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**DETERMINING THE MAJOR SOURCES OF PM₁₀ THROUGH MODELING AND ITS
IMPACT ON THE QUALITY OF THE AMBIENTAL AIR IN BITOLA**

-Doctoral Thesis-

ABSTRACT

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INTRODUCTION

Air pollution is a very important element of the environment and at the same time is a complex problem with a great challenge to reduce and manage. Air quality continues to be an important issue related to public health, the economy and the environment.

Air pollution problems already transcend local significance and are an issue that is being addressed in a wider regional context. Air pollution can be formed and transported over long distances and can affect a large area. Transport of pollutants, trans-boundary pollution caused by atmospheric movements, presents a huge challenge for the countries of the region. Air pollution has adverse effects at the regional level and has several significant environmental impacts and can directly affect vegetation, water quality and soil.

For complete risk management, the key issue is the identification of particulate sources. Sources affecting particulate matter concentration at a given location can be determined by applying methods based on the chemical composition of the particulate matter (PM), air pollution measurement data and meteorological data. The studies provide an estimate of the average impact of local air quality sources as well as temporal data on specific PM concentrations. Therefore, particle segregation by source and analysis of their individual impacts can yield results that can be directly used in risk assessment, risk management, and in setting air quality guidelines and standards.

Thus, the idea of defining the main sources of air pollution in the urban part of Bitola came up. Monitoring stations set up by the Ministry of Environment of the Republic of North Macedonia continuously measure concentrations of suspended particulate matter PM₁₀ (suspended particle size from 0 to 10 micrometers) without information on sources of pollution. In this way, the concentration was measured at different locations in the city so that the conceptual model could identify the main sources. The measurements and analysis resulting from the measurements are an integral part of this doctoral thesis.

The purpose of this doctoral thesis is to determine the impact of the main PM₁₀ suspended particle sources in the urban part of Bitola and to reflect current conditions with suspended particle concentrations. The content of this paper covers only the impact of wood burning used for home heating and the impact of traffic through exhaust and non-exhaust emissions. Other sources such as the impact of the construction industry, other industrial facilities, metal processing and power plant-thermal power generation are not an integral part of this thesis.

The doctoral thesis contains eight chapters - five relate to the theoretical section and three to the research section. The first chapter elaborates the subject of the doctoral thesis and its aims, together with the methods used in the research and the expected results. Also, an overview of current research findings has been made from a global and local perspective.

The second chapter reviews the EU air quality legislation and national legislation. This chapter also describes the methods for measuring concentrations of pollutants with constant measurement stations and with indicative measurements.

The third chapter gives a theoretical description of the PM₁₀ suspended particles and defines the particle categories in terms of the sources they emit, natural and anthropogenic, primary and secondary. This chapter also covers the particle size which together with the mass concentration are the most important physical properties of the particles. The chemical properties are not covered in this paper because the chemical composition of the particles varies depending on the presence of the sources. In terms of the topic of the doctoral thesis this chapter describes the mechanisms of the formation of suspended particles with an emphasis on physical friction and combustion.

The fourth chapter covers the sources of particulate matter emissions. This chapter covers only the wood burning for heating homes in winter and the impact of traffic on exhaust and non-exhaust emissions.

The fifth chapter describes the health and environmental effects of air pollution. This chapter presents the impacts of the physical and chemical properties of the particles. In terms of health effects, physical properties have been increasingly researched by researchers as their impact on human health is greater than the chemical composition of the particles. Possible diseases depending on the sources of emission are also shown. The impact of vegetation on air pollution is also an integral part of this chapter.

After the measurements, data were processed showing maximum, minimum, average values and standard deviation and are presented in tabular and graphics in order to analyze them, to determine sources of biomass combustion in winter and impact of traffic during the period when there is no biomass combustion. The conclusions about the current situation in the urban environment of the city of Bitola are shown in Chapter Six. This chapter shows in tabular and graphically the measurements of TSP (Total Suspended Particles - particle size between 50 and 100 micrometers), PM₁₀, PM_{2.5} (suspended particles from 0 to 2.5 micrometers, and PM₁ (particulate matter with size between 0 and 1 micrometer) from five measuring locations for the period October 2016 to March 2017.

The seventh chapter presents the analysis that emerged from the measurements. The analysis is made for the two sources separately. Three locations were used to analyze the impact of wood burning on home heating and traffic impact. In this research conceptual modeling techniques are applied. Also, the impact of biomass combustion on household heating has been taken into considered before the start of the heating season and during the heating season, while the impact of traffic has been considered in a location with no traffic and compared to locations where the impact of the traffic is the largest source.

The eighth chapter gives an overview of the conclusions reflecting overall progress and the benefits of research and analysis within the doctoral thesis, as well as recommendations for further work and future scientific research activities in the subject area.

1. BASIS FOR THE REALIZATION OF THE DOCTORAL THESIS

Air quality is an important issue related to public health, the economy and the environment. Poor air quality can cause deterioration of health, premature death and damage to ecosystems. Thus, enormous economic damage to the state can be caused expressed through reduced labor productivity and worsening environmental conditions.

Effective solutions to reduce air pollution require a good understanding of its contributors, that is how pollutants are transported and transformed into the atmosphere and how they affect humans, ecosystems and the climate.

Effective air quality principles require co-operation and action at global, national and local levels. Air pollution reduction policies and strategies are primarily aimed at reducing emissions in the combustion process, using alternative energy sources, renewables and biofuels as primary processes to improve air quality, opposed to inappropriate waste treatment, the use of wood for home heating in winter, incomplete combustion of fuels in vehicles and more.

By setting up measuring stations that monitor air quality and measure mass concentration, important information on ambient concentrations can only be obtained at certain locations in a given time, without describing the sources of emissions and the causes of poor air quality. Therefore, source identification plays an important role in air quality management, as it can provide a complete description of current conditions.

1.4. OVERVIEW OF CURRENT KNOWLEDGE IN THE AREA OF RESEARCH

There is little research on the quality of air in the urban part of Bitola. One is the Pilot Air Quality Improvement Program in Bitola, developed as part of the activities of the Twinning project "Strengthening the capacities at local and central level for environmental management in the area of air quality" funded by the European Union. This program is prepared in accordance with the Law on Air Quality which transposes the requirements of the European Union air quality legislation. Based on this program, the most critical pollutants in Bitola are suspended particles up to 10 micrometers in size – PM_{10} . PM_{10} concentrations exceed both daily and annual limit values at both stations. Identifying sources is a key issue in determining the origin and characteristics of PM_{10} in order to be able to prepare planned short- and long-term environmental improvement activities.

The Doctoral Thesis DETERMINING THE MAJOR SOURCES OF PM_{10} THROUGH MODELING AND THEIR IMPACT ON THE QUALITY OF THE AMBIENTAL AIR IN BITOLA is a continuation of the research in this field.

2.LEGISLATIVE FRAMEWORK FOR AIR QUALITY

2.1.EU DIRECTIVES ON AIR QUALITY

Council Directive 96/62 / EC of 27 September 1996 on the assessment and management of ambient air quality was published on 21 November 1996. This Directive represents a milestone in the area of European Union air quality regulations (EU 1996). The new Framework Directive replaces the Directives on SO₂ and particulate matter (80/779 / EEC), Pb (82/884 / EEC), NO₂ (85/203 / EEC) and O₃ (92/72 / EEC) that were previously active in the European Union.

With several sub (daughter) directives, it was the basis for a new EU air quality policy. The purpose of the Framework Directive was to set the basic principles of a common strategy, while the sub (daughter) directives set air quality standards (limit and target values and in some cases warning thresholds) for 13 pollutants.

In May 2008, Framework Directive 96/62 / EC, the first three sub (daughter) directives and the Information Exchange Directive were replaced by the new EU Directive 2008/50 / EC. Restricted and target values, information threshold and alert were retained except for the second phase of the PM₁₀ limit value which was removed. Additionally, based on recent health research on the harmful effects of PM_{2.5}, monitoring requirements, as well as limit and target values have been established for this pollutant.

European Community legislation on ambient air quality mainly consists of Directives 2008/50/EC and 2004/107/EC.

2.2.NATIONAL LEGISLATION ON AIR QUALITY

Basic legislation in the field of air quality in the Republic of Macedonia is the Law on Ambient Air Quality (Official Journal 67/04, 92/07, 83/09, 35/10, 47/11, 100/12, 163/13 , 10/15, 146/15) which is based on EU air regulation.

The purpose of the law is to reduce and prevent harmful effects on human health and environment and also to maintain good air quality where there isn't. According to this law, everyone is obliged to act carefully and responsibly to prevent ambient air pollution and the harmful effects on human health and the environment.

According to this law, the authorities are obliged to take all necessary measures, so the target values of pollutants in the ambient air do not exceed the target values.

Based on the law on ambient air quality bylaws have been prepared. According the Regulations and methodology for ambient air quality monitoring, the measurements of pollutants are taken at fixed locations or at random sampling.

3. PARTICULATE MATTER PM₁₀ IN THE AIR

The composition and distribution of particle size, and hence their harmful effects, depends on the processes of particle formation and particle sources, and numerous studies have been investigated concerning PM₁₀ and PM_{2.5}. In these studies, the main sources of RM are traffic, energy production using fossil fuels and biomass, industrial sources, soil resuspension, and others, but the relative contribution of different sources varies greatly depending on time and location. Resuspension of the accumulated dust from the streets due to traffic and wind is a specific problem in urban areas. Also, the resuspended dust may cause the limit values to be exceeded, which are primarily directed at regulating the concentrations of suspended particles from anthropogenic sources.

3.1.PARTICULATE MATTER CATEGORIES

Atmospheric particles can be categorized as primary and secondary particles. Primary particles are emitted directly into the atmosphere from anthropogenic particle sources. The main anthropogenic sources of PM include transport, stationary combustion sources, space heating combustion, biomass combustion and dust. Urban PM emissions from anthropogenic sources are a complex mix, as most sources emit both primary particles and precursor gases to form secondary particles. Most of the PM anthropogenic emissions are emitted within relatively small urban and industrial areas, resulting in hot spots of high concentrations of particles and other air pollutants. Primary and secondary particles from anthropogenic sources also affect regional PM concentrations, as some of the emitted particles may remain suspended for several days and travel up to thousands of kilometers in the atmosphere.

Secondary particles are formed in the atmosphere by reactions of emissions of primary gaseous substances. As for the secondary particles, particular attention is given to PM_{2.5} particles that are formed as new particles mainly as a result of the reactions of the primary gases or react with other already formed particles. The most common gases that lead to the formation of secondary particles are SO₂, NO_x, NH₃, and VOC (chemical compounds whose molecules contain carbon). The main precursor gases react in the atmosphere to form sulfate, nitrate and ammonia compounds. These compounds form new particles in the air or react with other particles to form so-called inorganic aerosols. Most of these gases are emitted from anthropogenic sources.

3.2.SIZE OF PARTICULATE MATTER

The suspended particles are a complex heterogeneous mixture of particles in the solid and liquid phase that vary in size, shape, color, chemical composition, physical characteristics and origin. Particle size is the most important parameter for determining the behavior of suspended particles in the air. All features of suspended particles depend on their size. The basic particle size unit is μm (micrometer).

Depending on the formation process and size, atmospheric particles are classified into ranges. We distinguish ultrafine particles up to 0.1 μm (nucleic range), fine particles from 0.1 to 2 μm (accumulative and nucleic range) and coarse particles over 2 μm .

Ultrafine and fine particles are formed mainly by chemical reactions between gases in the atmosphere, while coarse particles are formed primarily by physical friction and the combustion process. Nuclear range or ultrafine particles contain particles smaller than 0.1 μm in diameter that are directly emitted by the combustion

process. The life of nucleic particles after combustion is usually less than one hour because they coagulate rapidly with larger particles (Judith C. Chow, John G. Watson, 1998). Nuclear range can be detected by measuring post-combustion emission or when new particles are formed in the atmosphere (Lundgren and Burton, 1995).

The accumulative range contains particles of 0.1 to 2 μm . These particles are the result of the coagulation of ultrafine particles emitted by the combustion process, condensation of volatile compounds and dust. Particles in this range can contain ammonium nitrate, ammonium sulfate, ammonium bisulfate, sulfuric acid, organic carbon and elemental carbon.

Particles larger than 2 μm in diameter are called coarse particles. Coarse particles are mainly produced by mechanical processes and are emitted directly into the atmosphere from natural and anthropogenic sources. Sea salt particles, soil and road dust, sand, pollen and spores are natural sources of suspended particles in the atmosphere. Coarse particles from anthropogenic sources are the result of physical friction in various industrial and agricultural processes, the vehicles on the roads and more.

Large particles smaller than 50 μm in diameter are classified as total suspended particles (TSP). PM_{10} , are coarse particles up to 10 μm in size, while $\text{PM}_{2.5}$ are fine particles smaller than or equal to 2.5 μm . Particle size from 1 μm to 10 μm is particularly important in controlling air pollution. Most of the particles that form in some industrial processes are in this range. For human health, particles up to 10 μm in size are treated as a respiratory problem.

3.3.PARTICULATE MATTER FORMATION

The size range of the particle formed in the process largely depends on the type of mechanism by which the particle is formed. The most important mechanisms for particle formation in air pollution sources are:

- Physical friction / mechanical dispersion
- Particles from the combustion process
- Coagulation
- Condensation
- Nucleation

4.SOURCES OF PARTICULATE MATTER PM₁₀ IN THE URBAN AIR

The main sources of anthropogenic or artificial particles are formed by stationary combustion of coal for electricity production, firewood and pellets for household heating, burning of biomass for industrial processes and emitted traffic (street dust) emissions.

Suspended particles in the urban environment represent a complex mixture of particles from many types of sources, as most sources emit both primary particles and gases to form secondary particles.

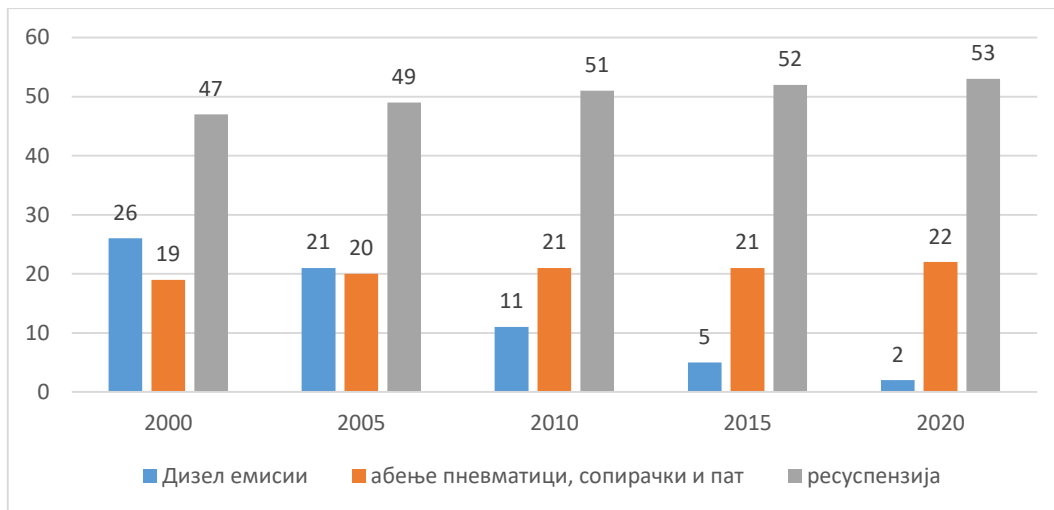
The distinction between sources, anthropogenic and natural particles and emitted particles is sometimes difficult to distinguish, for example, dust emissions and biomass combustion. Also, there are large differences in the relative importance of different sources from one geographical area to another. For example, most of the primary particulate emissions in eastern Europe come from stationary sources and combustion processes, while in western Europe emissions are evenly distributed across all economic sectors, although transport emissions play the most significant role in many locations (Vallius, 2005).

4.1.ROAD TRAFFIC EMISSIONS

Traffic-related particulate sources are a significant contributor to ambient air particulates especially in urban areas and major cities. Particulate emissions related to traffic can be classified into two groups: road emissions caused by exhaust gases as a result of incomplete combustion of fuel and evaporation of the engine lubricant during the combustion process and particulate emissions which are related to traffic and derive from non-exhaust (non-exhaust) sources of vehicles such as wear on brakes, tires, gear coupling and road surface. These sources contribute significantly to the concentration of PM particles in urban ambient air (Penkala et al. 2015). Particles from these sources are constantly generated in the ambient environment but also constantly deposited on the surface of the earth and re-suspended in ambient air due to the traffic.

There are several difficulties when studying emissions from non-exhaust sources. Primarily due to the lack of standardized measurement procedures, researchers have used many different sampling methodologies that often result in incomparable or even contradictory results and conclusions.

Research shows that the sources of exhaust and non-exhaust emissions from traffic contribute almost equally to the total emissions of PM₁₀ in ambient air. Much of the research over the last three decades has focused on exhaust emissions. Stricter regulations and new technological solutions have resulted in significant reductions in emissions. But despite these reductions, vehicle emissions are still present. Even with zero exhaust emissions, traffic will continue to be an important source of ambient air particles through processes of physical friction / mechanical dispersion and resuspended dust (Pant and Harrison, 2013). In the future, most of the emissions from road traffic into ambient air will be caused by non-exhaust sources. Rexeis and Hausberger predict that in central Europe, the concentration of PM from the contribution of non-exhaust emissions to total traffic emissions will increase by 80-90% (Harrison et al. 2012). As a result, research into this type of emissions will become increasingly important. Figure 4.1 shows a scenario of PM₁₀ emissions in Germany from 2000 to 2020 built in Gg/year (Jörß et al. 2010):



Graph 4.1. PM10 emissions according to PAREST-Referenzszenario, Jörß et al. 2010

A key reason for research into non-exhaust emissions is their tendency to act as carriers of heavy metals and carcinogenic compounds that violate existing ambient air standards and norms (Pant and Harrison, 2013). Although non-exhaust particulate matter is more aerodynamic in diameter than the exhaust particulate matter, it is still within the size that it can penetrate the respiratory system and cause adverse effects on human health.

Exhaust emissions are predominantly particles up to 2.5 micrometers, while non-exhaust sources are dominated by particles up to 10 micrometers (Pant and Harrison 2013; Abu Allaban 2003).

Gasoline and diesel fuel combustion in vehicle engines produce emissions of several potentially harmful substances. The primary emissions from motor vehicles come in two dominant forms: gaseous and particulate matter, which can be found in relatively high amounts in the atmosphere, and so called air toxins, which are usually found in small amounts in the atmosphere, but can have important health implications for people. Primary pollutants include carbon monoxide, nitrogen oxides and PM₁₀ and PM_{2.5} particles.

Road transport is known as a major source of air pollution. Although emission regulations for European passenger cars have been active for more than 20 years, the desired improvements in air quality have not yet been achieved (Hoofman et al. 2016). Therefore, the effectiveness of Euro-emission standards for road transport can be into questioned.

4.2.EMISSIONS FROM RESIDENTIAL WOOD COMBUSTION

In recent years, problems caused by atmospheric particles in urban air are receiving increasing attention. A significant source of winter emissions is the burning of firewood as a renewable and affordable fuel source for household heating. The combustion of firewood contributes to the growth of total atmospheric particles and is a noticeable source in temperate and cold climate regions.

Combustion of firewood for heating generation is a major source of emissions of fine particulate matter, poly aromatic hydrocarbon (PAHs) and certain gaseous pollutants, such as volatile organic compounds (VOCs). Particulate emissions from the combustion of firewood contain a large proportion of carbon materials consisting of elemental carbon and organic carbon as a result of incomplete combustion.

Wood combustion emissions can be divided into two groups: complete combustion emissions (oxidized pollutants) and incomplete combustion emissions (unburned pollutants). In the combustion of wood, combustion products are carbon dioxide, carbon monoxide, nitrogen, oxygen, water, hydrogen, unburned hydrocarbons, sulfur dioxide, nitrogen oxides, hydrocarbons and particles in solid and liquid phases.

When wood is completely burnt, the products of combustion are only carbon dioxide, water and a small number of particles (most are organic). The most important parameters for complete combustion are high combustion temperature, sufficient air intake and adequate mixing of combustion air and fuel. Complete combustion can only happen under ideal conditions. In practice this never happens and incomplete combustion is always present which results in incomplete combustion emissions.

As a result of the incomplete combustion of wood, there is large particle formation and poses a serious environmental problem. Incomplete combustion particulate emissions can occur as smoke, soot, or condensed heavy hydrocarbons (tar drops).

Soot is an agglomeration of carbon particles, which results from the lack of oxygen in the flame zone. There are mainly three sources of primary particle origin and formation mechanisms that are directly emitted into the ambient air by wood-burning devices: soot particles, organic particles, and ash particles.

The first fine particles formed in the process of combustion of wood are the particles of soot formed by hydrocarbons in the flames. PM emissions are increasingly dominated by soot during incomplete combustion with a lack of air at higher temperatures (800-1000) ° C.

Solid fuels such as wood contain a significant amount of ash in the form of inorganic elements. During combustion, these inorganic species in the combustion products produce ash. Most of the ash formed in the wood-burning process remains at the bottom of the furnace. Additionally, a small portion of the ashes called fly ash along with the exhaust gases are carried to the environment. Fly ash particulate emissions can be characterized as aerosols (<1µm in size) and coarse fly ash particles. The particle size of this coarse fly ash ranges from a few micrometers to about 200 micrometers (M. Obaidullah, 2012).

5. HEALTH AND ECOLOGICAL EFFECTS FORM PARTICULATE MATTER

Atmospheric particles are considered one of the most serious threats to human health, the environment and climate.

Atmospheric particles can persist for a long time in the atmosphere and are subject to dispersion and transport processes and thus cause harmful effects on human health and the environment. The harmful effects include health problems with the respiration and respiratory system, worsening of the existing respiratory and cardiovascular diseases, changes in the body's immune system, lung tissue damage, carcinogenesis and premature death. These effects are more expressed with the elderly and children (L. A. Jimonda 2012).

The term suspended particles refers to particles of all range. However, from the human health point of view, the most important are particles up to 10 μm (PM_{10}) and particles up to 2.5 μm ($\text{PM}_{2.5}$). PM_{10} can be easily transported through the upper respiratory tract causing direct health hazards. Recent research, however, focuses attention on fine suspended $\text{PM}_{2.5}$ particles due to their ability to penetrate deeper into the respiratory system (L. A. Jimonda 2012).

Suspended particles are associated with many negative effects on human health. A large number of studies conducted in different parts of the world indicate an important link between motor vehicle emissions and increased symptoms of asthma and rhinitis (Rai, 2015). Reports of the health effects of diesel engine exhaust particulate matter indicate that short-term exposure to diesel engine particles is associated with respiratory health effects such as allergic inflammation and asthma-typical symptoms, while long-term exposure is associated with an increased risk of lung cancer (US EPA, 2002).

Ambient particles resulting from the combustion of coal for generating electricity in thermal power plants contain directly emitted suspended particles and secondary particles formed by reactions of gaseous pollutants such as sulfur and nitrogen. In many industrialized countries coal combustion particles are a significant contributor to total air pollution and the health effects of exposure to this source have been extensively investigated (Hime et al. 2018).

Particles resulting from the combustion of wood for heating households significantly contribute to the increase in ambient air concentrations (Trajkovski et al. 2017). Much of the information regarding the health effects associated with firewood burning is related to respiratory problems especially with children, while the impact of cardiovascular disease is smaller (Brauer et al. 2007).

Air pollution caused by traffic, coal combustion for power generation and combustion in industrial processes can be important contributors to asthma and other respiratory problems. The wood burning, leaves and other agriculture debris creates large amounts of PM_{10} and other air pollutants that can aggravate asthma and cause breathing problems. Symptoms such as tachycardia and elevated blood pressure have been reported as a consequence of heavy metal pollution (Kampa and Castanas 2007).

6. EXPERIMENTAL RESEARCH ON PM₁₀

Five measuring locations have been designated to measure the mass concentration of suspended particles in the urban part of Bitola. The locations are set at relatively equal distances along the city. The results of the mass concentration measurements will be used to determine the impact of the major sources of PM₁₀ particles in ambient air. The main sources of this research relate to the combustion of biomass for household heating and the impact of traffic. For this purpose, three measurement sites have been designated to determine the impact of biomass combustion and two measurement sites to determine the impact of traffic.

Three sites are designated to measure the concentrations of suspended particles and determine the impact of biomass combustion. The first measuring site is located at the suburb Brusnic - location 1 - the urban periphery of the city. This location is characterized by no traffic and no industrial combustion processes near the site of measurement. As for the particle emissions, this location's characteristics is the burning of biomass in winter.

The second measuring site is located in the Park near the Primary School Stiv Naumov - location 2 and is located in the urban part of the city in the downtown area and is characterized by a relatively low frequency of traffic and no industrial combustion processes near the measuring site. Also in winter, this site is characterized by particle emissions from biomass combustion.

The third location is located in the lower peripheral part of the town in the suburb Streliste - location 3. This location besides biomass combustion is also affected by the traffic emissions as it is near the boulevard entrance to the city with a high frequency of vehicles.

Two measuring locations have been established for determining the impact of traffic. One is located on the ring road near Vero1 - location 4. This location is characterized by the high frequency of vehicles throughout the day. At this location, the vehicles are mostly passenger. Near this location, there are no industrial or other combustion processes during the measurement. The second location is located on the ring road near the gas station Bit-Oil - location 5. This site is characterized by a high frequency of vehicles of all types (passenger, trucks and buses).

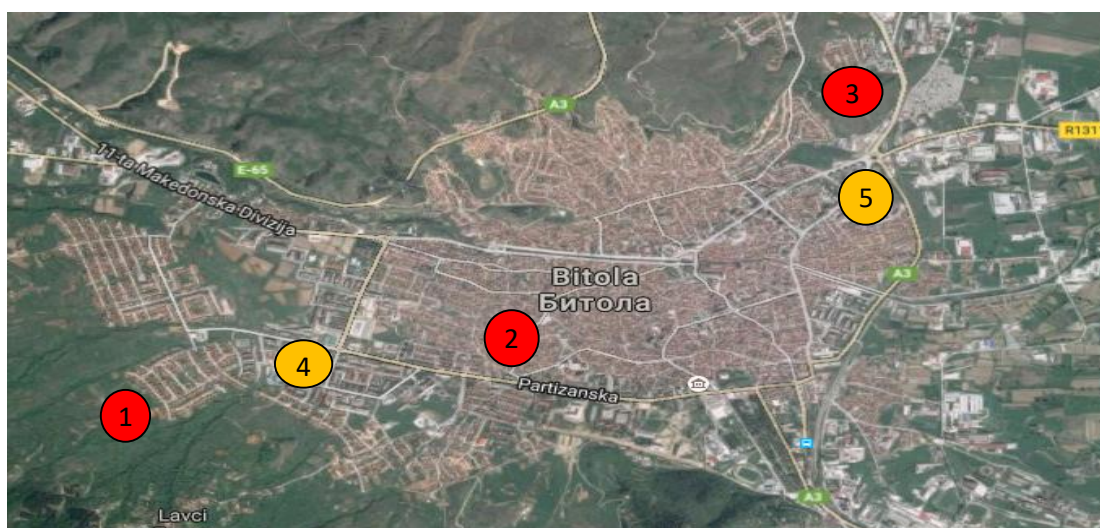


Figure 6.1: View of measuring locations for determination of PM₁₀ concentration in Bitola

8. CONCLUSION

Firewood combustion proved to be the dominant source of PM₁₀ emissions in total emissions in the urban part of the city of Bitola during winter. The average values of the measurements indicate consistently high concentrations during the measurement period at all measuring locations.

From the individual measurements, it was noticed that the highest concentration of PM₁₀ was measured at Site 3 (suburb Streliste) in November 2016, and the value is 4 times higher than the concentrations measured at Site 1 (suburb Brusnicka) in the same month. At measuring location 2, the measured concentrations are 3 times higher than at measuring location 1 in the same period. In October 2016 which is not heating season the measured values at location 2 are about 2.8 times higher than at location 1 and at location 3 also the measured values are about 2.5 times higher than at location 1. Unlike location 1, at locations 2 and 3, higher values were measured indicating that the downtown area had higher particulate emissions from the periphery of the city during the entire measurement period.

In terms of particle distribution, it is evident that during the heating season of Site 1 in February 2017, the fine particles up to 2.5 and 1 µm increased by about 4 times compared to October when the heating season of the site has not yet started at the same location. During this period, particles up to 10 µm in size increased by 15%. The ratios at locations 2 and 3 for the same period are similar. During the heating season at the three locations in the particle distribution, we have an increased percentage of fine particles indicating the presence of combustion as the dominant source. In the non-heating season, the particle distribution is within the model of Watson and Chow (2000), which points to soil and road dust as the dominant source. Particle allocation at locations 4 and 5 with increased vehicle frequency resulted in an increase in PM₁₀ coarse particles by nearly 10%, while fine particles up to PM_{2.5} increased by 8% and PM₁ remained approximately equal, indicating that that resuspended dust is a dominant source of exhaust gases at traffic frequencies.

From the ratio of particle size PM_{2.5} / PM₁₀ to the period when it's not heating season in October 2016, it can be noticed that at all locations the values are approximately equal and relatively low compared to the values obtained in the heating season in November 2016, February and March 2017, indicating a higher percentage of PM₁₀ coarse particles. That is, the resuspended dust from the soil and roads is more dominant source of combustion. When the heating season starts, in most locations, an increase in this ratio is observed, indicating an increased value of fine particles as a result of combustion.

If we look at the differences of the measurement locations, according the measured values over the entire measurement period, the impact of PM₁₀ emissions from biomass combustion on total PM₁₀ emissions in the air is 58-70% at site 1, 60-65% at site 2 and 69-76% at site 3. From these differences, it can be concluded that combustion during the heating season for home heating increased PM₁₀ concentrations by at least 58% and is a dominant source of particles in winter.

From the differences shown by locations 1,2 and 4 in October 2016 when the heating season has not started, it results that the particle concentrations of measuring location 4 which has bigger vehicle frequency are higher than location 1 for about 60% and 65% respectively at site 2. The impact of traffic has led to an increase of PM₁₀ concentration for at least 60% and is a predominant particle swarm in the urban part of the city during the non-heating season.

Conceptual modeling is a modeling techniques, that does not require modern and relatively expensive sampling equipment and complex software systems for processing and is therefore easily applicable. Unlike other

modeling techniques it can lead to larger deviations in the presented analyses. New modeling techniques such as receptor models are the most widely used modern sampling systems and with chemical analysis source identification is more accurate. The use of new modeling techniques and the use of other equipment open the possibility for further research in this area.