje, does not differ significantly, especially during the heating season, Positive Matrix Factorization was performed on background data, as more representative for wider city area. Statistical description of the input data including average, maximum, and median concentrations of species used for source apportionment, as well as standard deviations, average uncertainties and limits of detection are given below (Table 1). As only 54 valid samples were available, stretched over a 12-month period, PMF exercise should be seen as indication for dominant sources and cannot replace full scale source apportionment study.

Table 1. PMF input data (54 samples)

	Unit	Average	Maximum	Median	Standard Deviation	Average Uncertainty	Detection limit
PM_{10}	µg/m³	52	187	36	36	3	3.0
$PM_{2.5}$	µg/m³	36	174	14	38	2	3.0
EC (PM_{10})	ng/m³	15034	43550	10712	9548	752	100
Na (PM_{10})	ng/m³	76	624	25	114	30	39
Cl (PM_{10})	ng/m³	94	763	90	92	141	181
K (PM_{10})	ng/m³	477	2216	216	535	108	108
Ca (PM_{10})	ng/m³	1232	2911	1214	757	212	118
Mn (PM_{10})	ng/m³	26.1	204.5	6.9	44.6	4.4	5.1
Fe (PM_{10})	ng/m³	707	1513	677	338	71	73
Ni (PM_{10})	ng/m³	14.0	75.2	5.8	16.8	1.6	1.6
Cu (PM_{10})	ng/m³	17.9	195.9	8.1	30.1	4.1	5.1
Zn (PM_{10})	ng/m³	34.6	401.4	0.9	90.1	6.1	1.9
As (PM_{10})	ng/m³	0.09	1.17	0.01	0.23	0.03	0.20
Cd (PM_{10})	ng/m³	0.50	0.50	0.50	0.00	0.83	1.00
Pb (PM_{10})	ng/m³	18.6	139.8	2.6	29.0	4.2	5.2
Si (PM_{10})	ng/m³	176	658	109	164	61	118

As large numbers of species usually encountered in particulate matters were not quantified, including often dominant water-soluble ions (NH_4^+, SO_4^{2-} and NO_3^-), reconstructed masses determined using the elemental data accounted for only 28.4 % of the PM_{10} mass. Preforming multiple PMF runs to elemental data give optimal solution with 4 factors, identified as biomass burring (high EC content, K, Na and Si), industrial (Ni, Si, Na, Cu, As), traffic (Zn, Pb, Cu, EC) and crustal (Si, Ca, Fe, K) (Figure 5). Some of the elements have contribution in several sources, as some processes, like resuspending road dust or common combustion sources, contribute to a mixed source profiles (Ca, Na and K in traffic and crustal matter or EC in traffic and biomass burning).

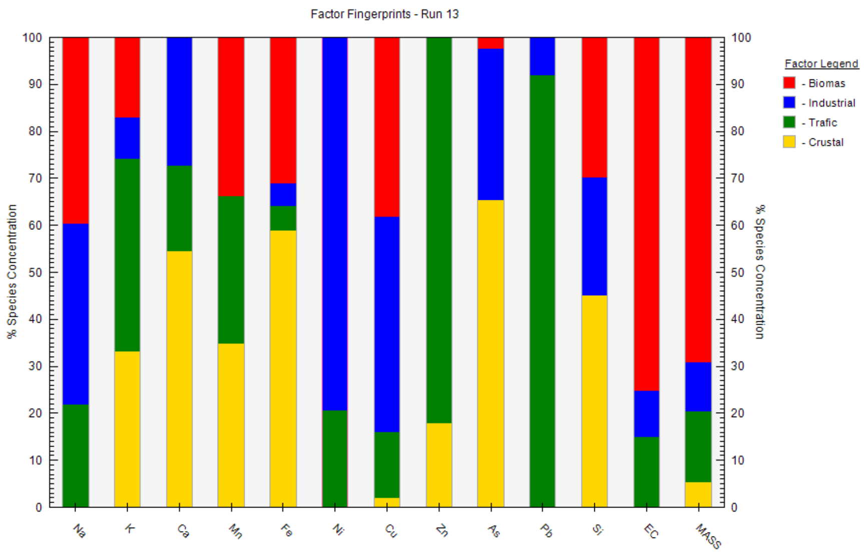

Figure 5. Factor fingerprints

The first factor, characterized as biomass burning has by far highest contribution to PM_{10} mass reaching 69.2 %, while traffic, industrial and crustal maters contribute 15.2 %, 10.4 % and 5.3 % respectively (Figure 6).

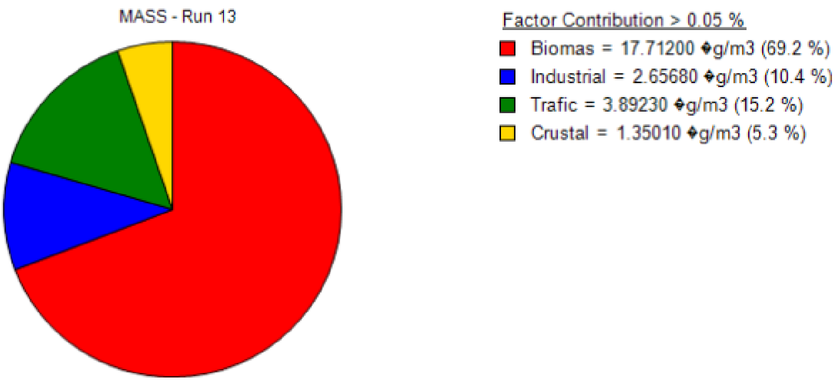

Figure 6. Factor contributions to PM_{10} mass

Such high contribution from biomass burning is not surprising, having in mind Skopje agglomeration emission inventory for reference 2014, where domestic heating participates with 91%, in total PM_{10} emissions, while industry, energy production, traffic, waste management, agriculture and construction have altogether with about 9 % (FMI & MOEPP, 2015). Adding the temporal distribution of this emissions to the picture, with biomass burning narrowed to in less than six months, this could be easily determined as the only significant source, that should be somehow altered in order to reduce frequent wintertime pollution episodes.

4. Conclusions

We have aimed to indicate sources that contribute to high particulate concentration during wintertime pollution episodes, by sampling and analyzing suspended particulate matters and concentration data in Skopje and several urban zones throughout the country during the last two years (2018-2019).

Concertation data collected confirm that most of the urban areas manifest PM_{10} yearly average above the TLV of 40 µg/m^3, like Skopje agglomeration (53.6 µg/m^3), Bitola (44 µg/m^3), Kumanovo (52 µg/m^3), Tetovo (49 µg/m^3), Kicevo (43 µg/m^3) and Kavadarci (55 µg/m^3), but values out of the heating seasons are mostly well within the TLV's. Distinct diurnal cycles are also common for most the cities covered with the monitoring systems, and they all exhibit bimodal pattern, with two peaks in morning and late evening. Such patterns are driven with natural changes in boundary layer height, but are also in direct conjunction with patterns of home heating usage, which also peaks in the morning and evening hours. In addition, there is an excellent correlation between carbon monoxide (as tracer for combustion related sources) and particulate concentration during the heating season (autumn/winter) with Pearson coefficient value r = 0.88, and no correlation during the spring/summer season (r = -0.12).

Positive Matrix Factorization was performed on background data, as more representative for wider city area, identifying 4 factors as biomass burring (high EC content, K, Na and Si), industrial (Ni, Si, Na, Cu, As), traffic (Zn, Pb, Cu, EC) and crustal (Si, Ca, Fe, K). The first factor, characterized as biomass burning have by far highest contribution to PM_{10} mass reaching 69.2 %, while traffic, industrial and crustal maters contribute 15.2 %, 10.4 % and 5.3 % respectively (Figure 6).

All this strongly indicate biomass burning as the only significant source, that should be somehow altered in order to reduce frequent wintertime pollution episodes.

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