Interreg – IPA CBC

EUROPEAN UNION

CCI 2014 TC 16 I5CB 009

ERFC

DELIVERABLE 3.1

"IDENTIFICATION OF THE EMISSION SOURCES"

Interreg – IPA CBC

EUROPEAN UNION

CCI 2014 TC 16 I5CB 009

TRAP

The project is co-funded by the European Union and by National Funds of the participating countries

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1. Environmental Centre of Western Macedonia
2. Municipality of Florina
3. European Regional Framework for Cooperation
4. Center for Climate Change
5. Ministry of Environment and Physical Planning

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Table of Contents

Table of Figures

Table of Tables

1. About the Project

Real-time air pollution monitoring is now more necessary than ever before. Air pollution is one of the most significant factors posing threat to the health of people worldwide. It is associated with a range of diseases, symptoms and conditions that impact quality of human life. According to WHO, outdoor air pollution was responsible for the deaths of some 3,7 million people under the age of 60 around the world in 2012, representing 6.7% of the global disease burden while indoor and outdoor air pollution combined are among the largest risks to health worldwide. Apart from population air quality has negative impacts on natural environment and biodiversity. The main sources of air pollution at participants countries are caused by industrial activities, transportation and heating systems. Air quality problems from industrial sources mainly concern areas with thermoelectrical power stations and industrial units located close to residential areas. Other sources (e.g. transport and dust from deserts) and natural conditions (e.g. local topography and climatic conditions) also worsen urban air quality. Local meteorological conditions and topography have a major impact on air quality and contribute to the generation of air pollution episodes. Air quality is also strongly influenced by pollutants trapped due to thermal inversions caused from sea/land breezes and thermal internal boundary layers. Strategic objective of TRAP project is the creation of an ICT application integrating air quality monitoring with health impact indexes in the participating countries and Cross Border area. The project aims to evaluate current situation regarding air quality in partners' areas, install air quality monitoring stations and create public health indexes for assessing air quality impact on human health and natural environment. TRAP will extend the already established network of air quality to both countries and cross-border area by establishing 4 new monitoring stations (2 per country). Moreover, health indicators based on the effects of air pollution on human health, will be displayed on the project's website. TRAP is expected to go a step beyond the current state of the art in air quality by correlating air quality and health and providing an integrated air quality – health index ICT tool which will be accessible by stakeholders and general public (vulnerable target groups).

More information about the project is available at:

http://www.ipa-cbc-programme.eu/approved-project/75/

https://trap-project.eu/

2. Objectives of the Deliverable 3.1

Deliverable 3.1 entitled as "Identification of emissions sources", has as main target to be identified the air pollution sources of the intervention area of the Project using bibliographical sources. It belongs to the Work Package 3 "Air quality impact assessment" in which the targets are to be identified the air pollution sources, to be created a regional emission inventory, to be identified the contribution of the emissions and to be developed an air quality plan for the intervention area. The major pollutants from which the emission sources are identified in this study are: PM_{10}, $PM_{2.5}$, NO_x, O_3, CO, SO_x. In the first chapter is presented the TRAP project. In the second chapter are included the objectives of the deliverable. Third chapter gives a description about the Policy context. In the Introduction Chapter (4^{th}) are referred the basic information points of the subject such as: emissions sources and air pollutants, Gothenburg Protocol to the Long-range Transboundary Air Pollution (LRTAP), air quality standards and background information for the area of intervention. In the next chapter (5^{th}), there is a more analytical description about the basic air pollutants and in the 6^{th} chapter is presented the identification of emission sources of air pollution in the Greater Area of Thessaloniki. The final chapter (7^{th}) presents a discussion about the results of the study.

Figure 1- The cooperation area of Interreg – IPA CBC "Greece – North Macedonia" 2014-2020

Source: http://www.ipa-cbc-programme.eu

3. Policy Context

Several control measures have been implemented since the 1990s, mainly related to traffic management and vehicle emission reductions, as well as reduction in fossil fuel use due to energy efficiency improvements or interventions in favour of alternative fuels and renewable energy sources. These resulted in reductions of atmospheric pollution and changes in emission source contributions; however, the financial crisis which started on 2009 has significantly altered this picture (Diapouli, et al., 2017).

On particulate matter the European Union has legislated a series of limits and guidelines on controlling the mass concentration of particles in ambient air. Specifically, a 24-h mean concentration of 50 μg/m^3 not to be exceeded more than 35 times per year and an annual mean concentration of 40 μg / m^3 have been imposed for particles with aerodynamic diameter below 10 μm, PM_{10} (Council Directive 1999/30/EC). In addition, the recent directive on ambient air (Council Directive 2008/50/EC) has imposed an annual concentration cap of 25 for particles with diameter below 2.5, while Member Stated are required to reduce exposure to $PM_{2.5}$ in urban areas by an average of 20% by 2020 based on 2010 levels (Kassomenos, et al., 2011).

Local authorities and policy-makers throughout the European Union are therefore asked to perform the necessary monitoring and apply environmental policies, in order to comply with the above limits and objectives. Under this perspective, characterizing the physical and chemical properties of the particles, as well as identifying major particle sources and quantifying their contribution are some of the issues that need to be addressed in urban areas in the European Union (Chaloulakou, et al., 2003).

4. Introduction

Millions of people worldwide live in cities where air pollution is so severe that hundreds of thousands die prematurely every year and dozens of others experience chronic or other acute illnesses (Elsom, 2014). Coal is still the major source of energy, constituting about 75% of all energy sources. Consequently, air pollution in China predominantly consists of coal smoke, with suspended particulate matter (PM) and sulfur dioxide (SO2) as the principal air pollutants. In terms of PM and SO2, China may have the worst air pollution level in the world (Kan, et al., 2012). In large cities, however, with the rapid increase in the number of motor vehicles, air pollution has gradually changed from the conventional coal combustion type to the mixed coal combustion/ motor vehicle emission type. Also, the characteristics of outdoor air pollution (e.g. air pollution level, and fate and transport of pollutants), meteorological conditions and socio-demographic patterns in China are different from North America and Western Europe (Chen, et al., 2011).

4.1. Emission sources and air pollutants

According to European Environment Agency (2017), air pollutants are emitted from a range of both man-made and natural sources including:

- Burning of fossil fuels in electricity generation, transport, industry and households
- Industrial processes and solvent use, for example in the chemical and mining industries
- Agriculture
- Waste treatment
- Natural sources, including volcanic eruptions, windblown dust, sea-salt spray and emissions of volatile organic compounds from plants.

Air pollutants can be released directly into the atmosphere (primary emissions) or can form as a result of chemical interaction involving precursor substances. The air pollutant emissions cause air pollution, however, reductions in emissions do not always automatically result in similar cuts in concentrations. There are complex links between air pollutant emissions and air quality. These include emission heights, chemical transformations, reactions to sunlight, additional natural and hemispheric contributions and the impact of weather and topography. Significant cuts in emissions are essential for improving air quality (European Environment Agency, 2017) (European Environment Agency, 2018).

Figure 2 - Sources of air pollution in Europe

Source: European Environment Agency, 2017

4.2. Gothenburg Protocol to the Long-range Transboundary Air Pollution (LRTAP)

The Gothenburg Protocol to the Long-range Transboundary Air Pollution (LRTAP) Convention sets emission ceilings. Parties to the convention must reduce their emissions to these levels. These ceilings, for 2010 and after, are for the pollutants nitrogen oxides (NOx), non-methane volatile organic compounds (NMVOCs), Sulphur oxides (SOx) and ammonia (NH3). In addition to the ceilings for individual countries, the protocol also specifies ceilings for the EU, which is a Party to the protocol in its own right. The protocol was amended in 2012. The ceilings set for 2010 and years thereafter are still in place, but the amended protocol also specifies new emission reduction commitments in terms of percentage reductions by 2020, relative to base year 2005. Parties are also encouraged to report primary particulate matter (PM) and black carbon (BC) emissions, in line with the revised emission-reporting guidelines. The EU ratified the amended protocol in 2017 (European Environment Agency, 2018).

4.3. Air quality standards

Humans can be adversely affected by exposure to air pollutants in ambient air. In response, the European Union had developed an extensive body of legislation which establishes health standards and objectives for a number of pollutants present in the air. These standards and objectives are summarized in the table below. These apply over differing periods of time because the observed health impacts associated with the various pollutants occur over different exposure times (European Commission, 2018).

Table 1 - Air quality standards

Pollutant	Concentration	Averaging period	Legal nature	Permitted exceedances each year
Fine particles ($PM_{2.5}$)	25 µg/m3***	1 year	Target value to be met as of 1.1.2010 Limit value to be met as of 1.1.2015	n/a
Sulphur dioxide (SO2)	350 µg/m3	1 hour	Limit value to be met as of 1.1.2005	24
	125 µg/m3	24 hours	Limit value to be met as of 1.1.2005	3
Nitrogen dioxide (NO2)	200 µg/m3	1 hour	Limit value to be met as of 1.1.2010	18
	40 µg/m3	1 year	Limit value to be met as of 1.1.2010 *	n/a
PM_{10}	50 µg/m3	24 hours	Limit value to be met as of 1.1.2005 **	35
	40 µg/m3	1 year	Limit value to be met as of 1.1.2005 **	n/a
Lead (Pb)	0.5 µg/m3	1 year	Limit value to be met as of 1.1.2005 (or 1.1.2010 in the immediate vicinity of specific, notified industrial sources; and a 1.0 µg/m3 limit value applied from 1.1.2005 to 31.12.2009)	n/a
Carbon monoxide (CO)	10 mg/m3	Maximum daily 8 hour mean	Limit value to be met as of 1.1.2005	n/a
Benzene	5 µg/m3	1 year	Limit value to be met as of 1.1.2010**	n/a
Ozone	120 µg/m3	Maximum daily 8 hour mean	Target value to be met as of 1.1.2010	25 days averaged over 3 years
Arsenic (As)	6 ng/m3	1 year	Target value to be met as of 31.12.2012	n/a
Cadmium (Cd)	5 ng/m3	1 year	Target value to be met as of 31.12.2012	n/a
Nickel (Ni)	20 ng/m3	1 year	Target value to be met as of 31.12.2012	n/a
Polycyclic Aromatic Hydrocarbons	1 ng/m3 (expressed as concentration of Benzo(a)pyrene)	1 year	Target value to be met as of 31.12.2012	n/a

Source: European Commission, 2018

Directive 2008/50/EC introduced additional $PM_{2.5}$ objectives targeting the exposure of the population to fine particles. These objectives are set at national level and are based on the average exposure indicator (AEI). This is determined as a 3-year running annual mean $PM_{2.5}$ concentration averaged over the selected monitoring stations in agglomerations and larger urban areas, set in urban background locations to best assess the $PM_{2.5}$ exposure of the general population.

Table 2 - Air quality standards for PM2.5 in a 3-year average

Title	Metric	Averaging period	Legal nature	Permitted exceedances each year
$PM_{2.5}$ Exposure concentration obligation	20 µg/m3 (AEI)	Based on 3-year average	Legally binding in 2015 (years 2013,2014,2015)	n/a
$PM_{2.5}$ Exposure reduction target	Percentage reduction* + all measures to reach 18 µg/m3 (AEI)	Based on 3-year average	Reduction to be attained where possible in 2020, determined on the basis of the value of exposure indicator in 2010	n/a

Source: European Commission, 2018

* Depending on the value of AEI in 2010, a percentage reduction requirement (0,10,15, or 20%) is set in the Directive. If AEI in 2010 is assessed to be over 22 µg/m^3, all appropriate measures need to be taken to achieve 18 µg/m^3 by 2020.

**Under Directive 2008/50/EU, the Member State was able to apply for an extension until three years after the date of entry into force of the new Directive (i.e. May 2011) in a specific zone. The request was subject to assessment by the Commission. In such cases within the time extension period the limit value applies at the level of the limit value + maximum margin of tolerance (35 days at 75µg/m^3 for daily PM_{10} limit value, 48 µg/m^3 for annual Pm_{10} limit value).

***Standard introduced by Directive 2008/50/EC

4.4. Background information for the area of Thessaloniki

Thessaloniki is suffering severe air pollution problems during the last decades, mostly related to PM_{10} levels. The air quality, combined with the strong hot season of the Mediterranean climate, is known to be one of the worse in Europe, especially in summer, and leads to serious sanitary concerns.

Thessaloniki is the second largest Greek city and has the second largest commercial port. The city is characterized by intense vehicular traffic in the center and several industrial units located in the West, North-West and North. Prevailing winds are weak (1-2 m/s) strongly influenced by the sea breeze, while frequently occurring calms result to inadequate dispersion of atmospheric pollutants and short-range transport processes (Diapouli, et al., 2017).

Thessaloniki is located on the northern part of Greece, while the plundering of permanent residents is almost 800,000 (Vouitsis, et al., 2011) and significant industrial activity (Kassomenos, et al., 2011). It is a major transportation hub for south-eastern Europe and despite the present economic situation of Greece it still hosts 20% of the country's industrial activity (Chatzimichailidis, et al., 2014). It is considered one of the most polluted cities in Europe in terms of particulate matter (PM) concentration as PM_{10} levels exceed the limits of atmospheric pollution set by the European Union (Vouitsis, et al., 2011). The factors that affect this phenomenon are the climate, the geography, the high traffic density, the industrial activity and the maritime transport. Specifically, in the winter is also added the domestic heating as a responsible factor. The climate is temperate with weak prevailing winds (sea breeze) and frequent calms resulting to inefficient dispersion of atmospheric pollutants and short-range transport, in the city area (Vouitsis, et al., 2011).

The mountain of Hortiatis, reaching 1200 approximately, is located at the east of the area, while some more hills are located at the north. In the west, the area is flat and allows the connection of the city with the rest of the Macedonia mainland. Finally, the rather shallow gulf of Thermaikos is in the south (Kassomenos, et al., 2011). Metropolitan Urban areas in Greece have been known to suffer from poor air quality, due to variety of emission sources, topography and climatic conditions favouring the accumulation of pollution. While several control measures have been implemented since the 1990s, resulting in reductions of atmospheric pollution and changes in emission source contributions, the financial crisis which started in 2009 has significantly altered this picture.

5. Basic Pollutants

Table 3 - Sources of primary air pollutants

Pollutant	**Sources**
Carbon monoxide	Incomplete burning of fossil fuels
	Tobacco smoke
Hydrocarbons	Incomplete burning of fossil fuels
	Tobacco burning
	Chemicals
Particulates	Burning fossil fuels
	Farming operations
	Construction operations
	Industrial wastes
	Building demolition
Sulphur dioxide	Burning fossil fuels
	Smelting ore
Nitrogen compounds	Burning fossil fuels

(Mabahwi, et al., 2014)

5.1. Particulate matter

Particulate matter emitted directly into the air:

- PM with a diameter greater than 2.5 microns ($PM_{2.5}$ also called fine particulate matter)
- PM with a diameter greater than 10 microns (PM_{10})
- BC, the most strongly light-absorbing component of PM

According to Kassomenos, et al. (2015) particles can be either primary (i.e. emitted directly from the source, such as road traffic and resuspended dust), or secondary (i.e. formed within the atmosphere through chemical reactions). According to Diapouli et al. (2017) particulate matter is a key atmospheric pollutant for local and regional air quality, with major implications to human health, degradation of natural and built environment and climate change. Urban

centers have been known to present increased PM levels due to the high degree of urbanization and dense vehicular traffic. PM is a complex mixture of chemical species, originating from a variety of sources, both anthropogenic (traffic, industrial activities and combustion processes in general) and natural (wind-blown dust, sea salt

, forest fires, volcanic eruptions), and may be emitted as primary pollutant or formed as a secondary product through atmospheric processes from gaseous precursors.

PM can be identified in many types, so the fate depends on the particular chemical composition of the PM being considered. The organic types of PM may be destroyed or transformed into other chemicals while in the atmosphere by the hydroxyl radical. Many others settle onto earth and water and are transformed there by microorganisms or by several other reactions (Hill, 2010)

Dust and dirt raised by vehicles or wind from farms (fertilizer, dried manure, or dried crop residues) are the major sources of PM_{10}. A big contribution comes from roads, unpaved and paved and construction sites which release large amounts of dust. PM_{10} also includes pollen. Burning fossil fuels have a 6% contribution to the emissions of PM_{10}. A natural PM source found in coastal areas is sea salts, which have high levels of chloride and can corrode local buildings and statuary (Hill, 2010).

Most $PM_{2.5}$ and the even tinier ultra-fines originate from fossil-fuel combustion, (e.g. diesel motor vehicles, electric power plants) and industrial operations such as steel mills emitting sulfur dioxide (Hill, 2010).

Power plants and other incinerators also produce fly ash (whose fine particles contain carbon, hydrocarbons, metal oxides, and silicon dioxide). Silicon dioxide in window glass is benign, but in fly ash or fine blowing sand, it is dangerous. Fly ash particles also trap dioxins formed during combustion.

Locales with large numbers of inefficiently burning wood stoves contribute to particle levels. Rural areas generate airborne particles when burning biomass.

The most organic material, by an efficient combustion, is converted to carbon dioxide and water, with little particulate matter and soot while more particles form when combustion is inefficient (Hill, 2010).

5.2. Nitrogen oxide (NO_x)

A group of highly reactive gases, some of which play an essential role in pollution problems, concludes oxides of nitrogen (namely photochemical smog, ground level ozone, acid rain, acidification and eutrophication of water and soils, visibility impairment, and damage to natural ecosystems, cultural heritage and crops) (Zandaryaa & Buckens, 2014), although trees do not emit the NOx that interact with VOCs to form ozone (Hill, 2010). Major pollution concern is in nitric oxide (NO) and nitrogen dioxide (NO_2). NO is formed at high temperature, but immediately turns into NO2 in the atmosphere. Nitrous oxide N_2O is a greenhouse gas. The dominant part of anthropogenic emissions is attributed to combustion and transportation (Zandaryaa & Buckens, 2014) that emit more than 50% of NOx (Hill, 2010). The NOx are produced by combustion of nitrogen-containing fuels and high-temperature oxidation of molecular nitrogen in air (Zandaryaa & Buckens, 2014).

Emissions of NOx are hard to control because they result from a reaction between atmospheric oxygen and nitrogen at high temperatures (Hill, 2010).

Seven different oxides of nitrogen are known to occur in the atmosphere. Nitric oxide (NO) and nitrogen dioxide (NO_2), together referred to as nitrogen oxides (NOx), are the major air pollutants, for their role in atmospheric oxidation/reduction reactions with ozone, forming hazardous or detrimental substances. The most current unit of measure for NOx is NO_2 mass equivalent (mg per Nm^3 (normal cubic meter)) of NO plus NO_2 (Zandaryaa & Buckens, 2014).

Nitric oxide is a colorless, tasteless, toxic, nonflammable gas, almost insoluble in water. At the point of discharge, NO is the predominant form of NOx. In the atmosphere, it is readily converted to the much more harmful NO_2 (Zandaryaa & Buckens, 2014).

Nitrogen dioxide is a yellowish-orange to reddish-brown toxic, strongly oxidizing and corrosive gas with a characteristic pungent, irritating odor. In the atmosphere a portion of NO_2 is converted into nitric acid (HNO_3) and ammonium salts. Nitrate aerosol is removed from the atmosphere through wet or dry deposition processes. At high concentrations, both NO and NO_2 can be toxic to humans, animals, plants and other living organisms and in the atmosphere react with other pollutants, forming toxic and irritating chemicals, such as peroxyacetal nitrate (PAN) and fine atmospheric particles that cause respiratory and bronchial health problems (Zandaryaa & Buckens, 2014).

The main environmental problems associated with NOx emissions are (Zandaryaa & Buckens, 2014):

- photochemical smog;
- ground level ozone;
- acid rain;
- visibility impairment;
- acidification and eutrophication of water and soils; and
- damage to natural ecosystems, cultural heritage and crops

5.3. Ground-level Ozone (O_3)

Ozone is a dangerous pollutant, in the ambient air, but it operates as a protective shield from the ultraviolent radiation rays by the stratosphere (Mabahwi, et al., 2014). Tropospheric ozone (O_3) is a photochemical air pollutant which is not directly emitted in the atmosphere and thus, the ecological problems related to O_3 formation from complex chemical reactions, can't be easily solved (Kolev, et al., 2011). It is formed in the atmosphere by the reaction of volatile organic compounds (VOCs) and nitrogen oxides (NOx) in the presence of sunlight (Liu, et al., 2017). Both NOx and VOCs originate from either anthropogenic source sectors (e.g., industrial and transport sectors) or from biogenic sources. Both anthropogenic and biogenic sources play a role in O_3 formation and accumulation. While the impact of biogenic emissions on surface O_3 levels has been thoroughly examined in many studies, the role of various anthropogenic emission source sectors in the production of ground-level O_3 has not been extensively investigated (Poupkou, et al., 2008). Ozone acts as a strong photochemical oxidant and causes serious health problems and damages materials and ecosystems. The effects of ozone in human health are more dangerous in children than in adults. Ozone in the presence of SO_2 and NO_2 lead to increased corrosion on building materials like steel, zinc, copper, aluminium and bronze (Leeuw, 2000). Ozone exposure of ecosystems and agricultural crops results in visible foliar injury and in reductions in crop yield and seed production. For vegetation a long-term, growing season averaged exposure rather than an episodic exposure is generally of concern. Adverse effects on vegetation can be noted at relatively low ozone levels (Leeuw, 2000).

Table 4 - Ozone levels and air quality

Concentration	**Air quality**
0.000-0.050 ppm	**Good:** No health impacts expected.
0.51-0.100 ppm	**Moderate:** Unusually sensitive people should consider limiting prolonged exertion outdoors
0.101-0.150 ppm	**Unhealth for sensitive groups.** Active children and adults, those with respiratory problems should avoid prolonged outdoor exertion.
0.151–0.200 ppm	**Unhealthful.** Active children and adults, and those with respiratory disease should follow the advice for 0.101–150. Others, especially children, should limit prolonged outdoor exertion.
0.201–0.300 ppm	**Very unhealthful.** Active children and adults, and people with respiratory disease should avoid all outdoor exertion. Others, especially children, should limit outdoor exertion.

(Hill, 2010)

5.4. Carbon Monoxide (CO)

Carbon monoxide intoxication is one of the most common inhalative poisoning worldwide which can lead to high morbidity and mortality involving multiple organ systems (Reumuth, et al., 2018). Carbon monoxide (CO) is a gas, which by its nature is colourless, odourless and tasteless, making it an invisible threat. It arises from the incomplete burning of hydrocarbons (Reumuth, et al., 2018) and the incomplete combustion of fossil fuels such as motor vehicles (Liu, et al., 2018). Carbon monoxide is formed anywhere that a carbon-containing material is burned. Thus, CO exposure happens anywhere that combustion occurs (Hill, 2010).

- Major sources in urban areas, motor vehicles emit up to 95% of CO. Drivers stalled in traffic or driving in highly congested areas can have high CO exposure. So, can traffic control personnel, mechanics working inside garages, and parking garage attendants.
- Lesser sources Cigarette smoke contains CO. Individuals exposed to CO on the job who also smoke, increase their risk of adverse effects.
- Facilities burning coal, natural gas, or biomass are CO sources.
- Burning biomass (wood, dried manure, other dried vegetation) for cooking, heating, and light is a CO source. These lesser sources can be significant in certain locales or situations.
- Environmental fate CO can persist in the atmosphere for one to two months before it is converted to CO_2 – thus CO contributes to CO_2 in the atmosphere (Hill, 2010).

Its affinity to hemoglobin molecules is roughly 240 times higher than that of oxygen (O_2), leading when present to the formation of carboxyhemoglobin by replacing the bounded O_2 (Reumuth, et al., 2018). In urban areas, CO is primarily generated by motor vehicle emission. Previously, epidemiologic studies have reported short-term associations of ambient CO with daily mortality and morbidity from cardiovascular diseases (Chen, et al., 2011). The process that leads to negative health results in human health by the presence of carbon monoxide is referred below. The blood protein hemoglobin picks up oxygen in the lungs and transports it to the tissues where it releases the oxygen. When carbon monoxide (CO) is inhaled, it binds to hemoglobin much more strongly than oxygen and so blocks the oxygen binding, lowering the amount of oxygen available to the body. Too much blocking of hemoglobin by CO can lead to death. Smaller doses can cause headache, nausea, and other flu-like symptoms. Chronic exposure is implicated in the development of heart disease (Hill, 2010).

5.5. Sulfur oxide (SO_x)

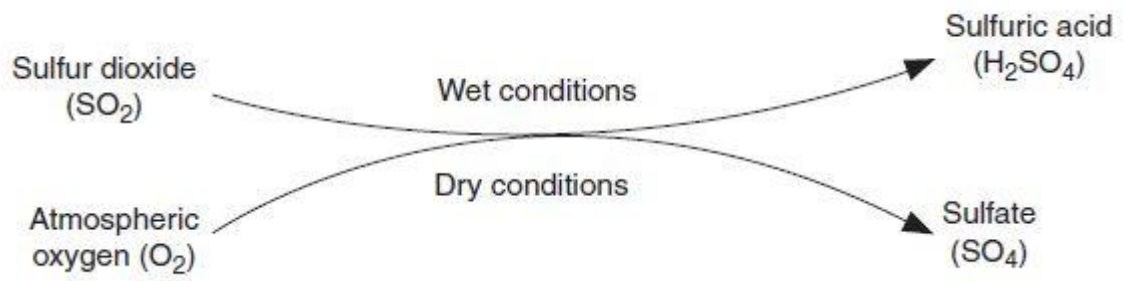
Sulfur dioxide is the main source of atmospheric sulfur. Significant amounts of SO_2 near the ground have a direct effect on human health, leading to respiratory symptoms, difficulty in breathing, even premature deaths in extreme cases, and inhibit plant growth. SO_2 oxidizes in the troposphere to form sulfuric acid (H_2SO_4) which is mostly deposited as acid rain, being responsible for the contamination of soil and for the degradation of marble monuments. Moreover, SO_2 plays an important role in cloud formation physics, leading to clouds of high reflectivity. In the stratosphere SO_2 is also oxidized and combines with water to H_2SO_4/H_2O

aerosols (Bekki, 1995). These aerosols scatter solar radiation and absorb long-wave radiation, causing heating in the stratospheric region and net cooling at the Earth's surface. Anthropogenic SO_2 and by extension sulfur emissions, are estimated to be larger than emissions from natural sources. The estimated difference between anthropogenic and natural sulfur emissions varies, since major natural sources like volcanic eruptions are occasional emitters (Georgoulias, et al., 2009).

Studies have only focused on the analysis of near surface concentration measurements showing a very strong seasonal variation with higher values during the cold period (November–February) and much lower values during the hot period (May–August) (Georgoulias, et al., 2009).

Sulfuric acid and sulfate are aerosols. These wash out with rain, or slowly settle out by gravity. Both are acidic deposition.

Figure 3 - Transformations of SO_2 and O_2



(Hill, 2010)

Human sources In the industrialized Northern Hemisphere, human activities produce five times more SO_2 than do natural sources.Worldwide, the figure is about two times as much. Sulfur is found in all raw materials – including coal, crude oil, and gas. Sulfur is converted to SO_2 when fossil fuels are burned. The EPA reports that 87% of the SO_2 released into US air is attributable to fuel combustion. In 2002, electric utilities burning fossil fuels produced about two-thirds of the anthropogenic SO_2 in the United States. The worst offenders are utilities burning high-sulfur coal.

- Many metal ores including aluminum, copper, zinc, lead, and iron contain sulfur, so metal smelters emit SO_2.
- As oil is processed into gasoline, SO_2 is also formed. However, sulfur is more readily removed from petroleum during its refining than is the case for coal; thus, motor vehicles account for a lower percentage of SO_2 emissions.

Other sources of SO_2 emissions include industrial facilities such as petroleum refineries and cement manufacturing operations. Natural sources Most SO_2 is anthropogenic, but many natural sources exist including sea water, marine plankton, bacteria, plants, and geothermal emissions. Erupting volcanoes are a major but periodic source. In 1991, the Filipino volcano Mt. Pinatubo ejected about 20 million tons (18.1 million tonnes) of SO_2 into the atmosphere. This huge quantity is believed to have been responsible for cooling the Earth’s climate for several years thereafter.

Table 5 - Sulfur dioxide, nitrogen oxides, and global change

Issue	Role of sulfur dioxide
Acid deposition	Sulfur dioxide emissions are converted in the atmosphere to sulfate and sulfuric acid – major contributors to acid deposition
Stratospheric ozone depletion	Volcanic inject sulfur dioxide into the stratosphere where it is converted to particles. Analogous to ice particles at the poles, these provide surfaces on which ozone depleting reactions occur.
Global climate change	Sulfur dioxide is converted to sulfate or sulfuric acid particles. These have an "anti-greenhouse" effect. They do so by absorbing part of the sun's radiation, preventing it from reaching and warming the Earth's surface.
Issue	Role of nitrogen oxides (NOx)
Acid rain, stratosphere ozone depletion, and climate change	NOx emissions are converted in the atmosphere to nitrate and nitric acid, which contribute to the problems shown on the left.
Ground-level ozone	NOx but not sulfur dioxide, is converted to ground-level ozone.
Nutrient pollution	NOx is converted to nitrate and nitric acid. After deposition to earth and water, these can "over-fertilize" water leading to eutrophication.

(Hill, 2010)

6. Identification of emission sources of air pollution: Case study of Thessaloniki

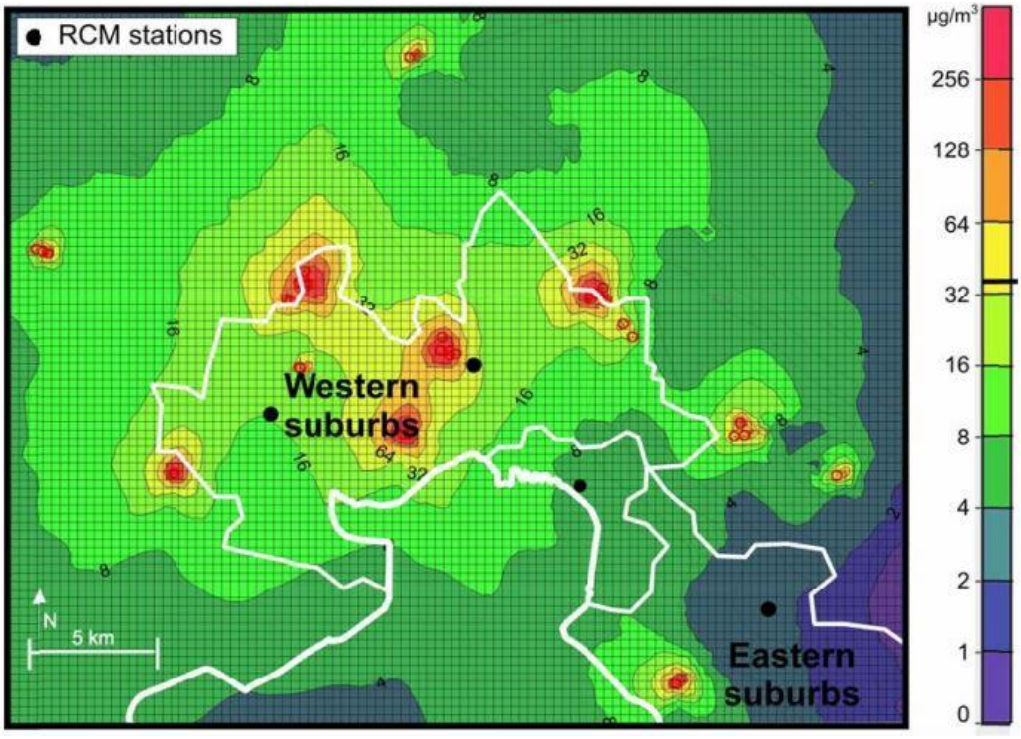
6.1. Industry

According to Petrakakis et al., (2008), the main air pollution sources are the industrial activities taking place in the western part of the city and the automobiles, with more than 500,000 registered in the area. The central heating also plays a significant role during the cold period of the year (October-April) (Petrakakis, et al., 2008).

An extended flat area at the west and north-west of Thessaloniki hosts the largest part of the industrial activity in the region. It stretches over 20 km in a bowl formed by low hills facing a bay that opens into Thermaikos gulf (Moussiopoulos, et al., 2009).

In a study (Kakosimos, et al., 2011) that deals with the assessment of the primary particulate matter pollution from industrial activities close to the city of Thessaloniki, a PM_{10} emission sources inventory was prepared with the main industrial plants of the area and the yearly and monthly average concentrations between 2003 and 2007. The atmospheric dispersion was estimated by AEROMOD (by EPA U.S.). Furthermore, it was estimated that the primary PM_{10} industrial contribution on the western suburbs of the city is approximately 30%, while in the city center about 7%. It was also illustrated that the frequent south-west winds present higher concentration levels than the strong north ones. Using the air dispersion model, it was calculated the mean annual and monthly concentrations of PM_{10} from 2003 to 2007.

Figure 4 - Concentrations levels PM10 in the Greater Thessaloniki Area (2007) and the RCM stations



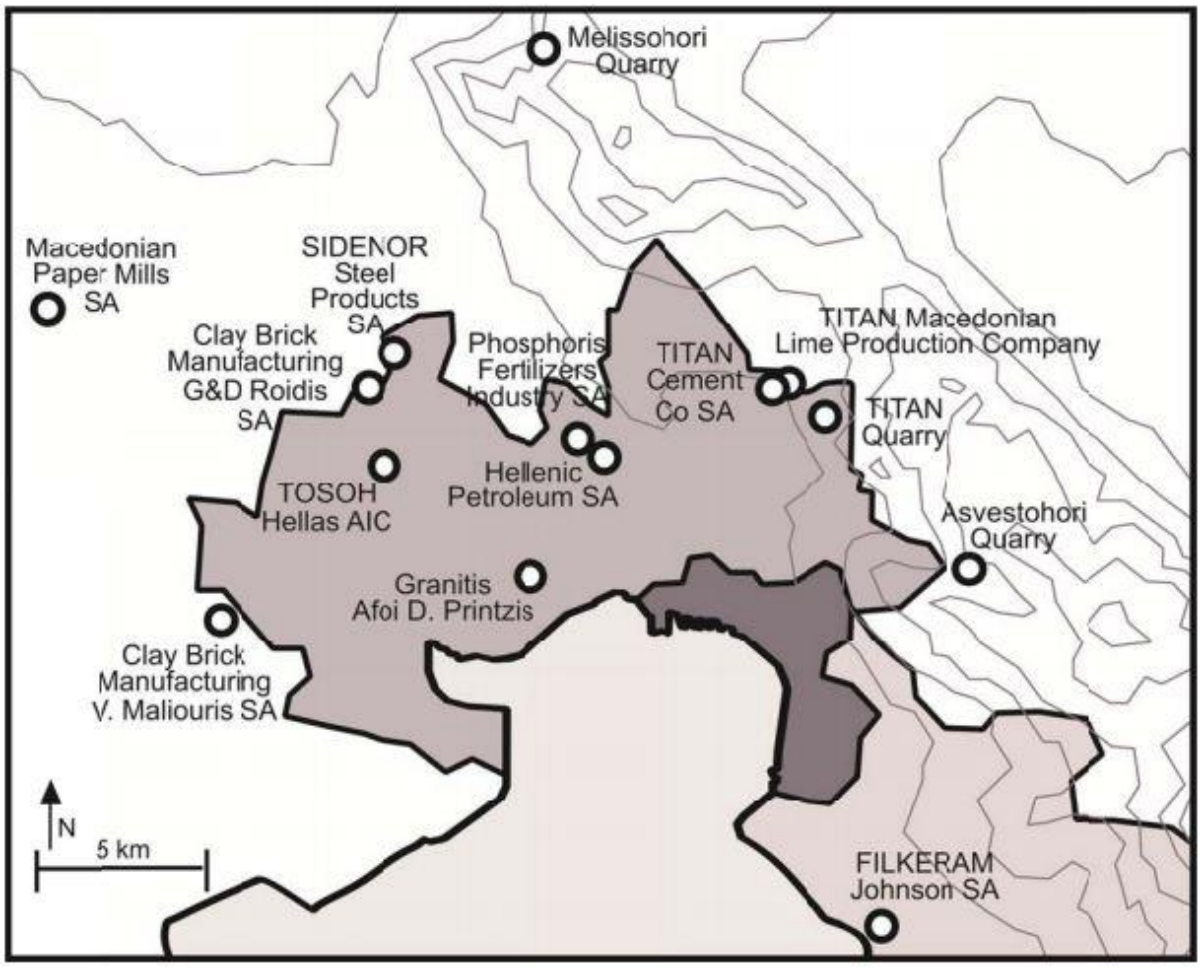
(Kakosimos, et al., 2011)

Observations from the above map:

- Nearly to the emission sources the concentration's values approach a maximum of 130 μg/m^3. In the center of the city the contribution of industrial activities reaches in a level of 8 μg/m^3. In the west suburbs of the city, the contribution of industrial activities ranges from 16 to 65. These high values are explained due to the fact that the west side of the city is surrounded by the industrial air pollution sources.
- Chortiatis mountain, north-western of the greater Thessaloniki area, creates a natural border which acts like an obstacle to the air pollution masses. The contribution of industrial activities to the eastern suburbs have a limitation of 4 μg/m^3 (Kakosimos, et al., n.d.).

According to Kakosimos et al., (2011), the major industry pollution sources for the area of Thessaloniki are depicted to the following map:

Figure 5 - The 13 major industrial sources in the Greater Area of Thessaloniki (2011)



(Kakosimos, et al., 2011)

The map is referring to 10 small and large industrial activities plus 3 quarries. These activities varied from processes, stacks and open areas. All the plants and quarries were asked to provide details of their PM_{10} emissions or sufficient data for the purposes of the study of Kakosimos et al. (2011). Details of the emission sources are given in Table 6 and their location is shown in Figure 5.

Bourliva et. al., (2018), found from road dust samples that were collected during 2010-2011 in Thessaloniki, that high loadings of Cd, Mn and Ni were observed, suggesting mixed sources of oil/fuel combustion and industrial activities. Cadmium is commonly used in lubricating oil and coal combustion could release Cd into the environment including road dust. Industrial activities such as oil refining and petrochemical production, could produce enhanced concentrations of Ni in road dust (Bourliva , et al., 2018).

Table 6 - Details of the emission sources from 13 industrial activities in the Greater Area of Thessaloniki

	Industrial source	**Type of source**	**Area (m^2)**	**Emission Factor ($kg \cdot t^{-1}$)**	**Emission Rate ($g \cdot s^{-1}$)**	**Rate/area x 10^6 ($g \cdot s^{-1} m^{-2}$)**	**Production in Tones/Year**
1	Macedonian Paper Mills SA	Process	351x154	1.0	2.89	54	120000 Coated Paper
		Stack	-	0.2	0.58	-	
		Open area	97x85	-	0.05	6.6	
		Open area	85x96	-	0.05	6.6	
2	Phosphoric Fertilizers Industry SA	Process					Status Shutdown
3	TOSOH Hellas AIC	Process	252x202	-	-	19[2]	electrolytic manganese dioxide (EMD), 60000
		Stack	-	-	0.27[2]	-	
4	Hellenic Petroleum SA	Process	230x190	-	0.0003	0.001	polypropylene production 220000
		Process	137x241	-	0.1	0.24	
		Process	112x80	-	-	1.01	
		Process	100x80	-	-	0.54	

5	TITAN Quarry	Open area	213x238	-	1.01	20	
		Open area	143x190	-	0.54	20	
6	Asvestohori Quarry	Open area	1143x217	-	4.96	20	
		Open area	108x130	-	0.28	20	
		Open area	157x116	-	0.36	20	
		Open area	355x201	-	1.43	20	
7	Melissohori Quarry	Open area	251x435	-	-	2.18	
8	TITAN Cement Co. SA	Process	262x283	0.05	2.74	37	Cement 1500000
		Open area	93x77	-	0.05	6.6	
		Open area	54x52	-	0.02	6.6	
		Open area	34x39	-	0.008	6.6	
9	TITAN Macedonian Lime Production Company	Process	99x227	0.94	37.1	1700	Lime production 80000
		Stack	-	0.2	7.8	-	
		Open area	59x38	-	0.15	6.6	
10	SIDENOR Steel Products SA	Process	373x119	2.76	53.3	1200	Steel production 600000
		Stack	-	0.01	0.2	-	
		Stack	-	0.3	6.15	-	

		Stack	-	0.3	6.15	-	
		Open area	144x120	-	0.11	6.6	
		Open area	157x120	-	0.12	6.6	
11	Afoi D. Printzi Granitis	Process	105x120	3.0	4.7	370	
12	Clay brick Maliouris manufacturing: V. SA	Process	123x119	3.0	19.0	100	
		Open area	64x80	-	0.03	6.6	
13	Brick manufacturing G&D Roidis SA	Process	133x72	1.5	2.3	240	
		Stack	-	0.5	0.79	-	
		Stack	-	0.5	0.79	-	
		Stack	-	0.5	0.79	-	
		Open area	76x72	-	0.04	6.6	

(Kakosimos, et al., 2011)

The amounts of production for each industry has been identified by the official websites of the industries for the latest years.

6.2. Production of energy

The city of Thessaloniki is very sensitive to transboundary transport of gaseous pollutants and aerosols from Central and Eastern Europe (Katragkou, et al., 2009). North-eastern flows increase the mass of PM with a possible explanation of the transport of dust from local and regional sources with most probable being the power plants of Maritsa, Bulgaria (Katragkou, et al., 2009).

The cold period from January to March can be attributed to the wide use of oil burning heating systems during the Greek winters. The peak of July–August can be attributed to lignite-burning power stations to the WSW of the city and trans-boundary transport from SO_2 sources in Bulgaria, Romania and the former Yugoslavia (Georgoulias, et al., 2009).

In addition, 4 biogas plants are established in the area of Thessaloniki, producing electricity. The plants are presented in the following table.

Table 7 Biogas Plants in the area of Thessaloniki

Biogas Plant	**Biomass use in tons (Full production) ***	**Power (MWe)**
Helector Thessaloniki Tagarades Landfill	125000	5 MWe
EYATH	65000	2.6 MWe
kIEFER TEK	21100	0.844 MWe
BIOGAS Lagadas SA	25000	1 MWe
TOTAL	**236100**	**9.44 MWe**

The amounts of production for each Biogas Plant has been identified by the official websites of the industries for the latest years.

*Biomass Capacity is calculated using the established Power production by the assumption that for 1 MWe production it is needed 25000 tons of biomass (manure) under a 70% humidity environment.

6.3. Transportation

Road traffic is a major pollution problem for modern cities. PM is produced both by mechanical means (road abrasion, tire-wear and break lining), and by fuel combustion (Chatzimichailidis, et al., 2014).

Moussiopoulos et al., (2009) using a network of air monitoring stations have led to the next results. After 2001, SO_2 concentrations have stabilised at considerably lower levels due to de-sulphurization of fuels which resulted in the minimization of traffic-related SO_2 emissions. Furthermore, CO concentrations since 2000 have a significant reduction due to the introduction of the 3-way catalyst converter which resulted in the minimization of traffic related CO emissions. In addition, NO_2 concentration level has a reduction since 1995 but it is not as significant as it should have been because of the number of vehicles that has increased considerably over this period. During 2001–2006, NO_2 annual average concentrations stabilised at levels close to the EU limit (40 μg/m_3) in most stations, except Ag. Sofias Sqr which approaches 60 μg/m_3 (Moussiopoulos, et al., 2009).

According to Kassomenos et al., (2011), both PM_{10} and $PM_{2.5}$ are positively correlated to the NOx concentrations, reflecting their common road traffic origin, as a conclusion by data that were analysed and they were ejected from two air pollution stations which were located in two different sites in Thessaloniki. It is noted that the frequency of diesel taxis and buses circulating in the city centre is very high, as these are the only means of public transportation in Thessaloniki (Kassomenos, et al., 2011).

Road dust is a composite environmental medium consisting mainly of solid matter (composed of both natural and anthropogenic constituents) and its emissions have been widely documented to be the main components of atmospheric particulate matter (PM) (Bourliva , et al., 2018). Manoli et al., (2003) found that road dust dominated the coarse particle fraction (3.0-10μm) in the center of Thessaloniki, accounting for 57% to its ambient levels, while it was also an important contributor (28%) to the fine size fraction (<3.0 μm) (Bourliva , et al., 2018).

Bourliva et. al. (2018) found (from samples in different areas in Thessaloniki) that from March 2010 to March 2011, some important components of road dust exhibit significant loading for Cu, Cr and moderate loading for Pb. The presence of widely known as traffic-related elements reveals that their possible source is vehicular emissions (Bourliva , et al., 2018). The high

loading that was found for Cu known as the most abundant element in brake lining materials, along with the high loading for Cr and Pb, typically used as frictional additives in brake friction materials (Bourliva , et al., 2018). So, an important source of PM are the brake lining materials and the brake friction materials and in Thessaloniki play an important role to air pollution emissions by transportation. Finally, Zn was also an element that was found which can be found in road dust and has been linked with tire and brake wear, diesel exhaust emissions and corrosion safety fence (Bourliva , et al., 2018).

Table 8 - Motor vehicles in operation, by category and use of the region of Thessaloniki for the year 2018

Type of Vehicle	**Year**	**Quantity**
Passenger	Total	525056
	Private use	522601
	Public use	2455
Trucks	Total	98040
	Private use	94060
	Public use	3980
Buses	Total	2397
Motorcycles	Total	131103
	Freight private use	130230
	Freight public use	238
Nuts	EL522	

(Hellenic Statistical Authority , 2018)

Port activities in the greater Thessaloniki area is a serious reason for particulate matter pollution which is caused by stevedoring activities, such as lifting and stowing dry bulk cargo (minerals, scrap metals, grain, pet coke and other) (Chatzimichailidis, et al., 2014). The port of Thessaloniki is one of the largest Greek seaports and one of the largest ports in the Aegean Sea basin, with a total annual traffic capacity of 16 million tones. (Valavanidis, et al., 2015)

Another possible emissions source is the International Airport “Macedonia” which is located south-east of the city’s center (Chatzimichailidis, et al., 2014).

6.4. Heating

During 2012-2014, wood began to be employed for heating as a result of the economic crisis. Burnt wood particles contribute further to the already over polluted greater area of Thessaloniki. There are no valid data on how much wood is burned during the winter months (Chatzimichailidis, et al., 2014).

Areas close to the Caspian and Aral seas over Kazakhstan are well documented as arid regions with potential to contribute significantly to the total dust budget and have the potential to add particle mass in the city of Thessaloniki, especially from May to August. Also, measurements have shown that easterly winds especially during summer months are responsible for long-range transport of particulate matter and identify the PM mass as product of biomass burning (Katragkou, et al., 2009).

A result after the analysis of data that were used in the study of Kassomenos et. al., (2011), was that the relatively high coefficients between PM_{10}-SO_2 and $PM_{2.5}$ – SO_2 during the cold season reflect the oil combustion for domestic heating and its impact on SO_2 and PM emissions.

According to Diapouli, et al., (2017), during 2011-2012 Thessaloniki had a significant increase of carbonaceous particle concentrations during the cold period, especially in the residential urban background sites, pointed toward domestic heating and more particularly wood (biomass) burning as a significant source. After the execution of the PMF analysis by Diapouli et al., (2017), the analysis supported the finding that biomass burning was the largest contributing source. Also, a major contributing source was the secondary aerosol formation. Finally, at the urban traffic site, vehicular traffic (exhaust and non-exhaust emissions) was the source with the highest contributions (Diapouli, et al., 2017).

A study (Saffari, et al., 2013) has found a 30% increase in the concentration of fine particle ($PM_{2.5}$) emissions associated with wood smoke from residential heating in 2012 and 2013, with implications for the health of local residents. The increase in organic and elemental carbon (OC/EC) ratios during the cold period may associated with biomass burning aerosols and suggests intense use of fireplaces and wood stoves at the residential areas in Greece (Saffari, et al., 2013). Since 2011 citizens of Thessaloniki switched burning of fuel for domestic heating to burning of uncontrolled wood and biomass. This phenomenon appears to have caused significant air quality deterioration. Burning of distilled fuel oil has been the most popular

method of residential heating in Thessaloniki during recent years, while wood burning has also been used in a small number of houses as a supplementary heating source, mainly in the suburbs of Thessaloniki. Wood smoke and biomass burning have been identified as potential sources of urban $PM_{2.5}$ emissions in several studies (Saffari, et al., 2013).

The results revealed there had been a substantial increase in fine particle emissions over the two years, with the total $PM_{2.5}$ (particles smaller than 2.5 micrometres in diameter) increasing by 30% from 2012 to 2013, from 26 to 36 micrograms per cubic metre. Levels of $PM_{2.5}$ were twice as high in the evenings compared with the mornings in 2013, illustrating the effects of higher wood smoke emissions from residential heating in the evening. Organic matter was the most common component of the $PM_{2.5}$ samples. Evening samples contained a higher proportion of organic matter (74%) compared with the morning samples (58%), again suggesting more wood and biomass were used for heating in the evening. Potassium concentrations were two to three times higher in 2013 compared with 2012 and were also two to three times higher in the evening compared to the morning. Analysis revealed the potassium most likely came from wood smoke. Furthermore, the concentrations of vanadium and nickel, which indicate combustion of residential fuel oil and industrial activity, were 30-40% lower in 2013 compared with 2012. The concentrations of the chemicals levoglucosan, mannosan, and galactosan, commonly found in wood smoke, were four to six times higher in 2013 compared with 2012 and the concentrations of all three compounds were three to four times higher in the evening compared with the morning (Saffari, et al., 2013).

In 2007 the calculated OC/EC ratios were similar (during the cold and warm period), something that proves that residential biomass burning was not so pronounced before the financial crisis (Diapouli, et al., 2017)

Significant trans-boundary SO_2 transport has not been recorded and even though a fraction of the SO_2 can be transported above the boundary layer, the winter conditions are not the most favorable for such a transport. However, there is a very strong seasonal variability with a winter peak due to the use of heating systems and lower values during the summer (Georgoulias, et al., 2009).

The drastic decreases in SO_2 level during the warm part of the year can be attributed to both the shutting-off of central heating burners and faster atmospheric removal the warm part of the year. Both these factors contribute also to the observed decrease of benzene and toluene during summertime (Kourtidis, et al., 2002). It can be inferred that a strong seasonal variability could

be partly responsible for the satisfying correlation between ground-based and satellite observations (Georgoulias, et al., 2009).

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6.5. Other sources

An important component of PM is coming from the sea salt (Diapouli, et al., 2017) and as a result the sea spray contributes constantly a small part in the whole problem (Chatzimichailidis, et al., 2014). Water bodies like sea release airborne particulate matter with physical and mechanical ways. In coastal environments such as Thessaloniki area, particles originating from sea are expected to be a significant source of atmospheric pollution, still with high temporal variation (Chatzimichailidis, et al., 2014).

Mineral dust contribution, according to a study which investigated the air pollution in Thessaloniki during 2007 and 2011-2012, was significant for the PM_{10} fraction. Its concentration was larger during the warm season when dry conditions favour soil dust resuspension (Diapouli, et al., 2017).

Agricultural activities affect mainly the rural areas of the greater Thessaloniki area (regardless the fact that produce huge amounts of PM, in the months they occur (Chatzimichailidis, et al., 2014). There are areas of irrigated land with a variety of different cultivations, such as wheat, cereals, corn, vineyards and other products. The agricultural activities, such as tilling and harvesting can be a major particulate matter pollution factor, on the months they are carried out. Each cultivation type has a different timeline of activities and emission factors (Chatzimichailidis, et al., 2014).

Measurements with lidar system situated in Thessaloniki combined with back-trajectory analysis and studies with satellite data attributed part of particulate load to natural sources, namely transport of dust from Sahara and maritime aerosols (Katragkou, et al., 2009). During spring the central part of northern Africa appears to be a potential source of PM (Katragkou, et al., 2009).

7. Discussion – Results

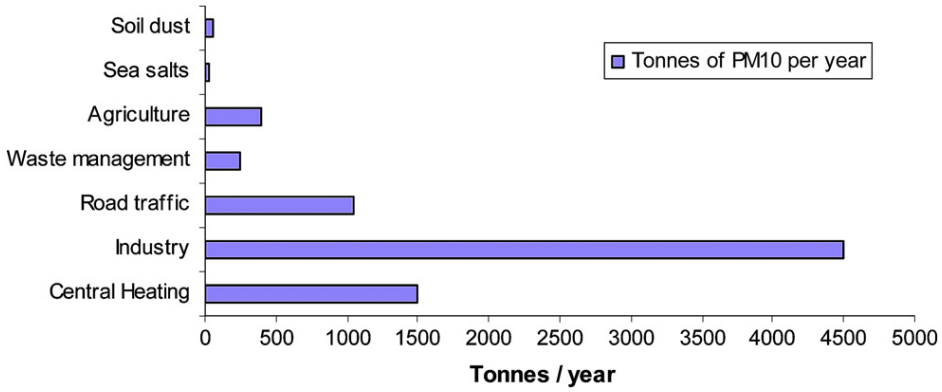
On 2009, Mousiopoulos et al. referred that vehicle and industrial emissions are the two main sources of air pollutants in the greater Thessaloniki area.

Conclusions about the year 2002 in the greater Thessaloniki area: Road transport is the dominant source of NO_x emissions, with industrial activities also having a considerable share. Road transport is also responsible for most NMVOC emissions, with solvent use and industry to impose a significant burden. Industrial activities dominate PM_{10} emissions with road transport and space heating also having a considerable share (Moussiopoulos, et al., 2009).

According to Chatzimichailidis et al., (2014), and their study with data from 2003 to 2007, the main causes of local high levels of PM are the industrial activities, followed by the car traffic. The PM_{10} from residential heating significantly impacts air quality during winter months and its contribution tends to grow following the increased use of wood for heating. Agricultural activities affect mainly the rural areas of the greater Thessaloniki area (regardless the fact that produce huge amounts of PM, in the months they occur. Finally, the sea spray contributes constantly a small part in the whole problem (Chatzimichailidis, et al., 2014).

During 2006-2008, the main particulate pollution sources, namely road traffic, natural or other fugitive sources such as dust, soil resuspension and sea-spray, industrial activity, oil combustion for domestic heating and photochemical activity were identified (Kassomenos, et al., 2011).

Figure 6 - The main PM10 pollution sources in the greater area of Thessaloniki during 2006-2008



(Kassomenos, et al., 2011)

In conclusion, Thessaloniki has a low air quality especially in the western suburbs due to the industrial activities and the big use of automobiles in the center of the city. Furthermore, a significant factor during the winter months is the use of organic material for domestic heating.

Figure 7 - Emission sources in the Greater Thessaloniki Area

The bibliographical sources that were examined in this study mentioned that economic crisis was the major reason for the additional use of organic material for domestic heating. Meteorological factors play a significant role in the whole problem. Other factors that act with a negative way are the transportation (port and airport), the agricultural activities, the mineral dust, the sea salts, the dust from Sahara, the oil combustion for central heating, the photochemical activities, the soil dust resuspension, the construction activities, the neighbor regions and countries lignite-burning power stations for energy production plants (trans-boundary air pollution from Bulgaria, Romania and former Yugoslavia).

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