MACEDONIAN AIR QUALITY ASSESSMENT REPORT FOR THE PERIOD 2005–2015

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1. INTRODUCTION

Air pollution in the country is a cause of serious concern as the limit values set for protection of human health, especially for particulate matter, are exceeded significantly. The situation is the worst in the largest urban settlements, especially in Skopje and Tetovo. Several sources and causes of the severe air quality problems are identified and may vary within and between cities. Household heating with wood during the winter period causes severe air quality problems in densely populated residential areas as major part of the households in the country still use wood as the primary source of heating. Road traffic is also a source of air pollution in urban locations, due to high volumes of traffic and partially old and poorly maintained vehicle fleet. Energy production and industry can have local impacts to the air quality, especially in the vicinities of old industrial installations lacking modern emission reduction systems. Furthermore, development of densely built urban areas including decrease of green areas in cities can affect the formation of pollution.

The air quality in the country has been monitored with modern methodologies for more than 10 years. In this report, the trends in the air pollution levels during the period of 2005-2015 are analysed. For some pollutants, including sulphur dioxide, there can be seen a clear decreasing trend in the concentration levels during the ten year period. For other pollutants, the decreasing trend is only slight or the concentration levels have remained in the same level. The particulate matter concentrations have remained in a very high level during the period with no significant declining trend.

According to the legislation, measures to improve the air quality must be implemented when limit values of pollutant concentrations for protection of human health are exceeded. In order to successfully decrease the emissions to air, efforts by the central and local authorities, companies and citizens alike are needed. For successful improvement of air quality, the air quality policies will need to be harmonized with other policies, such as energy, climate and transport policies in national and local level.

The country should urgently take actions to improve the air quality with commencement of implementation of the measures defined in the national and local air quality improvement plans. The measures related to the decreasing of the emissions from domestic heating and road traffic should take priority due to the significant impact to air quality. In addition, the environmental permitting process for those installations with high emissions to air should be fast tracked.

For more than ten years the European Union has supported the country in strengthening the air quality administration. EU has provided support for improving the air quality monitoring network and calibration and chemical laboratories as well as air quality data management systems. Through three Twinning projects, support has been provided to strengthen the air quality management capacities including the development of air quality legislation, and improvement of air quality monitoring, assessments, data management and reporting and emission inventories. The current Twinning project ‘Further strengthening the capacities for effective implementation of the acquis in the field of air quality’ concentrates on further improving the capacities of the Ministry of Environment and Physical Planning, municipalities and the Institute of Public Health. One of the focus areas of the current project is the development of local level air quality improvement plans. This air quality assessment report was prepared as a part of the Twinning project activities.

2. EXECUTIVE SUMMARY

Currently the Macedonian air pollution levels are high for certain polluting substances and can be harmful to human health. Air quality can be improved with effective actions that reduce emissions which have the largest impact to the pollutant concentrations. Based on the air quality measurements, some improvement of air quality in recent years can be seen, especially related to the sulphur dioxide concentrations as result of emission reduction actions. Thus, continuous long-term efforts for cleaner air make a difference.

Air pollution levels are known to be high

Air quality monitoring has been performed in the country since 1965 with passive sampling of some pollutants. In late 1990's the monitoring was modernized and also spatially expanded to cover the whole territory of the country. With the State Air Quality Monitoring network the levels of air pollutants are continuously monitored in accordance with the EU air quality directives and national legislation. The monitoring program includes continuous measurements of sulphur dioxide (SO_2), nitrogen oxides (NO_x/NO_2), particulate matter (PM_{10} and $PM_{2.5}$), carbon monoxide (CO) and ozone (O_3) at seventeen monitoring sites in different parts of the country. Volatile organic compounds (VOCs), polycyclic aromatic hydrocarbons (PAHs) and heavy metals (HM) have been measured with short-term campaigns. Also dispersion model calculations have been prepared and applied to evaluate the air quality in relation to the limit values.

Since the autumn 2012 the results of all continuous measurements are available to the public on real time through an internet portal (http://airquality.moepp.gov.mk).

Based on the decade long monitoring of air pollution concentrations, it is now well known that urban air pollution is a significant problem in the country. In addition, even in rural areas the pollutant concentrations can be at times elevated, especially concerning ozone and fine particles ($PM_{2.5}$). National and European air quality standards set for the protection of human health are exceeded considerably, widely and for prolonged periods. The mountainous terrain and meteorological conditions in the country cause extra challenges for the air pollution management. Furthermore, easy access to the air pollution information has substantially increased the public awareness and raised serious and justified concerns of the air pollution.

Figure 1. In the winter time the particulate concentrations can be high effecting also the visibility (Photo: Pia Anttila).

Sources and causes of air pollution

Many sources and causes for the severe air quality problem have been identified. The causes and underlying processes may vary within and between cities, but nowhere there is a one simple explanation for the currently detected pollution levels.

Obvious source category for air pollutants that most cities share is the old and poorly maintained vehicle fleet. At national level, approximately half of the passenger cars and buses are old belonging to the high-emission vehicle categories. Congested traffic flows and poorly developed or totally missing public transport worsen the situation.

Inefficient small scale combustion in residential fireplaces and boilers together with the use of poor quality fuels like moist or treated wood or even waste materials cause problems especially in densely populated residential areas. Major part of the households in the country still consume fuel wood as the primary source of heat. Deteriorated and limited district heating systems and high price of electricity further increase the use of solid fuels for residential heating.

Energy production and distribution systems are often aged, inefficient, unreliable and polluting. Domestic electricity production relies mostly on poor-quality lignite in old thermal power plants. Combined heat and power generation as well as wind and hydro power generation are still rare in the country.

The absence of proper waste management and recycling systems increase the amount uncontrolled waste combustion such as open burning of household waste. Also agricultural biomass burning may cause local air quality problems.

Limited economic growth has closed numerous smokestack industries in the country during the past decades. Furthermore after 2006, the remaining manufacturing industries were subjected to the environmental permits or to the environmental impact assessments processes and some already have started the installation of best available technologies for reduction of air pollution. Nevertheless, there are still existing manufacturing industries that are old-fashioned without proper air pollution control, resulting risk of harmful impacts to air quality.

Focus on effective actions to reduce pollution

The decade long air quality monitoring now provides valid information of the magnitude of the problem, and therefore future priorities in air quality management should be targeted to the reduction of air pollution. The available monitoring data gives an opportunity to track trends in ambient air concentrations and identification of sources of the pollutants. These are useful in deciding where the reduction measures are most needed and likely to have the greatest effect.

3. AIR QUALITY POLICY AND STRATEGIES

Due to the active international cooperation during the last decade, the capacities in air quality management have been significantly strengthened in the country. Current air quality pollution levels and their causes are well known. There is reliable air quality data available for public and decision makers. However, air quality improvement requires high-level political commitment. Air quality policies and strategies for cleaner air have been created to set the objectives for future actions.

The objective of the European air pollution policy is "to achieve levels of air quality that do not result in unacceptable impacts on, and risks to, human health and the environment." In many countries in Europe further efforts are needed to achieve this objective.

European Community has adopted policies limiting the emissions from industry, traffic, energy production plants and agriculture, in order to limit the air pollution responsible for adverse impacts on human health and environment. The European directives for air quality protection and regulating emissions are presented in Annex I.

The European Union directives are transposed to the national legislation (Annex II). The main laws are the Law on Environment and the Law on Ambient Air Quality. The latter regulates in detail the following issues:

- air quality limit and target values;
- emission limit values from different sources of pollution;
- air quality management and assessments, air quality and air emission monitoring;
- planning of the air quality protection;
- supervision and competent bodies for the implementation of legislation.

In addition to the primary legislation, a number of bylaws have been prepared and are already in force.

In 2012 the first two national strategic documents for air quality protection were adopted by the Government. The National plan for protection of the ambient air presents the situation concerning air quality, defines the measures at national level for protection and improvement of the quality of ambient air per sector (energy, industry, traffic, agriculture and waste), and defines all the relevant institutions responsible for their implementation for the period 2013-2018.

National program for the gradual reduction of the quantities of emissions of the certain pollutants was prepared for the period 2012-2020. In this program, measures at national level to reduce emission of the air pollutants, sulphur dioxide, nitrogen oxides, ammonia, volatile organic compounds (VOCs), TSP and carbon monoxide to air, are identified. Additionally the program sets national emission projections for the period 2015 to 2020 for these pollutants.

In addition to the national plans, the local air quality improvement plans have to be prepared in the municipalities where the exceedances of the air quality limit values have been observed. These plans represent the practical actions to be taken to improve the air quality on local level. Until now the local air quality improvement plans have been prepared for the Skopje agglomeration and municipalities of Tetovo and Bitola.

4. METHODOLOGIES AND DATA USED IN THE ASSESSMENT

Reliable long-term air quality monitoring data and advanced air quality assessment tools are used to support decision making and development of air quality improvement plans. Air quality monitoring is the basis for the assessment of levels of air pollution, supported by emission inventories and dispersion modelling and source apportionment studies.

This assessment builds on the air quality monitoring data accumulated over the last ten years, updated emission inventories since 1990, results from a number of intensive measurement campaigns and case studies where dispersion of pollutants in the atmosphere is mathematically modelled. Also meteorological, geographical and land use data are used. Furthermore, sources of air pollution have been approximated with statistical modelling of the observed ambient pollutant concentrations.

Based on the emission information only, the impact of certain emission source to the air quality cannot be identified. For example, power plants may emit large amounts of pollutants but as the emissions are mostly released to air through high stacks, their impact on ground level concentrations, at human breathing height, remains minor. Sources like road traffic, however, may emit less pollutants but since the emissions are released near the ground level their impact to the air quality in the breathing level can be significant.

Prevailing weather conditions can decrease or increase the pollutant concentrations. Strong winds can rapidly transport the pollutants for hundreds of kilometres, whereas in calm weather, pollutants can accumulate around the source of the release. Complex terrain or blocks of buildings may further enhance the accumulation of the pollution. Some pollutants remain in the atmosphere days or even weeks and can be transported hundreds of kilometres, while others may transform to other substances or get deposited to the ground in minutes or hours.

4.1. Air quality monitoring

Air quality measurements are used to assess the air quality, particularly at those locations where exceedances of limit values are to be expected. The legislation defines the minimum requirements for the air quality measurements. The national air quality monitoring network includes 17 stations which measure air quality continuously in different parts of the country (Figure 2). The number of monitoring stations is sufficient according to the requirements defined in the national legislation and European directives. Atmospheric concentrations of the basic pollutants SO_2, CO, NO_2, PM_{10}, $PM_{2.5}$ and O_3 are measured and the information of the levels of the pollution is in real-time available for the authorities and the public. This network of monitoring stations is managed by the Ministry of Environment and Physical Planning (MEPP). For this assessment the air quality monitoring data from years 2005-2015 was used.

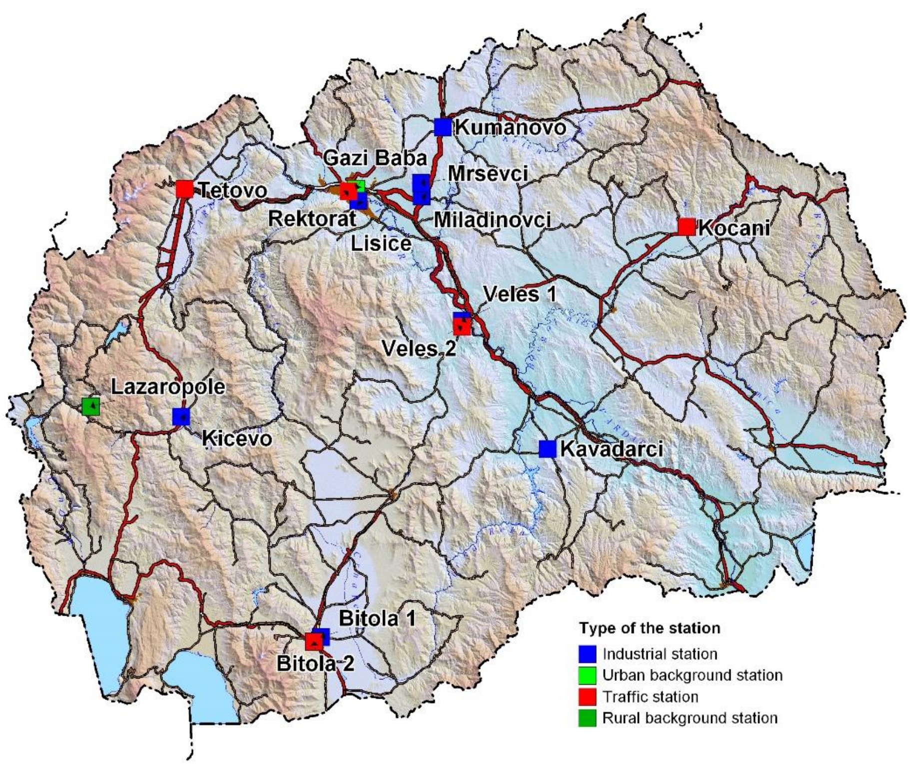

Figure 2. State air quality monitoring network managed by MEPP.

In order to achieve reliable results the monitoring equipment must be maintained and calibrated regularly. Unfortunately, the lack of regular maintenance of the instruments and the lack of spare parts for the aging instruments have decreased the data coverage, especially during the most recent years.

In addition to the continuous routine monitoring, the results of a measurement campaign of heavy metals (HM) and polycyclic aromatic hydrocarbons (PAHs) are used in this assessment. Campaign was organized in the urban background site in Karpos, Skopje, between August 2015 and March 2016. The sampling was done every three days by MEPP and the chemical analyses were carried out in the accredited laboratories in Finland and Serbia.

This data was also used to estimate the potential source categories of pollution by multivariate statistical modelling (Positive Matrix Factorization).

The results from a measurement campaign for volatile organic compounds (VOCs), which was conducted in Skopje in 2011-2012 were also used in the assessment.

4.2. Emission inventories

Emission information is important basis for national policies to reduce emissions. MEPP compiles annually the national emission inventory for SO_x, NO_x, CO, non-methane VOCs, NH_3, particulates, heavy metals and persistent organic pollutants (POPs). Emission inventories are based on the relevant activity data (fuel consumption, quantity of products, used consumables, number of livestock, arable land etc.), and the compliance monitoring conducted and reported by the major installations. Default emission factors are taken from the EMEP/EEA Guidebook (EMEP/EEA, 2013). In 2016, the inventories for the period 1990–2014 were assessed and recalculated by MEPP.

Emission inventory results are reported to UNECE on an annual basis. All inventories are available on the following website http://cdr.eionet.europa.eu/mk/un/UNECE_CLRTAP_MK/.

Figure 3. Energy production and traffic are important sources of emissions to air (Photo: Pia Anttila).

4.3. Dispersion modelling

Dispersion models can be used to estimate the impact of specific emission sources and source categories to the air quality. They can be used to support the decision making providing information of the impact of air quality improvement and emission reduction measures and for example to support the urban and traffic planning.

Achieving good quality results requires high quality meteorological observation data and detailed emission information. Poor availability of good quality input data has limited the use of dispersion models in the country. However, through initial applications, the foundation for future development has been created.

Dispersion modelling studies have been made to estimate the air quality impact of energy production and industrial installations and traffic in the City of Skopje. In addition, air quality forecasts are made with regional scale model. The models used are the local (UDM-FMI and CAR-FMI) and regional (SILAM) scale models developed by the Finnish Meteorological Institute for assessment of levels of air quality.

5. SOURCES OF AIR POLLUTION

5.1. Road traffic

The emissions from road traffic affect the air pollution the most from the transport sector. The impact of road traffic emissions is the highest in urban areas with dense road networks and high volume of vehicles. The road traffic contributes to the nitrogen oxides, carbon monoxide, benzene and particulate, heavy metal and polycyclic aromatic hydrocarbon emissions.

The emissions from road traffic depend on the type and age of the vehicles, mileage of each vehicle group and quality of the fuels used in the vehicles. Also driving cycle and speed have impact on the emissions; driving in the city area with low speed produces typically more emissions than smooth driving on the highway at a constant speed. In the European Union, the exhaust emission limit values of vehicles have been gradually tightened since 1992 with so called Euro class emission regulations. These continuously updated regulations from Euro 0 (no controls) to the current Euro 6 level have led to a significant reduction of the emissions produced by every new generation of vehicles.

Heavy traffic such as buses and trucks produce the largest amounts of emissions per vehicle. Diesel cars have higher NOx and particulate emissions compared to petrol cars with catalytic converters (Euro 1 onwards). Usually, when the vehicle gets older the emissions are increasing as the catalytic converter and other exhaust treatment systems lose some of the efficiency.

Based on the vehicle register information in year 2015, total amount of registered motor vehicles in the country was 436 502 (Figure 4).

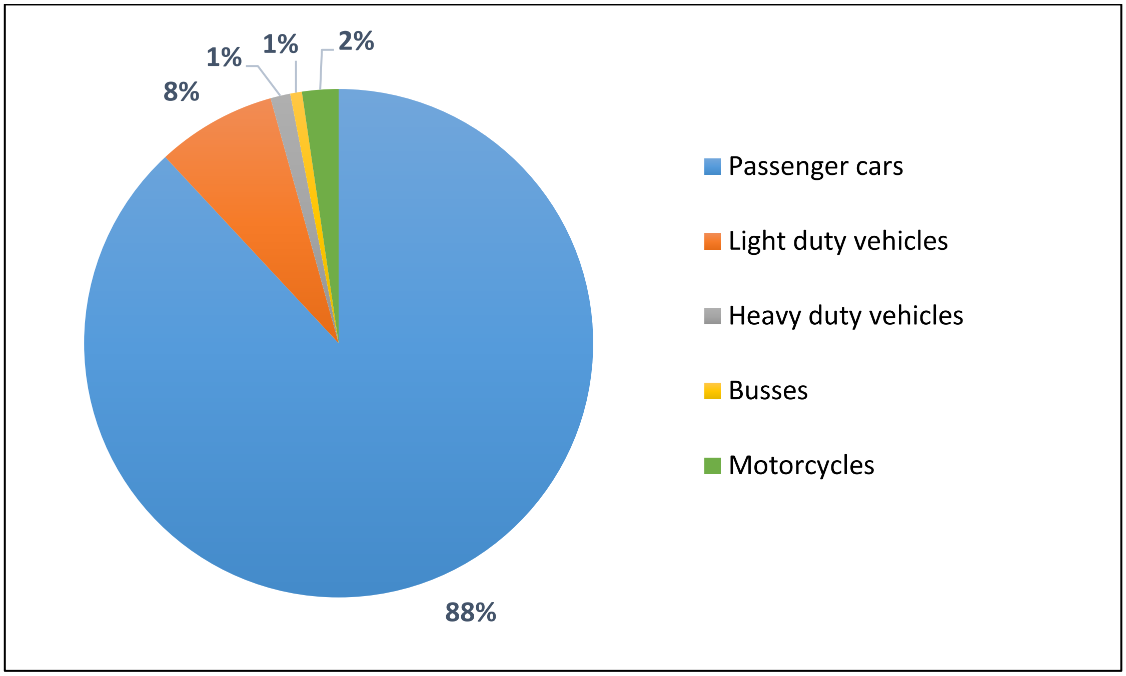

Figure 4. The share of different types of vehicles in 2015 (Ministry of Interior, 2015).

Approximately half of the passenger cars and buses still belong to the high-emission Euro 0 – Euro 2 classes whereas the share is less for light and heavy duty vehicles (Figure 5). According to the vehicle register information there is still a quite considerable share (approximately 10-18 %) of the passenger cars, light duty vehicles and buses that belong to the oldest category of the cars (Euro 0) with no treatment systems for the exhaust gases.

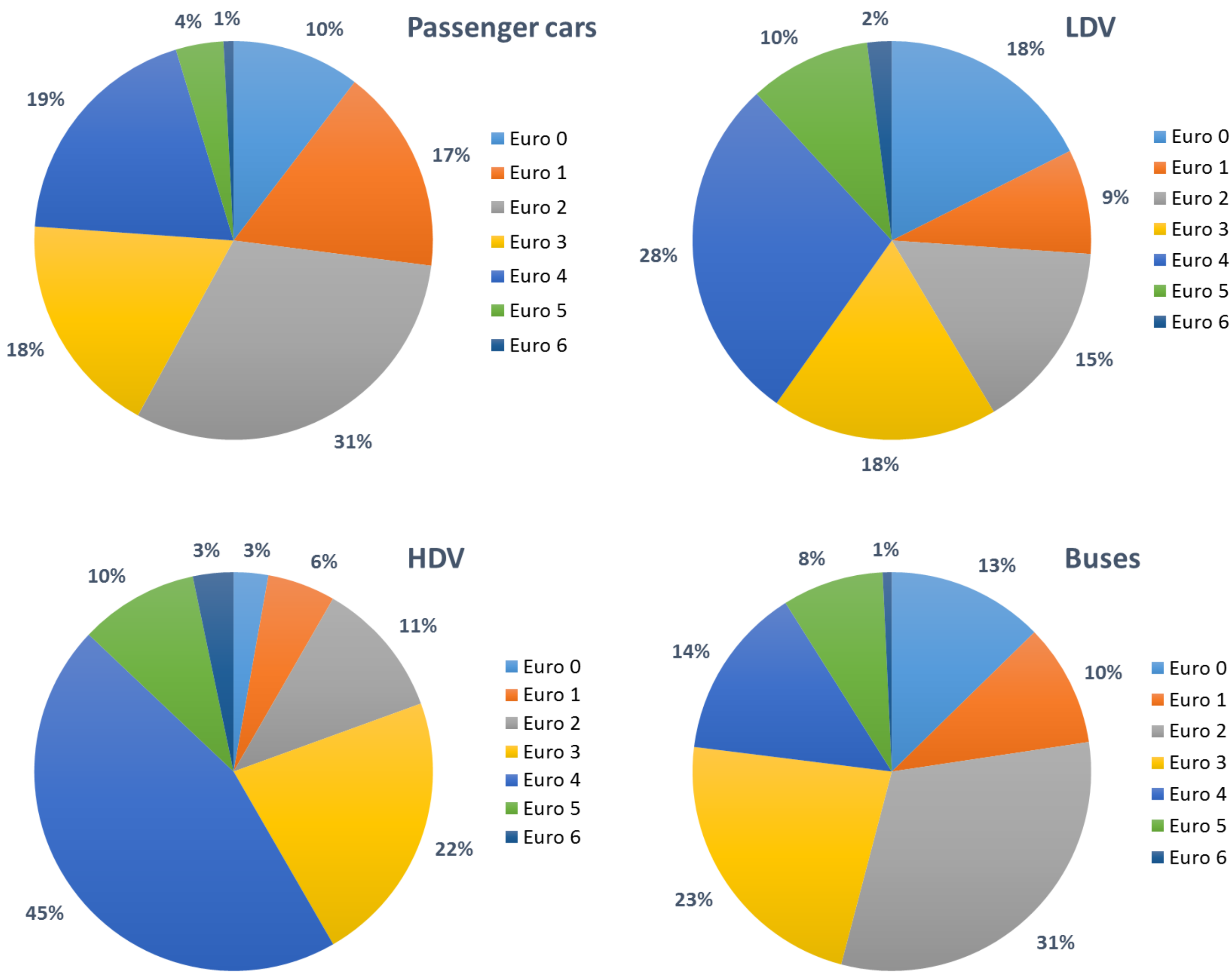

Figure 5. Share of the registered passenger cars, heavy duty (HDV) and light duty (LDV) vehicles and buses belonging to the various Euro emission classes in 2015 (Ministry of Interior, 2015).

5.2. Industry and energy production

Industry is an important source for emissions of particulates and heavy metals, while energy production is key source for SO_x, NO_x, particulate and CO emissions at the national level. Operations of industrial installations are regulated by environmental permits, which for the largest installations are issued by MEPP (so far 113 A licenses issued). Municipalities and the City of Skopje issue the licenses for smaller industrial facilities that emit less pollutants in the air.

Major installations which have significant contribution to the national emissions currently are:

- Power plants generating electricity using lignite and heavy fuel oil such as REK Bitola and REK Oslomej;
- Industrial sector producing ferroalloys (installation Jugohrom Alzar DOOEL);

- Other facilities including cement production installation Titan in Skopje, iron and steel production installations Makstil AD Skopje and Arcelor Mittal and Feni Industries for manufacturing of ferro-nickel.

Installations for the production of electricity and thermal energy located in Skopje utilize natural gas as a fuel and thus have a negligible impact to the air quality.

5.3. Residential heating

Residential heating and other small scale combustion can be a major source of air pollutants. In small stoves, fireplaces and heating boilers the combustion conditions may be inadequate (e.g. low temperature and low combustion air supply) and the quality of the fuel used may be poor (e.g. moist wood) or even unacceptable (e.g. waste). In addition, the emissions are released just at few meters height, near the air people breathe. Typical air pollutants resulting from incomplete, inefficient combustion in small combustion installations include CO, PM, NMVOCs and PAHs.

According to the last official census there are 559 187 dwellings in the country. According to the survey conducted in 2015 (State Statistical Office, 2015) of the total number of households, 62% consume fuel wood as the primary source of heat, 29% use electricity, 8% rely on heat energy from district heating systems, and the remaining 1% utilize other types of heating sources.

The minor contribution of district heat and high percentage of wood used for heating are due to wood being the cheapest fuel and limited availability of heat distribution networks. There are no domestic sources for production of natural gas and the supply of natural gas comes from imports.

5.4. Other sources (agriculture, waste, construction)

The agricultural sector is a major source category of ammonia (NH_3) emissions. Ammonia emissions are mostly originating from animal husbandry, manure management and use of inorganic N-fertilizers. Agriculture is also a minor source of NMVOC and particulate emissions. Most of farms in the country are small individual farms. Cattle and dairy farms are a major source of ammonia emissions.

Large scale controlled waste incineration is still rare in the country, as there is only one clinical waste incinerator operating. Instead, more than 99% of the municipal solid waste is landfilled, mostly in landfills which do not fulfil European standards (with exception of Drisla landfill located in Skopje). The waste sector however is not a major source of air pollutants. Unauthorized open burning of waste including agricultural waste, wood, prunings, slash, leaves, plastics and other general waste may occur which can have a local impact on air quality.

Emissions from construction and demolition activities are mainly particulates, but other pollutants may also be emitted, depending on the materials used. This source has minor contribution in national emissions, but can have an impact on local air pollution.

6. PARTICULATE MATTER (PM_{10} AND $PM_{2.5}$)

Particulates are the most critical air pollutant to affect human health as even small concentrations can pose a health risk. Particulate concentrations in the country are high particularly during the winter time, exceeding significantly the limit values set in the legislation. The main source of particulates is the residential heating. Industry and traffic are also important sources of particulates.

Particle pollution, especially fine particles, contain microscopic solid or liquid droplets, which are so small that they can penetrate deep into the lungs and cause serious health problems. Particle size, chemical composition and physical properties define particulates impact to the air quality and human health. Particulates have also an effect on climate change. Partly they are cooling the climate as they have role in the cloud formation processes and partly they contribute to the melting of the glaciers (black carbon on snow) and therefore warming the atmosphere.

The particles are classified according to their (aerodynamic) diameter, as either PM_{10} (particles with the diameter smaller than 10 µm) or $PM_{2.5}$ (diameter smaller than 2.5 µm). Coarser PM_{10} includes the finer $PM_{2.5}$ fraction.

6.1. Sources and emissions of particulates

There are both natural and anthropogenic sources for atmospheric particulate matter. Natural sources include for example sea salt, naturally suspended dust, pollen and volcanic ash. Anthropogenic sources include fuel combustion for power generation, incineration, domestic heating and fuel combustion for vehicles. Particularly in cities, important local sources are road traffic (vehicle exhaust and road dust) and burning of wood or coal for domestic heating. The height of the emission release of these is low, close to the breathing level. Thus, the impact of these sources to the air quality in ground level can be significant.

Figure 6. Traffic is one major source of emissions (photo: Aleksandar Ristovski).

Particulate matter, also known as aerosols, can be categorised as primary or secondary particulate matter. Primary particulates enter the atmosphere directly and secondary particulates are formed in the atmosphere after the oxidation and transformation of primary gaseous emissions (e.g. gaseous sulphur dioxide to particulate sulphate or gaseous hydrocarbons to secondary organic aerosols). During the worst air pollution episodes a mixture of primary and secondary particles and reactive gaseous pollutants are typically monitored.

Major components of atmospheric PM are secondary sulphate, nitrate, ammonium and organic aerosols, and primary sodium chloride, elemental carbon, mineral dust and water vapour.

There are wide range of sizes and chemical compositions of particulates. Figure 7 illustrates the sizes of particulates from different sources (the largest particulates are presented in the left side of the figure). The particulates included in traffic emissions and tobacco smoke for example are very small (under 2.5 µm of diameter), whereas cement dust and pollen are larger (over 10 µm diameter). Energy production and industrial emissions can include variety of different size particulates depending of the production process. The size of the particulates is very significant from the health point of view, as the finest particulates penetrate deeper in the human body causing more severe health impacts.

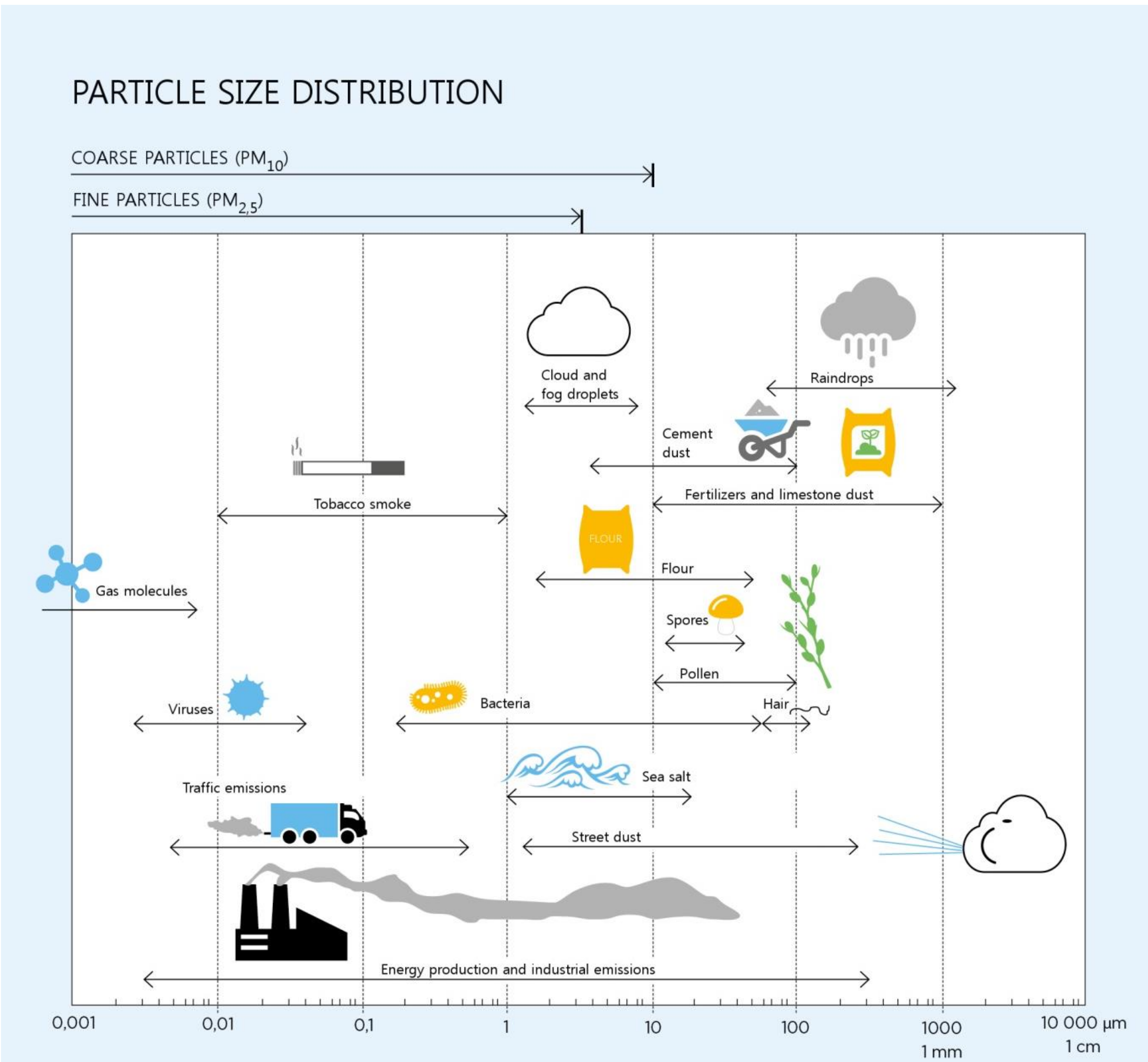

Figure 7. Illustration of the different particulate sizes and examples of their origins.

Figure 8 present the development of the primary (direct anthropogenic) PM_{10} emissions in national level in 1990–2014. In 2014, the major source sectors for particulate matter (PM_{10}) emissions were residential heating, industrial processes and energy industry, representing 36%, 33% and 20% of the total primary emission, respectively. The emissions from residential heating are based on the statistical data of wood, coal and oil consumption for the residential heating. Other emission sectors are less significant. For example traffic emissions were approximately 2 % of total PM_{10} emissions in 2014. Industrial processes produce significant amount of particulate emissions mainly due to the ferroalloys production and energy production utilizing low quality lignite (brown coal). However, these direct primary PM emissions are only a part of the anthropogenic PM load, and secondary aerosols and fugitive dust emissions are not inventoried.

The annual trends of national $PM_{2.5}$ and PM_{10} emissions are similar as the emission sources are mainly the same. Total PM_{10} emissions in year 2014 were 33 000 tons and total $PM_{2.5}$ emissions 22 000 tons. The year-to-year variations of the annual emissions are mainly due to the fluctuations in industrial production (ferroalloys) or mild winters reducing the need for residential heating.

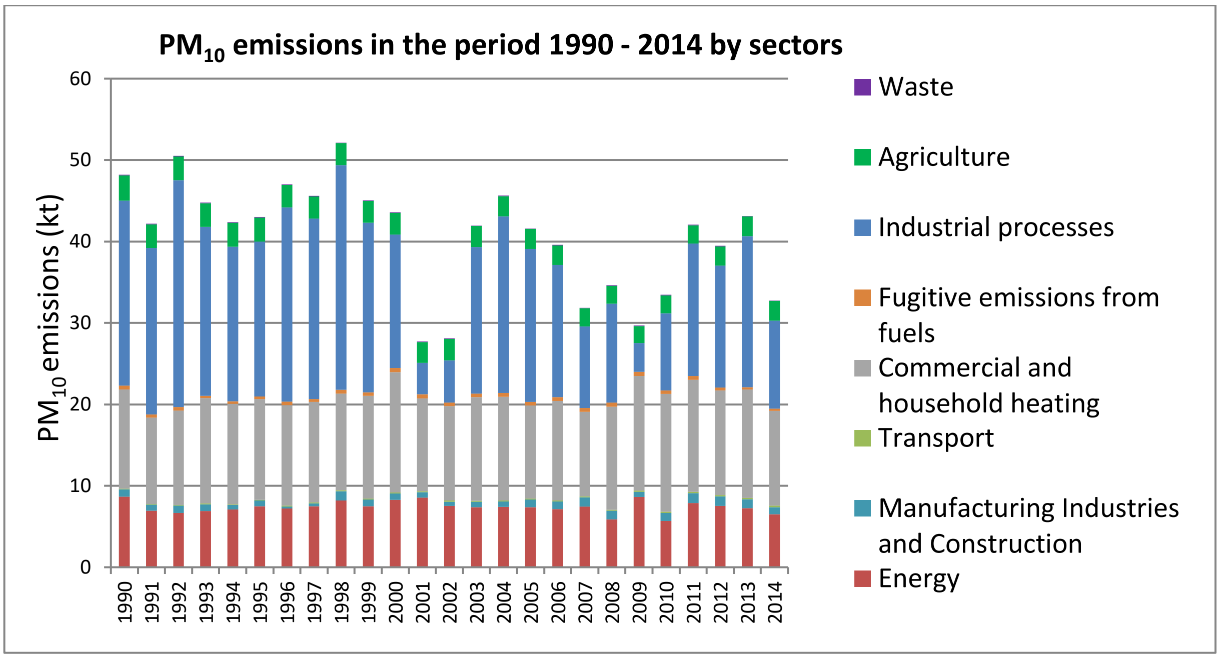

Figure 8. National primary PM_{10} emissions 1990–2014 by the sector (MEPP, 2016).

6.2. Air quality standards for particulates

Limit and target values for PM_{10} and $PM_{2.5}$ for health protection are defined in the national legislation, in which the air quality directive 2008/50/EC (EU, 2008) is transposed with prolongation when the limit value for $PM_{2.5}$ should be met (Table 1). For PM_{10} there are limit values for short-term (daily) and long-term (annual) concentrations. For $PM_{2.5}$ there is only a limit value for long-term (annual) concentration. The daily limit value for PM_{10} (50 µg/m^3) is most often exceeded in the country as well as in other European cities and urban areas.

Table 1. Air quality limit and target values for PM_{10} and $PM_{2.5}$.

Size fraction	Averaging period	Value	Comments
PM_{10} limit value	One day	50 µg/m³	Not to be exceeded more than 35 days per year
PM_{10} limit value	Calendar year	40 µg/m³	
$PM_{2.5}$ target value	Calendar year	25 µg/m³	
$PM_{2.5}$ limit value	Calendar year	25 µg/m³	To be met by 1 January 2020
$PM_{2.5}$ limit value*	Calendar year	20 µg/m³	To be met by 1 January 2025
$PM_{2.5}$ exposure concentration obligation	based on a three-year average	20 µg/m³	2020
$PM_{2.5}$ limit value**			To be met by 2025

***** Indicative limit value (Stage 2) to be reviewed by the Ministry of Environment and Physical Planning in 2018 in the light of further information on health and environmental effects, technical feasibility and experience of the target value in EU Member States.

****** 0–20 % reduction in exposure (depending on the average exposure indicator in the reference year)

6.3. Trend of particulate (PM_{10} and $PM_{2.5}$) concentrations 2005-2015

The particle pollution is on a high level, widespread and persistent in urban locations of the country. The annual mean concentrations of PM_{10} have exceeded the annual limit value (40 µg/m³) at all urban monitoring sites in all years since 2005 (Figure 9). Similarly, daily limit value is exceeded at all sites and all years, except for the monitoring station located in the village of Lazaropole (Annex III).

The highest annual average concentrations of PM_{10} have been measured in Tetovo and Skopje (Lisice) and exceed 120 µg/m³. Statistical trend analysis is not possible due to large amount of missing data. It can be nevertheless assessed that the concentration levels have remained quite stable between 2005 and 2015. An urban mean level of PM_{10} can be estimated to be approximately 80 µg/m³.

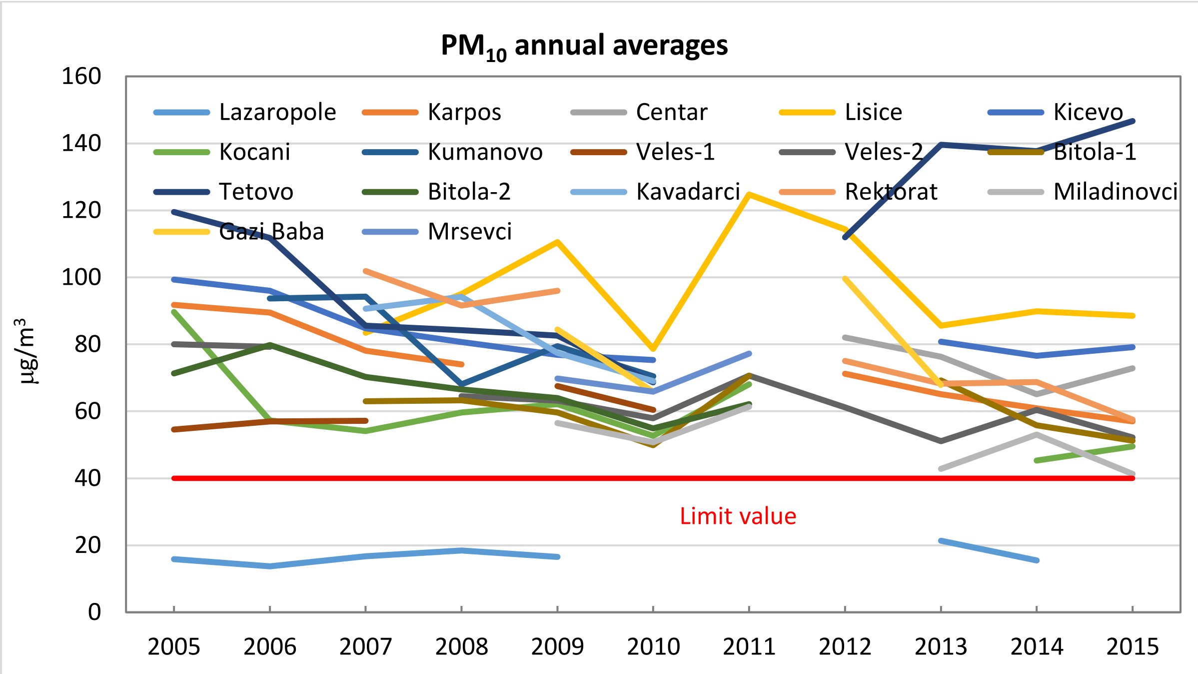

Figure 9. Annual averages of PM_{10} in 2005-2015.

The urban PM_{10} concentrations have a strong and regular seasonal variation; concentrations are at highest in December-January (Figure 10). The high winter time PM_{10} concentrations are related to the higher direct emissions (residential heating with wood in particular) but also meteorological conditions that limit the dispersion of emissions and facilitate chemical reactions that create more secondary particles from e.g. vehicle exhausts. Wintertime smog episodes are typical in cities which are located in valleys.

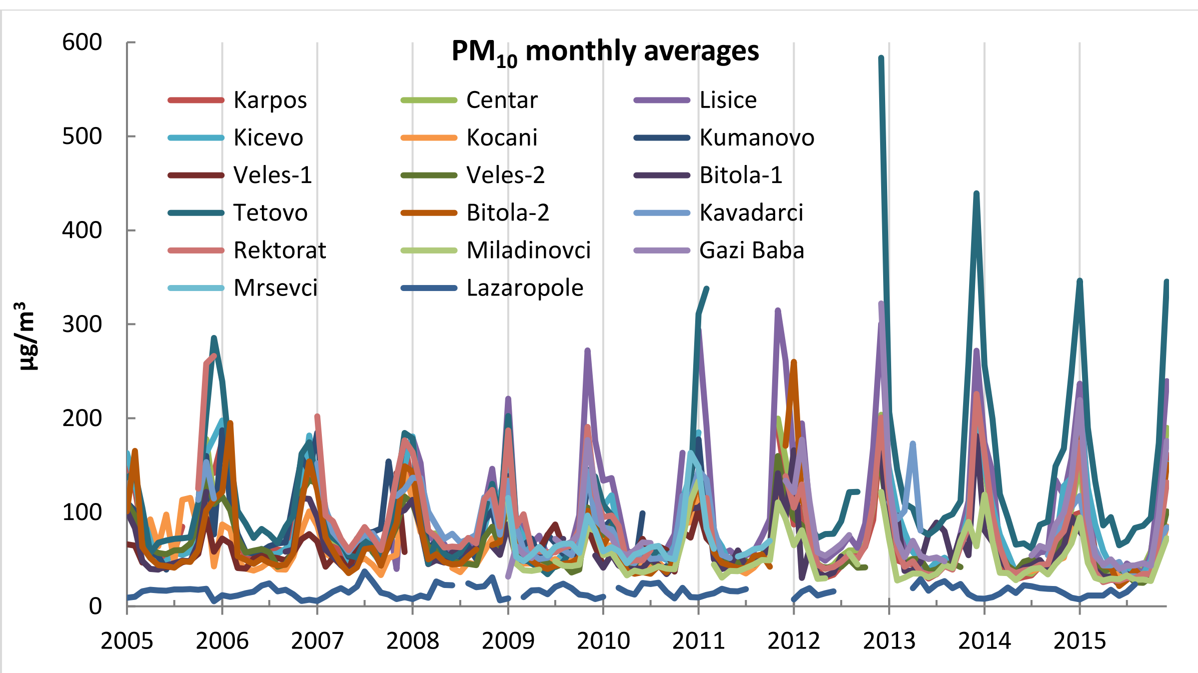

Figure 10. Monthly averages of PM_{10} between 2005 and 2015.

Mountains generally reduce the flows of air in valleys and allow pollutant levels to increase the ground level. Atmosphere tends to be stable (lower wind speeds) during night and neutral to unstable during days (higher wind speeds). This is why also the air pollution concentrations often increase during the night-time even though the emissions are not the highest at the same time.

In cities surrounded by mountain ranges the most severe pollution episodes are created during specific atmospheric conditions, so called temperature inversions. These inversions occur during the winter months when normal atmospheric conditions (cool air above, warm air below) become inverted and the normal vertical mixing of warm and cold air is prevented. Inversions trap a layer of cold air under a layer of warm air. The warm layer acts much like a lid, trapping pollutants in the cold air near the valley floor. Pollutants do not disperse or dilute but remain trapped at the ground level.

Fog further exacerbates the problem, facilitating rapid gas to particle reactions and condensation of semi-volatile species and thus higher particulate pollutant concentrations i.e. creation of smog. The longer the inversion lasts, the more the levels of pollution trapped under it increase. The warm inversion air layer is usually displaced only by a change of the weather pattern, manifested typically with higher wind speeds.

Figure 11. Lisice Skopje on Friday 20 November 2015 at 08:00. During the preceding week the pollutants accumulated in the Skopje valley. PM_{10} and CO hourly maximum levels were measured on Thursday evening; 500 µg/m^3 and 10 mg/m^3, respectively (Photo: Riste Stefanovski).

Also during the summer the PM_{10} concentration levels are relatively high: approximately 40-60 µg/m^3 as daily averages. These elevated PM levels are likely due to direct local emissions, photochemical particle formation from precursor gases, regionally dispersed particulates from forest and land fires and also background aerosols. At the rural background site Lazaropole, the average PM_{10} concentration level have varied between 14–21 µg/m^3, but there the maximum levels occur during the summer months. The average particulate concentration levels during the summer months (June–August, whole study period) was 20 µg/m^3, which can be considered as summer time background particulate concentration in this region.

Fine particle ($PM_{2.5}$) concentrations have been measured at two sites in Skopje since 2012. Annual average concentrations have been approximately 40-50 µg/m^3 (Figure 12) which is twice the level of the limit value.

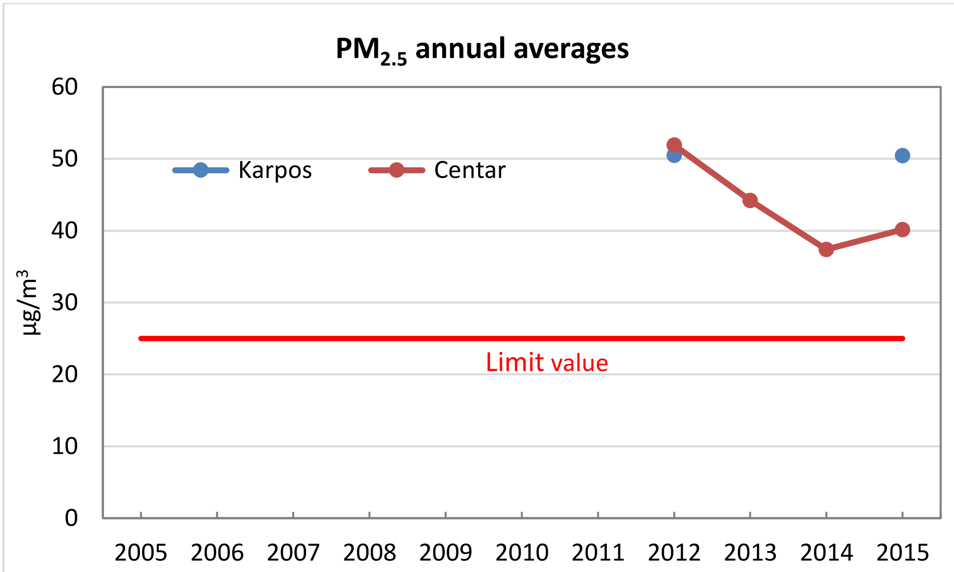

Figure 12. Annual averages of $PM_{2.5}$ in Skopje 2012-2015.

At Centar and Karpos sites in Skopje the fine particle concentrations correlate highly, however, at the Karpos site the concentration level is slightly higher (10%) than at Centar site. Comparison of the PM_{10} and $PM_{2.5}$ concentration shows that large proportion (approximately two thirds) of the respirable particles belong to the most harmful fine particle fraction (Figures 13 and 14).

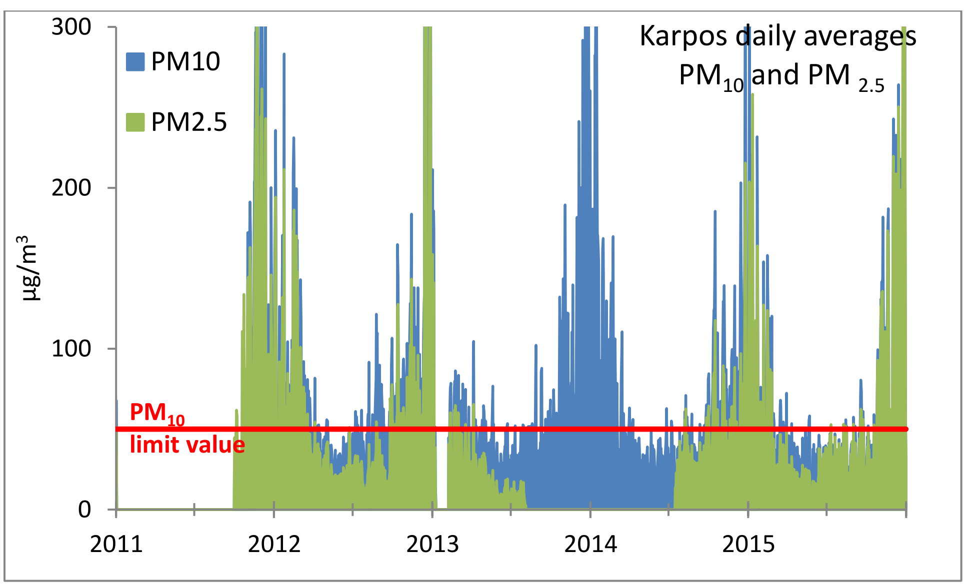

Figure 13. Daily averages of PM_{10} and $PM_{2.5}$ in Karpos.

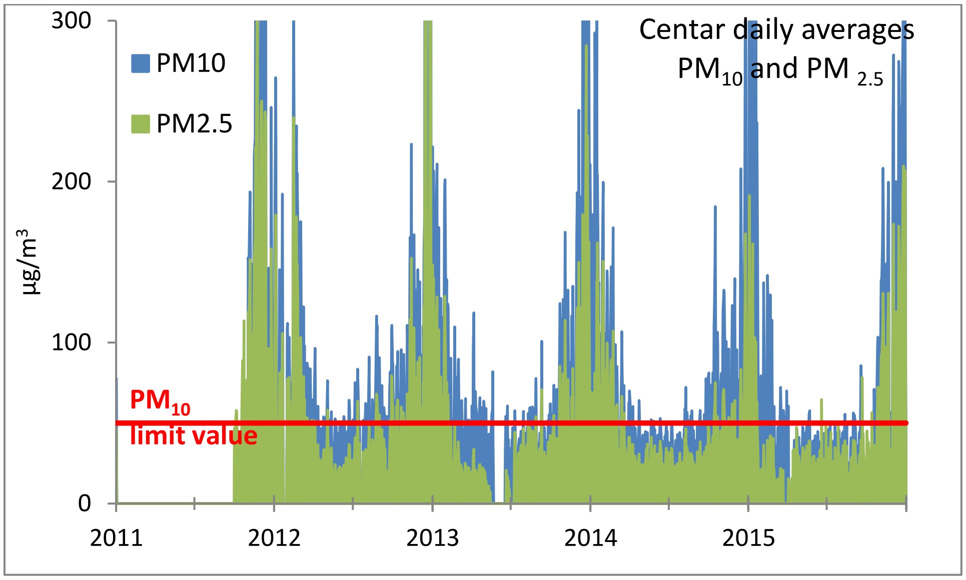

Figure 14. Daily averages of PM_{10} and $PM_{2.5}$ in Centar.

Source apportionment study for particulate matter

The contribution of different emission sources to the concentrations measured in Karpos station in Skopje was estimated with Positive Matrix Factorization (PMF) Model. Particulate matter, SO_2, NO_2, CO, O_3, heavy metal and PAH concentrations measured during the period of August 2015-February 2016 were used in the assessment.

The study shows prominent contribution of biomass burning to $PM_{2.5}$ and PM_{10} concentrations in Karpos. The biomass burning originating from domestic heating contributes to 32-36% of the particulate matter concentrations (graphs below). Other important sources of particulate matter concentrations are traffic with 16-19%, soil including road dust with 19-20% and industry with 18% of the contribution to the particulate matter concentrations.

The location of the monitoring station in Karpos represent a residential area which is not significantly and solely affected by any single emission source. Therefore it can be estimated that the concentrations measured in this location are similar to which majority of people living in Skopje are exposed to. However, the concentrations and the source contribution can be different in other areas for example close to major roads or where a large number of houses are using wood as heating source. Nevertheless the result indicate that the air quality improvement measures in local level in Skopje should be directed to domestic heating and traffic sectors.

Due to very limited dataset (heavy metal and PAH concentration data was available only for six months period) the results of the PMF modelling should be considered as indicative. To improve the results of the source apportionment studies, longer time-series of reliable air quality monitoring data should be utilized.

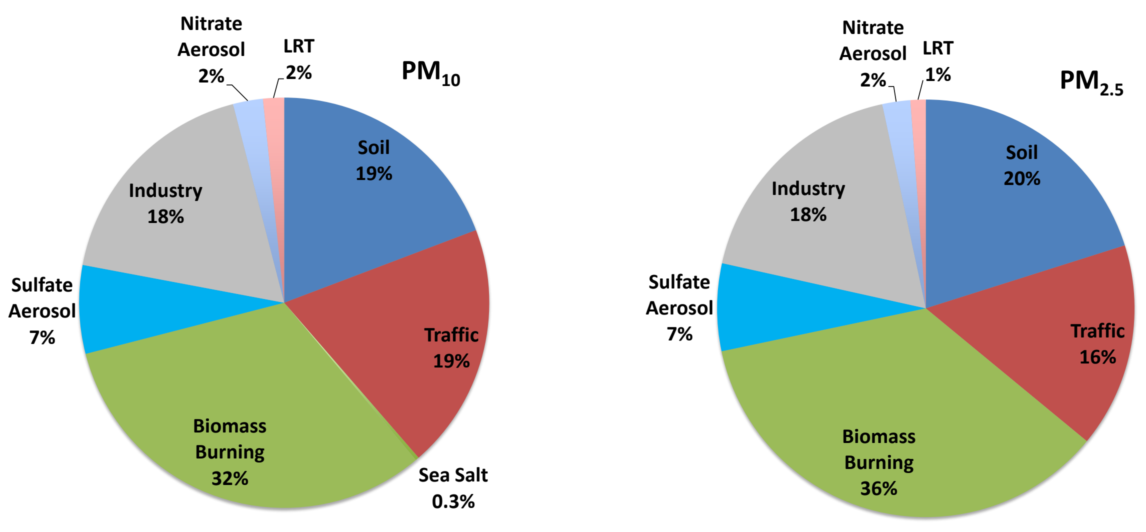

Figure 15. Contribution of different emission source sectors to PM_{10} and $PM_{2.5}$ concentrations in Karpos urban background station.

7. NITROGEN DIOXIDE (NO_2)

Traffic is causing the highest NO_2 concentrations in the city areas. By developing and promoting public transportation and cycling the amount of cars in city centres can be reduced and the air quality improved. The oldest cars have the highest emissions, thus renewal of the car fleet will reduce the traffic emissions.

Nitrogen dioxide is mainly formed from oxidation of nitrogen monoxide (NO). These two gases together are known as nitrogen oxides (NOx). Nitrogen dioxide is the main source of nitrate aerosols, which form particulates ($PM_{2.5}$ and PM_{10}) and in the presence of ultraviolet light, ozone (O_3). NO_2 can have adverse effects on ecosystems, as it can beside of its acidifying effects act as a nutrient. However, excess deposition of reactive nitrogen in ecosystems may cause eutrophication (nutrient oversupply).

7.1. Sources and emissions of NO_2

The major sources of anthropogenic emissions of NO_2 are high temperature combustion processes (heating, power generation and fuel combustion in vehicles' engines). Emission occurs mainly as NO, which rapidly transforms to NO_2 in the atmosphere.

Figure 16. The use of public transport is one way to reduce the traffic congestion and emissions (Photo: Aleksandar Ristovski)

Major share of the national NO_2 emissions is caused by the energy sector (41 % in 2014) and traffic (40 % in 2014) (Figure 17). Total amount of NOx emissions in 2014 was approximately 32 000 tons (MEPP, 2016).

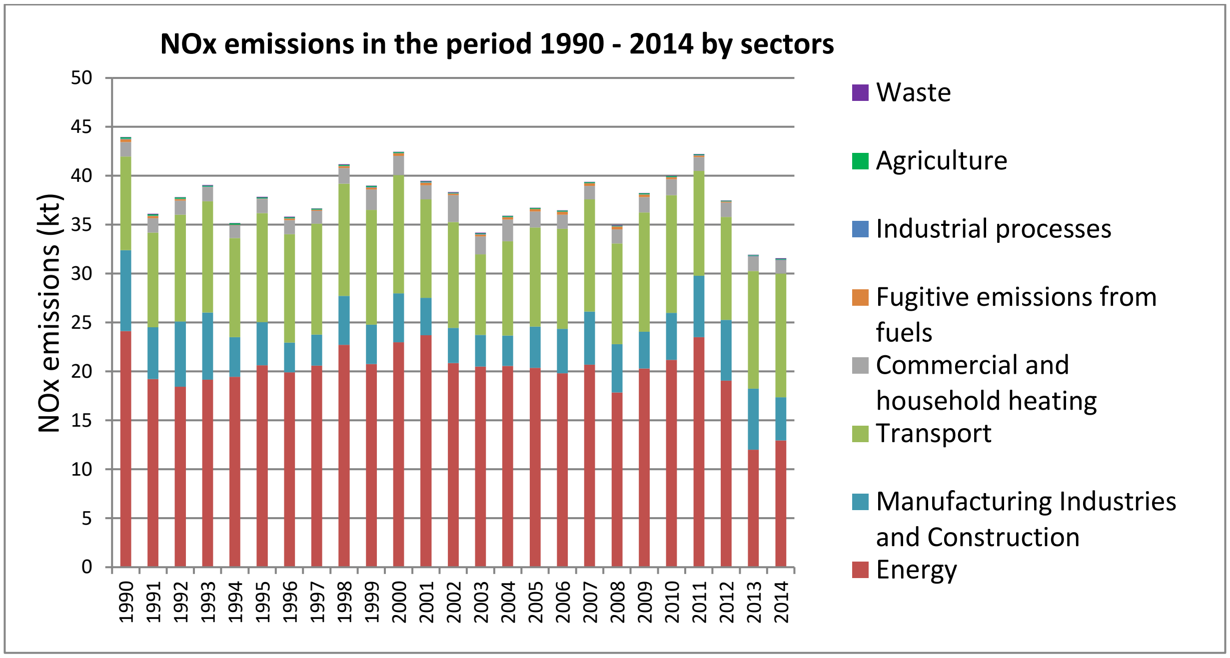

Figure 17. National NO_x emissions 1990-2014 by sectors (MEPP, 2016).

A decreasing trend is noticed for NOx emissions from 2011 (Figure 17), which is a result of the shorter operation of the power plant REK Oslomej and decrease in coal consumption and gasification of one heating plant. Lower NOx emissions in 2013 compared to 2012 are also a result of the modernization of the boilers in the major power plant REK Bitola as well as gasification of the existing heating plants. From 2013 to 2014 emissions remained quite stable.

According to the national emission inventory information the NOx emissions from transport have remained in the same level in recent years. In the national emission inventory system the transport emissions are calculated based on the energy balance (fuel consumption and assumptions on how the fuel usage is distributed by different vehicle categories), while the mileage of different vehicle types is not considered. This creates uncertainty in emission inventory system, which could be decreased using higher level calculations taking into account mileage and categorisation of vehicles by euro classes.

The ongoing renewal of the car fleet and the resulting adoption of lower-emission vehicles is not yet visible in the national emission estimates. There are many variables affecting the renewal rate such as the economic situation of the country and changes in it, legislation and regulation, incentives for transportation, taxation of new cars and different fuel types. Based on these consumers decide when and what type of vehicle they will buy and use, if any.

Assessment of impact of road traffic to NO_2 concentrations in Skopje

The dispersion of nitrogen dioxide emissions from road traffic was calculated using CAR-FMI dispersion model. The emissions were calculated for 475 road segments covering 113.5 km of roads in the Skopje city area. Data from automatic traffic counters, the vehicle fleet statistics and emission factors for different types of vehicles were used in the calculations. The total annual NOx emissions from the road traffic in the area were calculated to be approximately 500 t/a. In addition one year (2015) meteorological observation data was used in the calculations.

In figure 18 the annual average concentrations from the road traffic in Skopje are shown for 2015. According to the model calculations the NO_2 concentrations exceed the annual limit value (40 µg/m^3) in the vicinity of major roads and crossroads. In residential areas the annual concentrations are below the limit value.

The model calculation results should be compared to measured concentrations to estimate the reliability of the modelling results. For the year 2015 there is not available reliable NO_2 measurement data from the monitoring stations in Skopje due to malfunction of the instruments and poor time coverage. However, as during the previous years the annual average NO_2 concentrations have exceeded the limit value in traffic stations, it can be estimated that the modelling results are reasonably reliable. The modeling calculations should be repeated when reliable NO_2 monitoring data becomes available in Skopje.

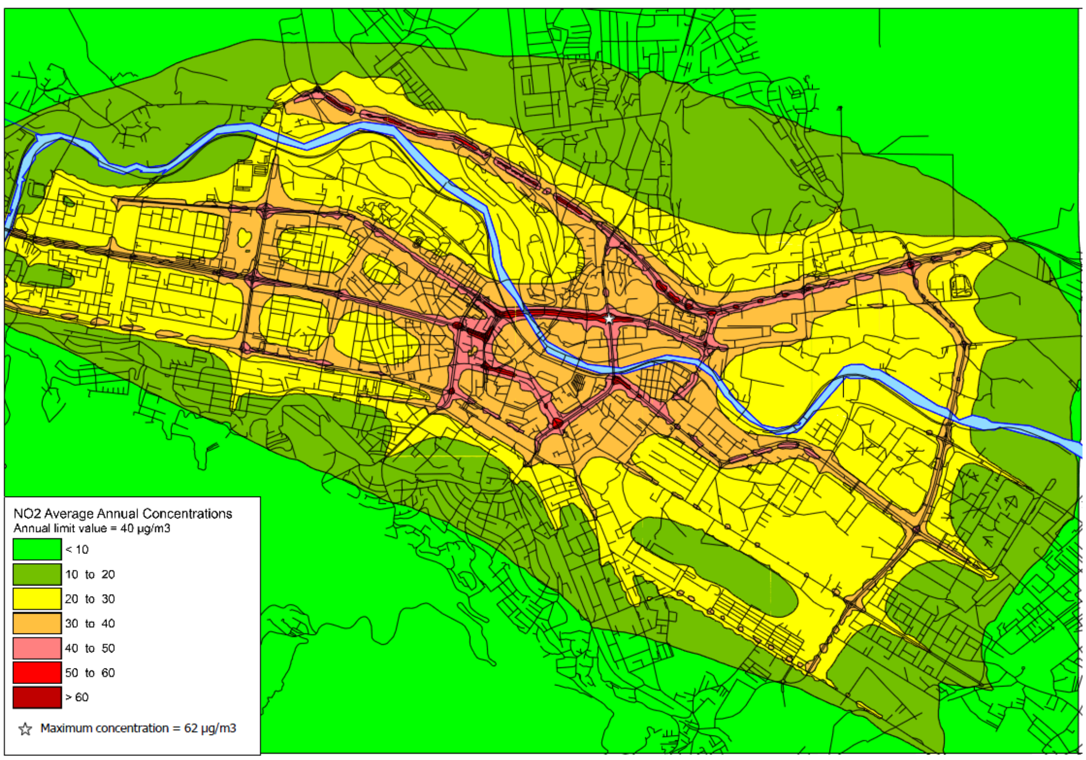

Figure 18. Annual average concentrations of NO_2 from road traffic in Skopje calculated with dispersion model.

7.2. Air quality standards for NO_2 and NO_x

Limit values, critical levels and thresholds for NO_2 and NO_x (Table 2) are defined in the national legislation, which fully transposed the Air Quality Directive 2008/50/EC. Hourly and annual limit values for NO_2 are set for health protection. There is also an alert threshold value for NO_2. When alert threshold is exceeded over three consecutive hours, authorities have to implement action plans. Critical level for NO_x is set to protect the vegetation.

Table 2. Air quality standards for NO_2.

Objective	Averaging period	Limit or threshold value	Number of allowed exceedances
Human health	One hour	200 µg/m^3	18 hours per year
Human health	Calendar year	40 µg/m^3	
Alert*	One hour	400 µg/m^3	
Vegetation**	Calendar year	30 µg/m^3	

*To be measured over three consecutive hours at locations representative of air quality over at least 100 km^2 or an entire zone or agglomeration, whichever is smaller.

**As oxides of nitrogen (NOx), expressed as µg/m^3, critical level for protection of ecosystems.

7.3. Trend of NO_2 concentrations 2005-2015

Monitoring of NO_2 concentrations has been seriously troubled by the insufficient maintenance and aging of the instruments, and as a result, the time series are often discontinuous (Figure 19). In the early years the annual limit values of NO_2 was exceeded at all monitoring sites in Skopje and at the one site in Kicevo. In the recent years the limit value has no longer been exceeded. However, as the results from monitoring of NO_2 concentrations include significant uncertainties, it cannot be confirmed that the NO_2 limit values would no longer be exceeded . In Skopje, the traffic amounts have not decreased or very significant renewal of the vehicle fleet has not occurred, and therefore it is likely that the limit value may still exceed. The NO_2 instruments should be regularly maintained in order to receive reliable information of the concentration levels.

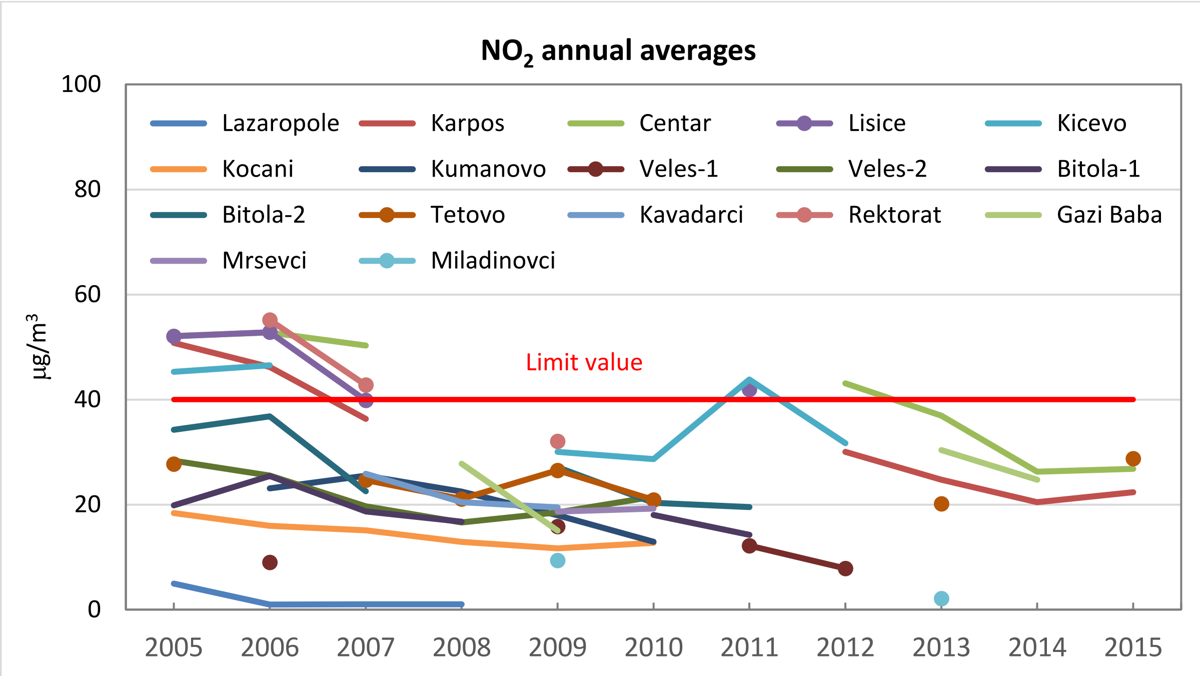

Figure 19. Annual averages of NO_2.

According to the air quality measurements, there is strong and regular seasonal variation of the NO_2 concentrations which can be related to the meteorological conditions (poor winter time mixing conditions) as the emissions (mostly traffic) do not have a similar seasonal variation.

The concentrations of nitrogen dioxides can be estimated to have remained at the same level in recent years. However, the assessment of the trends of NO_2 concentration is challenging due to the considerable uncertainties and low coverage of the monitoring data. The highest NO_2 concentrations are measured in centre of Skopje city close by the busy roads. The annual limit value of NO_2 has been exceeded on the traffic stations in city centre and the exceedances of the annual limit value of NO_2 are still probable. Therefore, the traffic has the most significant impact on NO_2 concentrations especially in the cities and close by the busy roads and intersection areas.

8. SULPHUR DIOXIDE (SO_2)

SO_2 concentrations in air have clearly decreased in recent years as the consumption of brown coal and heavy fuel oil has been reduced. However, the total national SO_2 emissions are still high. Thus, SO_2 emission reduction technologies especially for the main power plants are needed.

Sulphur dioxide (SO_2) is harmful to human health and the ecosystems. It is a major precursor to particulate matter, which is associated with significant health effects. When SO_2 combines with water, it forms sulfuric acid. It is the main component of acid rain, which is a cause of deforestation and acidification of soils and waters.

Sulphur can be stored in soils in certain biochemical conditions and cause postponed acidification process. Thus, SO_2 emission reduction measures may take many decades before they have a positive effect.

8.1. Sources and emissions of SO_2

SO_2 is produced from burning of fossil fuels and smelting of mineral ores that contain sulphur. Generally, the main anthropogenic source of SO_2 is the burning of sulphur-containing fossil fuels for domestic heating, power generation and motor vehicles.

Major share (over 90 % in 2014) of the national SO_2 emissions is caused by the energy sector including public electricity and heat production (Figure 20). Total amount of annual SO_2 emissions in 2014 was 83 000 tons.

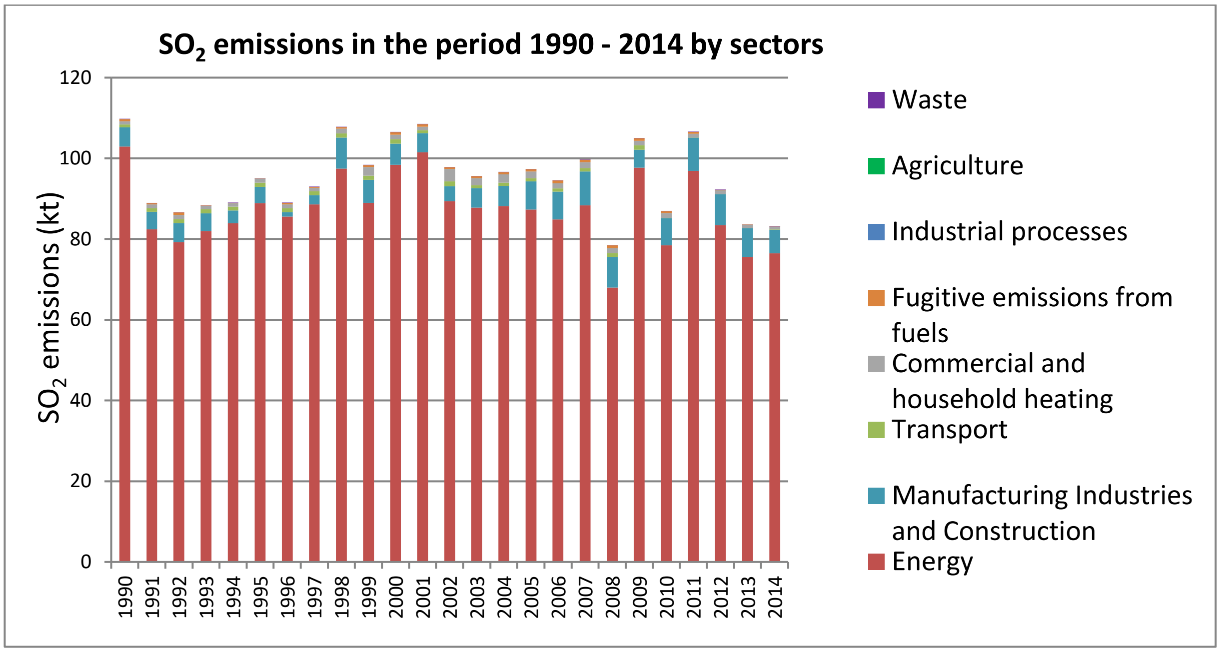

Figure 20. National SO_2 emissions by sectors 1990-2014 (MEPP, 2016)

Two largest coal powered energy production plants REK Bitola (675 MW) and REK Oslomej (125 MW) produce most of the SO_2 emissions due to the lack of desulphurization devices and the use of low quality lignite (brown coal) that consists approximately 2 % of sulphur. The SO_2 emissions of these plants are released through high stacks (250 and 185 meters above the ground level). Because of the high stacks, the emissions disperse and dilute in the atmosphere and as a consequence the ground level concentrations of SO_2 around the plants remain relatively low despite of the high emissions. There are plans to install the desulphurization unit to REK Bitola power plant which would reduce the national SO_2 emissions considerably.

Figure 21. Coal powered thermal heating power plant REK Bitola (Photo: MEPP, 2016).

In addition to the large energy production facilities there are smaller district heating plants using gas and light fuel oil particularly in Skopje area. These smaller energy production units have replaced the use of heavy oil with natural gas or light oil. This has improved the SO_2 concentration levels in Skopje area considerably. Part of the SO_2 emissions (under 10 %) are produced by the industry: steel factories, refinery and cement industry. Road traffic does not emit considerable amounts of SO_2 emissions as due to the use of low-sulphur fuels.

Fluctuations of annual SO_2 emissions (Figure 20) in recent years are caused by the changes in the consumption of coal by the major power plants REK Bitola and REK Oslomej. In 2009 and 2011 there was higher consumption of coal and in the period of 2012–2013 there was decrease of coal consumption. In 2013 lower emissions were also result of the modernization of the boilers in the power plant REK Bitola.

Dispersion of emissions from REK Bitola

The dispersion of sulphur dioxide emissions from the electricity production plant REK Bitola was calculated using UDM-FMI dispersion model. Annual emissions of 61 227 tons from two 250 meter stacks were taken into account in the calculations. According to the model calculations the maximum concentrations caused by the plant are below the limit values set for SO_2 concentrations. The maximum concentrations on the study area were 16 % (3.1 µg/m^3) of the critical level for vegetation protection, 25 % (31.6 µg/m^3) of the limit value for daily concentrations and 44 % (152.3 µg/m^3) of the limit value for hourly concentrations. The highest concentrations occur 2-8 km south or eastward direction of the plant. The City of Bitola is located 14 km west of the plant and due to the prevailing wind directions, the highest concentrations caused by the plant emissions are not occurring in the city. According to the model calculations, the SO_2 concentrations caused by the plant emissions in the City of Bitola are less than 0.5 µg/m^3 for annual average, less than 10 µg/m^3 for daily average and less than 30 µg/m^3 for hourly average concentrations. The modelled daily and hourly concentrations comparable to the limit value are shown in the maps below.

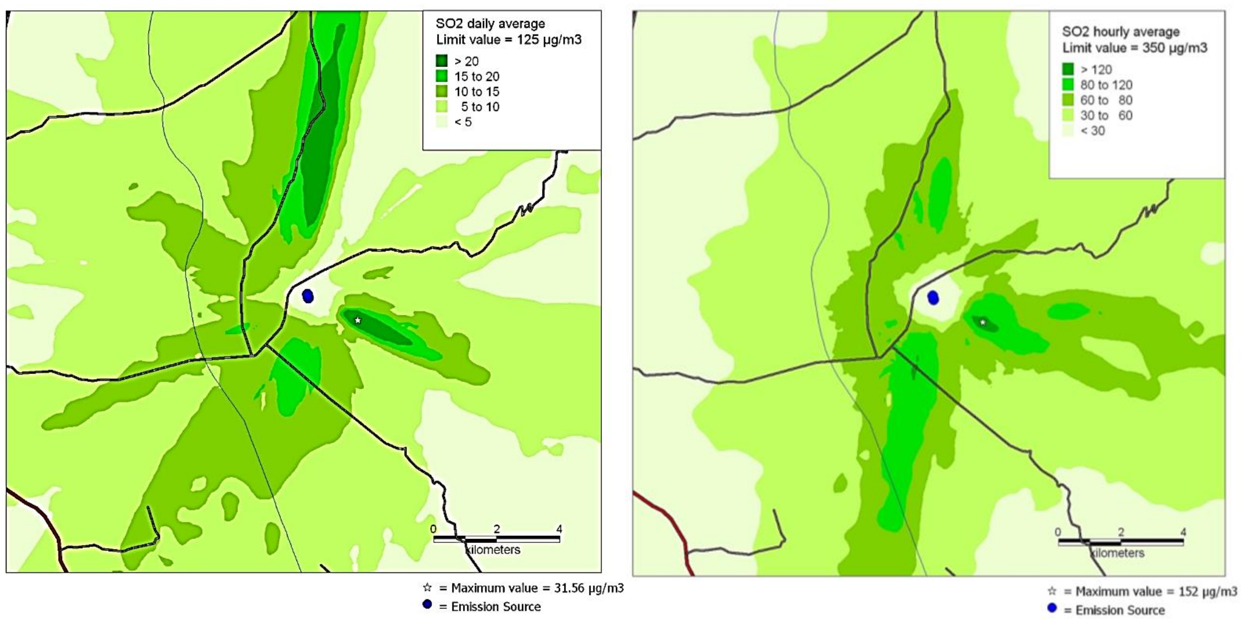

Figure 22. Daily and hourly average concentrations of SO_2 relevant to limit values calculated with dispersion models.

8.2. Air quality standards for SO_2

Limit values, critical levels and alert threshold for SO_2 (Table 3) are defined in the national legislation which was prepared according air quality directive 2008/50/EC (EU, 2008). Hourly and daily limit values are given for health protection. There is also an alert threshold value for SO_2. When alert threshold is exceeded for over three consecutive hours, authorities have to implement action plans to remedy high levels of SO_2 concentrations. Critical levels are set to protect the vegetation.

Table 3. Air quality standards for SO_2.

Objective	Averaging period	Limit or threshold value	Number of allowed exceedances
Human health	One hour	350 µg/m³	24 hours per year
Human health	One day	125 µg/m³	3 days per year
Alert*	One hour	500 µg/m³	
Vegetation**	Calendar year	20 µg/m³	
Vegetation**	Winter (1 October- 31 March)	20 µg/m³	

* To be measured over three consecutive hours at locations representative of air quality over at least 100 km² or an entire zone or agglomeration, whichever is smaller.

**EU critical level for protection of ecosystems.

8.3. Trend of SO_2 concentrations 2005-2015

The reduction in SO_2 emissions have clearly decreased the SO_2 concentrations in the air as well (Figure 23). In the beginning of the monitoring period, the annual means of over 20 µg/m³ were detected in Skopje and Veles. In 2006 in Skopje even the hourly and daily limit values of SO_2 were exceeded. Over the past ten years the decrease in SO_2 concentrations is relatively systematic at all monitoring sites. Since 2007 no exceedances of SO_2 limit values have occurred.

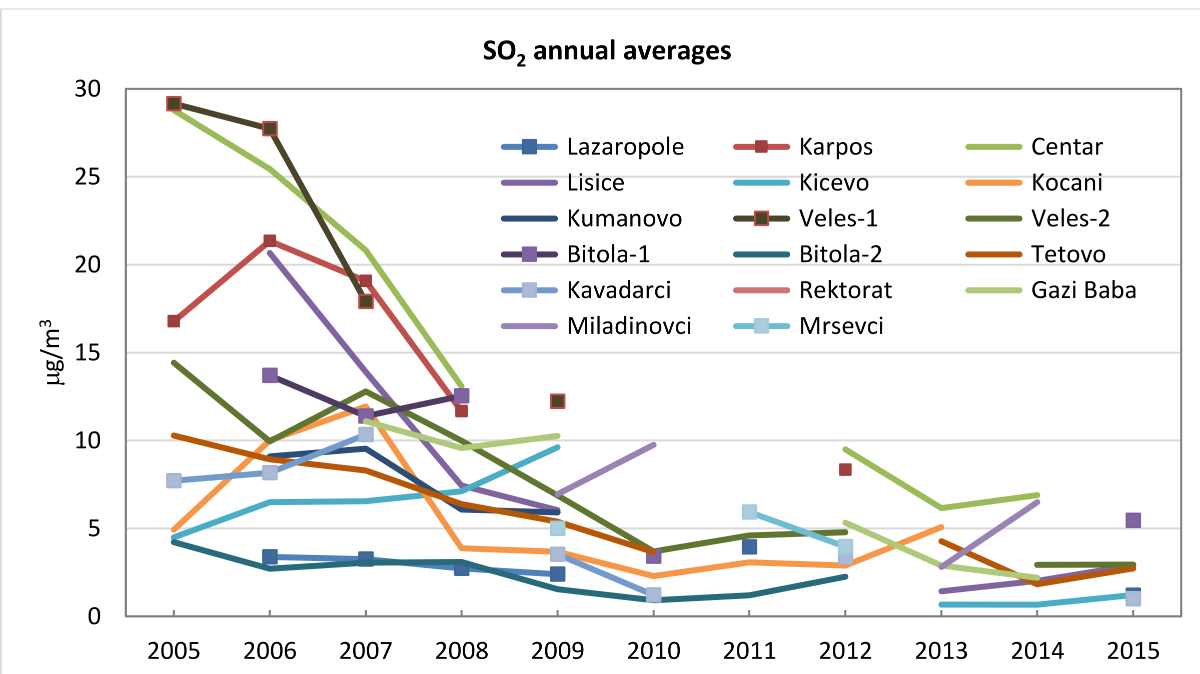

Figure 23. Annual average SO_2 concentrations.

9. OZONE (O_3)

On average the ozone levels in cities are relatively low due to the presence of other pollutants that consume the ozone from the air. However, as typical for these latitudes, short term ozone episodes are usual. Number of these episode days have decreased during the past ten years.

Exposure to ozone is considered to be most damaging to the vegetation compared to any other pollutant in the air. Ozone can have significant effects on the growth of trees, vegetation in general, and important crops such as wheat, soybean and rice. Due to this, high ozone concentrations can cause significant economic losses for forestry and agriculture. Ozone is also harmful to human health.

9.1. Sources of O_3

Ozone (O_3) is formed by complex chemical reactions with the emissions of precursor gases such as nitrogen oxides and hydrocarbons. Generally, in urban environments vehicular exhaust emissions are the most important contributor to the ozone concentrations. In addition to the precursor gases, the formation of O_3 requires sunlight. Therefore the O_3 concentrations are typically higher for example in the Mediterranean countries compared to the Northern European countries.

The concentrations of O_3 usually increase when the altitude increases (within couple of kilometres), thus in high altitude monitoring stations the O_3 concentrations can be higher than in the lower altitude stations. In urban environments the O_3 is consumed with chemical reaction of NO to form NO_2. Therefore, unlike other pollutants, the O_3 concentrations are generally the highest at rural locations, lower at urban sites, and even lower at traffic locations. Sometimes, during episodes of high solar radiation and temperatures, the high O_3 concentrations may occur also in urban environments.

In urbanised areas the reduction of NOx emissions can lead to increase of O_3 concentrations. Nevertheless, O_3 concentrations are not only determined by precursor emissions but also strongly by meteorological conditions. Episodes of elevated O_3 levels occur during periods of warm, sunny weather as the sunlight and high temperatures favour O_3 formation.

9.2. Air quality standards for O_3

Air quality standards for O_3 are defined for the protection of human health and for the protection of vegetation in the national legislation (Table 4). Maximum daily 8-hour mean threshold 120 $\mu g/m^3$ is set for health protection. Legislation includes also information and alert thresholds for health protection. When the information threshold is exceeded, the authorities are obliged to notify the citizens. When the alert threshold is exceeded a short-term action plan needs to be developed.

As the O_3 concentrations have negative impact on vegetation, the legislation also sets targets for the protection of vegetation from high O_3 concentrations accumulated during the growing season (defined as May to July).

Table 4. Air quality target values, objectives and thresholds for O_3.

Objective and legal nature	Averaging period	Target, objective or threshold value
Human health target value	Maximum daily 8-hour mean	120 $\mu g/m^3$ *
Vegetation target value	AOT40 accumulated over May to July	18 000 ($\mu g/m^3$) ·h **
Human health long-term objective	Maximum daily 8-hour mean	120 $\mu g/m^3$
Vegetation long-term objective	AOT40 accumulated over May to July	6 000 ($\mu g/m^3$) ·h
Information threshold	1 hour	180 $\mu g/m^3$
Alert threshold	1 hour	240 $\mu g/m^3$

Note: AOT40, accumulated O_3 exposure over a threshold of 40 ppb. It is the sum of the differences between hourly concentrations > 80 $\mu g/m^3$ (= 40 ppb) and 80 $\mu g/m^3$ accumulated over all hourly values measured between 8 and 20 Central European Time.

* not to be exceeded on more than 25 days per year averaged over 3 years.

** averaged over 5 years

9.3. Trend of O_3 concentrations 2005-2015

Ozone concentrations are being monitored at one remote background site, Lazaropole, and at fourteen urban sites. As expected, the highest concentrations are detected in rural station in Lazaropole, and lowest in the congested city of Skopje (Figure 24). At Lazaropole the concentration level is very high, even for a high altitude site, on average 120 $\mu g/m^3$. Lazaropole is located at 1 350 metres above the sea level. The mean ozone concentration is known to increase with altitude so this concentration level is representative only for the background ozone concentration at this this altitude and latitude. In other locations of the country, the ozone concentrations are significantly lower (Figure 25).

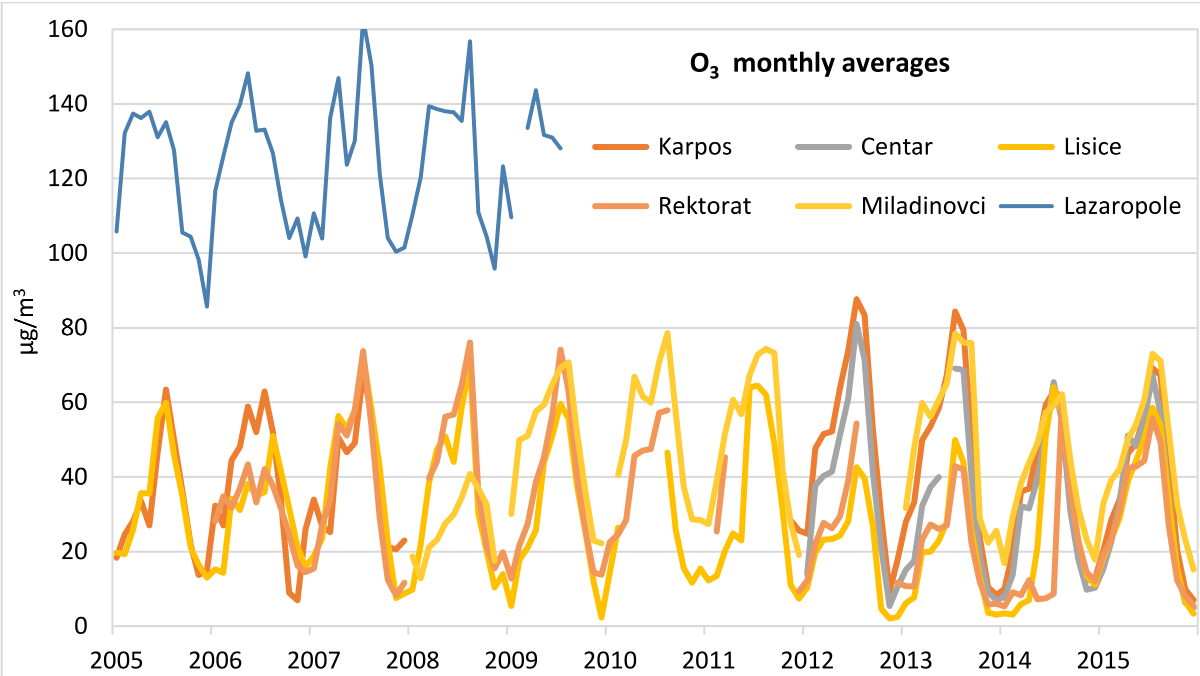

Figure 24. Monthly average concentrations of O_3 in Lazaropole and Skopje.

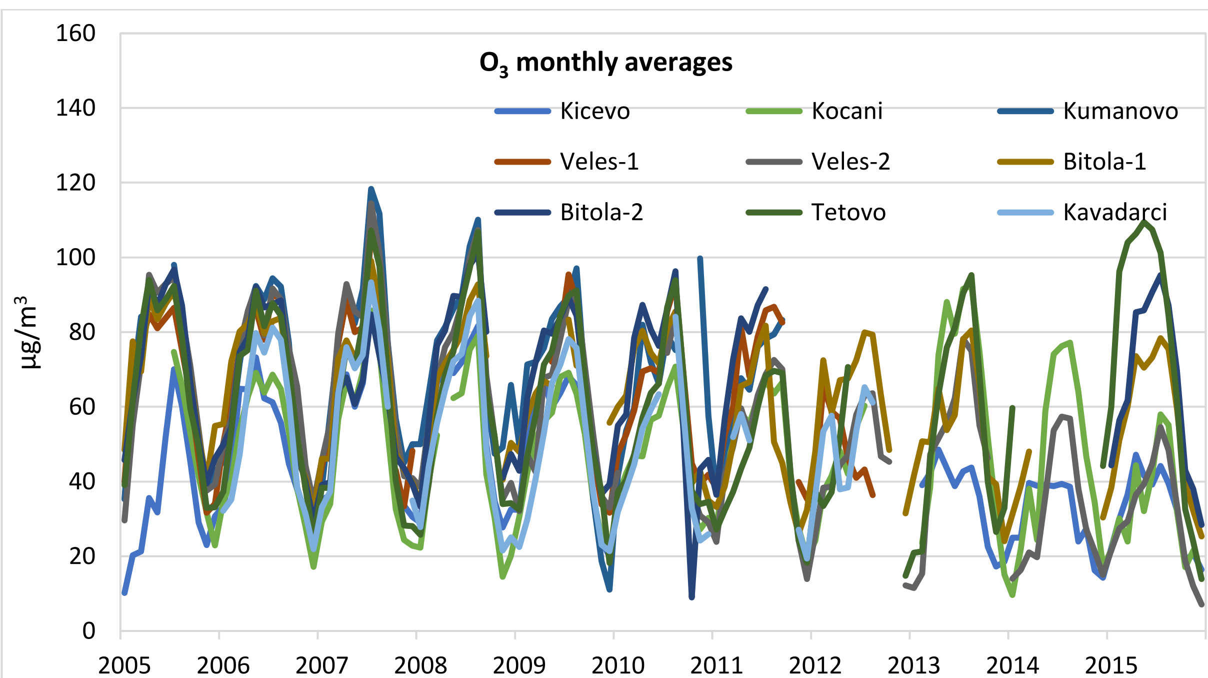

Figure 25. Monthly average concentrations of O_3 in other parts of the country.

In Skopje area the average ozone levels are approximately 30-40 µg/m^3 with no clear trend. In polluted areas (like Skopje) the ozone concentrations should increase with decreasing NO_x concentrations. However, this is not seen from the monitoring data, which suggests that in Skopje no significant NO_2 concentration decline has taken place yet.

In other smaller cities with less traffic, the average ozone levels are approximately 60 µg/m^3, and the concentration levels seem to be declining slightly since 2007 (except in Tetovo). This declining pattern is even clearer in the peak concentrations (daily maximum eight-hour ozone concentrations) (Figure 26). In-depth study would be needed to assess the factors behind this development.

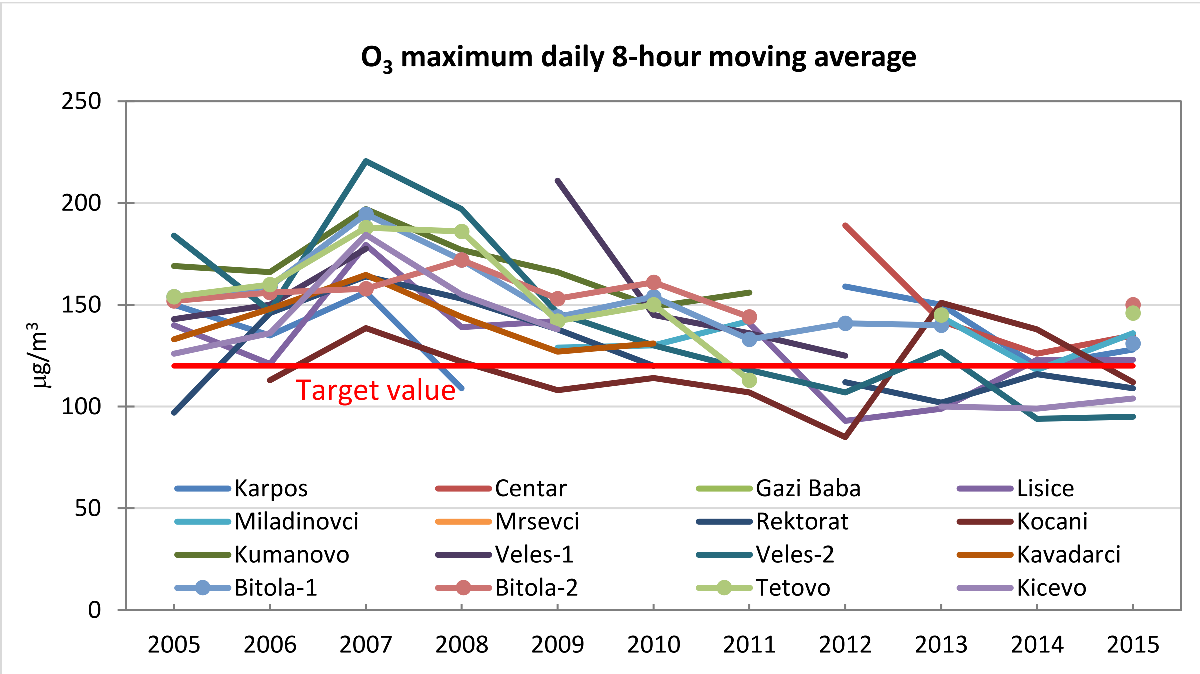

Figure 26. Exceedance of daily target value.

Due to the decrease in peak concentrations also the number of exceedance days of O_3 have declined in the past ten years (Figure 27).

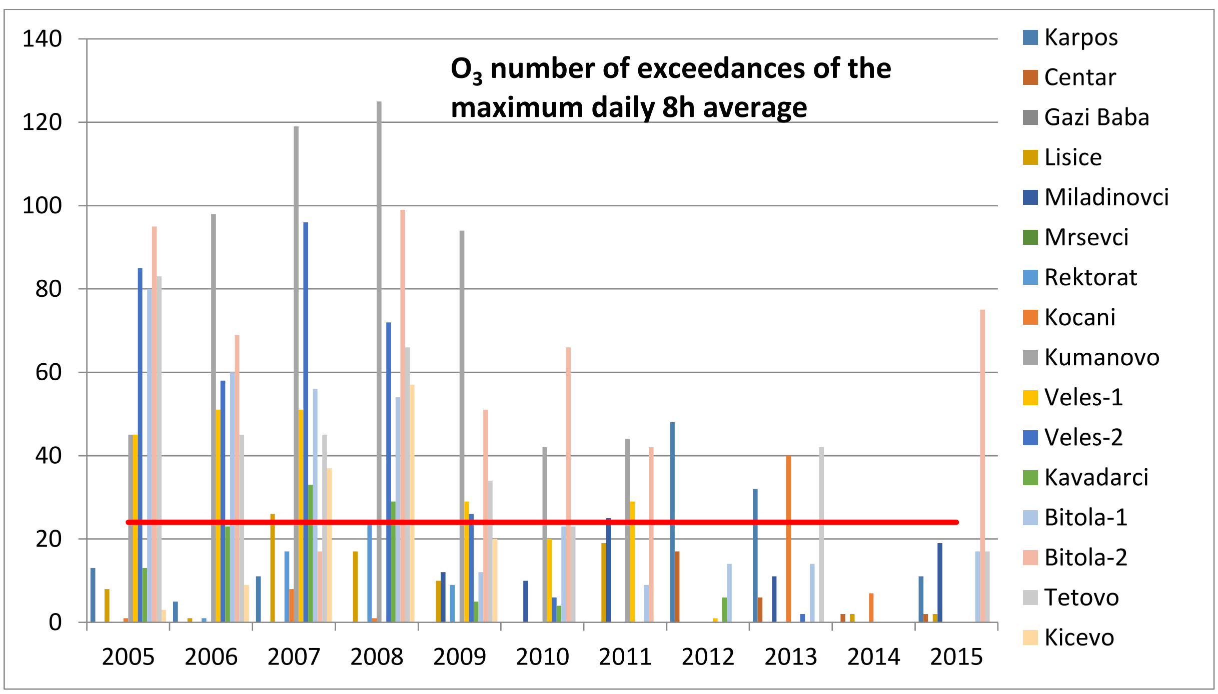

Figure 27. Number of exceedances of the maximum daily 8-hour average.

10. CARBON MONOXIDE (CO)

Unlike in most other European cities, the carbon monoxide limit value is still being exceeded in some Macedonian cities in several days per year. This is likely due to the old car fleet and the widespread use of wood for residential heating.

Carbon monoxide (CO) is a colourless, odourless, and tasteless gas that is slightly less dense than air. In the atmosphere, it is spatially variable and short lived, having a role in the formation of ground-level ozone.

10.1. Sources and emissions of carbon monoxide

Carbon monoxide (CO) is an air pollutant that is formed by combustion processes such as residential heating by solid fuels and road traffic particularly in poor combustion conditions. In European cities the CO concentrations have substantially decreased since the catalytic converters became mandatory for new petrol cars in 1992. The measured concentration levels of CO in the ambient air in Europe are nowadays well below the limit value and the population's exposure to high ambient concentrations of CO is very localised and infrequent.

Figure 28. Domestic burning of wood is an important source of CO emissions (Photo: Aleksandar Ristovski).

The major share of national CO emissions is caused by the residential heating (over 60% in 2014) and transport (27% in 2014) (Figure 29). The total CO emissions have decreased since 1990's approximately by 20%. The decrease is mainly due to the declining emissions from transport sector (road traffic). Total amount of annual CO emissions in 2014 was 112 000 tons.

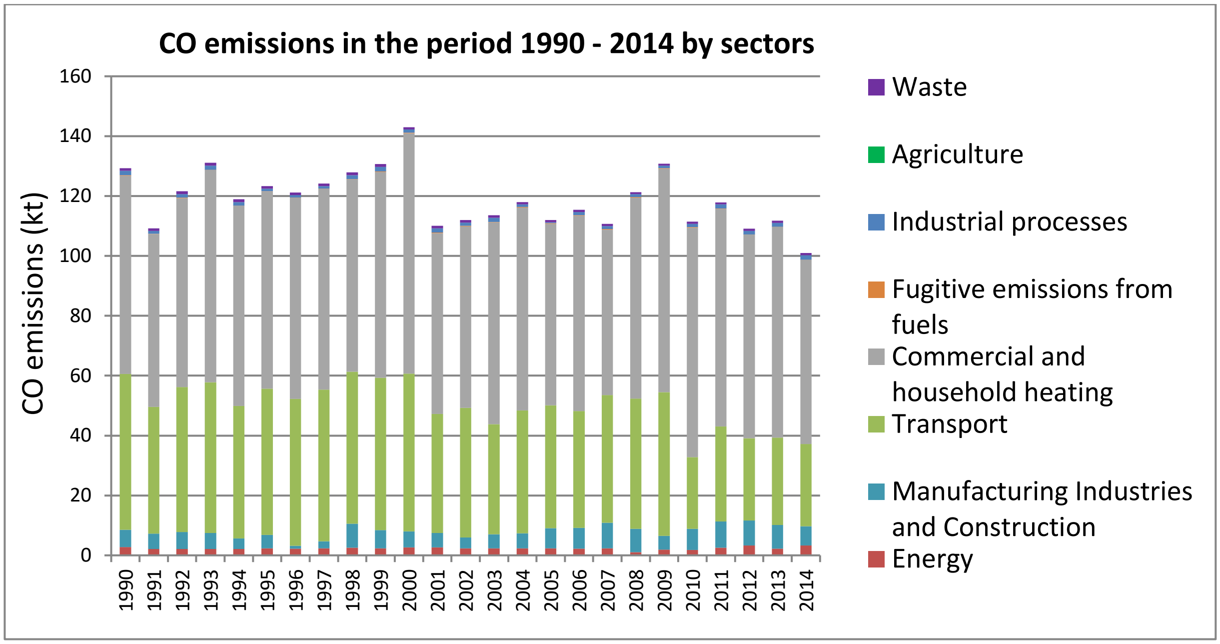

Figure 29. National CO emissions 1990-2014 by sectors (MEPP, 2016)

10.2. Air quality standards for CO

Limit value for CO (Table 5) is defined in the national legislation in which the air quality directive 2008/50/EC (EU, 2008) is transposed.

Table 5. Air quality standards for CO

Objective	Averaging period	Limit value
Human health	Maximum daily 8th hour mean	10 mg/m^3

10.3. Trend of CO concentrations 2005-2015

In Skopje and other urban locations in the country, the carbon monoxide concentrations occasionally exceed the limit value (Figure 30). Obvious causes for this are the old car fleet, and the widespread use of wood for domestic heating. Carbon monoxide concentrations do not show a clear sign of declining trend. In recent years the CO limit value has been exceeded in Skopje, Tetovo and Bitola, as bigger cities in the country with higher traffic density.

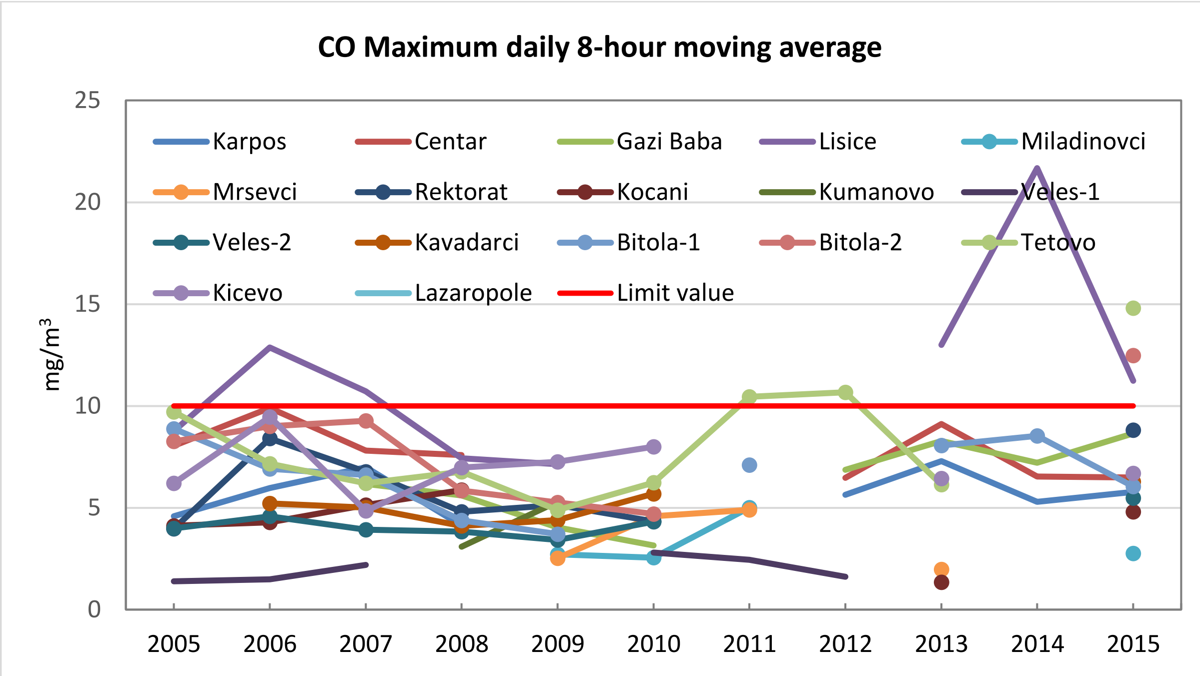

Figure 30. Exceedance of CO daily limit value.

11. POLYCYCLIC AROMATIC HYDROCARBONS (PAH)

The concentrations of polycyclic aromatic hydrocarbons including benzo(a)pyrene are likely to be high in Skopje and other locations in the country especially during the winter months when wood is widely used in residential heating.

Among the air pollutants, the PAHs are one of the most dangerous for human health, since many of them are carcinogenic. PAHs in the ambient air are attached to particles ($PM_{2.5}$ and PM_{10}). The best known and most studied PAH is the benzo(a)pyrene (B(a)P) and it is used to represent the PAHs. B(a)P is the only PAHs which has target value in the air quality legislation. Measured B(a)P concentrations are high across large parts of Europe, mostly as a result of emissions from the domestic combustion of coal and wood.

11.1. Sources and emissions of benzo(a)pyrene

Incomplete combustion of fuels generates the polycyclic aromatic hydrocarbons which are released to the air. Combustion of organic material always releases some PAHs compounds to air. However, typically in the urban areas the major sources of PAHs are the residential heating and exhaust gases from traffic. Some industrial processes, such as foundries and coking plants can also emit PAH emissions.

Figure 31. Wood burning is common in the country (Photo Aleksandar Ristovski)

The largest source of national PAH emissions is the energy sector (mainly residential heating), with approximately 90 % of the total national emissions in 2014 (Figure 32). The total PAH emissions have remained on the same level since 1990. Total amount of PAH emissions in 2014 was 12 t. Annual fluctuations are mainly caused by the yearly differences in meteorology, since colder winters create more need for residential heating. For example the higher emissions in 2013 compared to 2012 are due to the colder winter. A decline in biomass fuel consumption and increase of natural gas fuel combustion in the latest year are also reasons for the lower emissions.

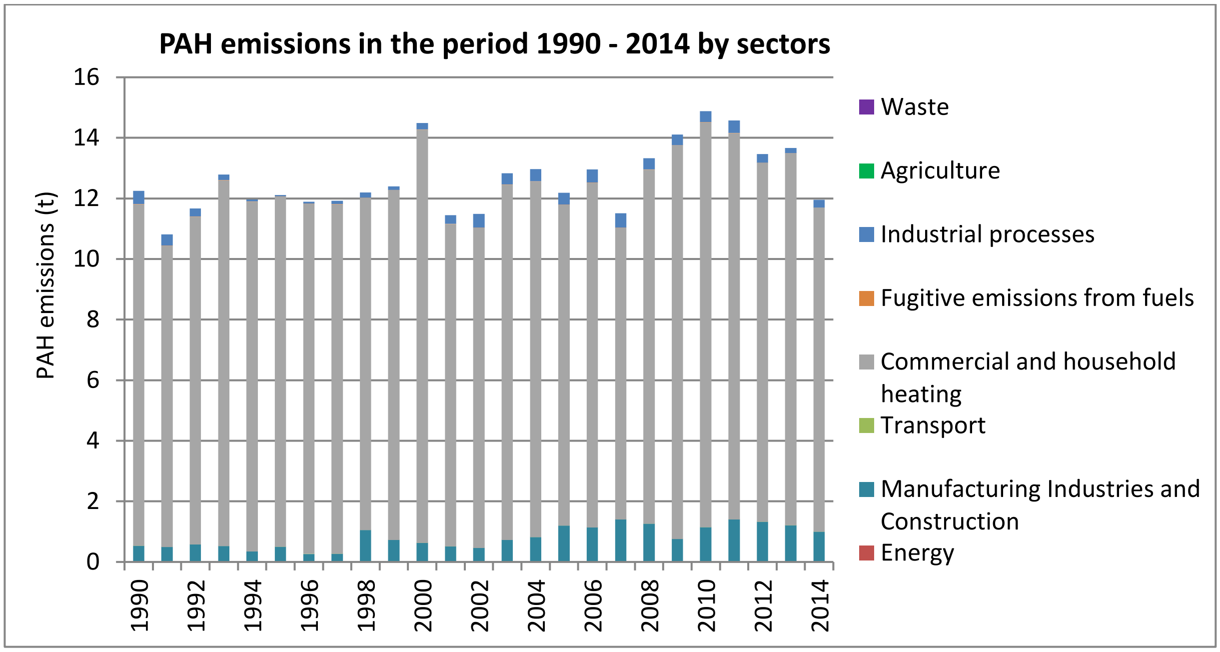

Figure 32. National PAH emissions 1990-2014 by sector (MEPP, 2016).

11.2. Air quality standards for B(a)P

Target value for B(a)P is defined in the national legislation, which has been prepared with the transposition of the air quality directive 2004/107/EC (EU, 2004). Target value is 1 ng/m^3 as annual average.

Table 6. Air quality standard for BaP.

Objective	Averaging period	Target value
Human health	Calendar year	1 ng/m^3 *

*Measured as content in PM_{10}

11.3. Benzo(a)pyrene concentrations

Polycyclic aromatic hydrocarbons in particulate matter (PM_{10}) were measured in a six months campaign in Karpos, Skopje, between August 2015 and March 2016 (Figure 33). Based on the monitoring campaign results, the BaP concentrations are clearly higher during the colder months (from November until February). This is likely due to the higher emissions from residential heating.

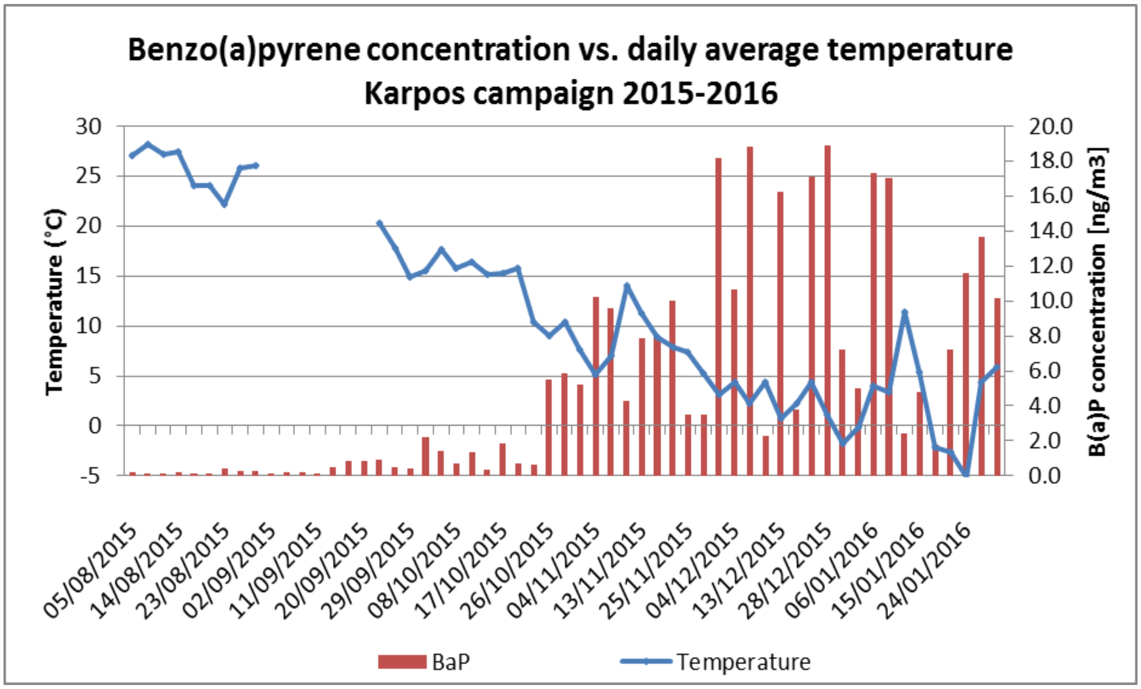

Figure 33. Benzo[a]pyrene concentrations in PM_{10} particles in Karpos during August 2015 – February 2016. Temperature is marked with a blue line.

Average benzo(a)pyrene concentration 8 ng/m^3 measured during a half year campaign exceeded clearly target value (1 ng/m^3). This clearly indicates that PAH concentrations can be elevated in Skopje during the heating season.

12. HEAVY METALS (ARSENIC, CADMIUM, LEAD AND NICKEL)

Since the early 2000s the heavy metal emissions have decreased due to closing of outdated manufacturing industry and introduction of unleaded petrol.

Heavy metals in ambient air are typically attached to particles ($PM_{2.5}$ and PM_{10}). In European level, the human exposure to arsenic, cadmium, lead and nickel ambient air concentrations above the limit or target values is a local problem, restricted to a few areas with specific industrial plants and activities.

12.1. Sources and emissions of heavy metals

Manufacture of basic metals by processing ores containing these substances, originates emissions of heavy metals to air. However, the emission volumes have declined significantly over the past two decades due to technological developments. Also, the use of fossil fuels, as well as the uncontrolled burning of waste can emit heavy metals to air.

The emissions of lead have decreased significantly (over 95 %) starting from 2003 as a result of the closure of the smelter company Zletovo in Veles and the use of unleaded gasoline in transport. The closure of the smelter company reduced also cadmium emissions. Currently the main sources of lead emissions are industrial processes (steel and iron production) and energy sector.

Also the emissions of cadmium and nickel have decreased clearly since the beginning of the 2000s, but for arsenic the emission reduction is not as clear. In 2014 the total national emissions of cadmium were 0.14 tons, arsenic 0.88 tons, lead 4.5 tons and nickel 2.3 tons.

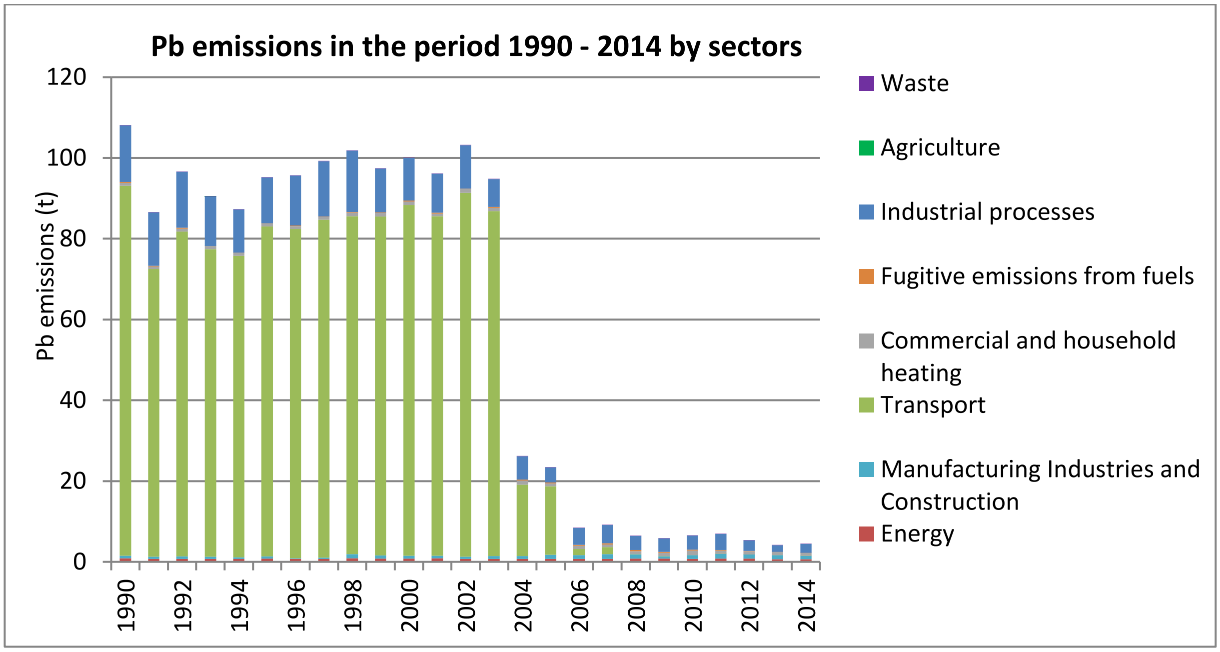

Figure 34. National lead emissions 1990-2014 by emission sector (MEPP, 2016).

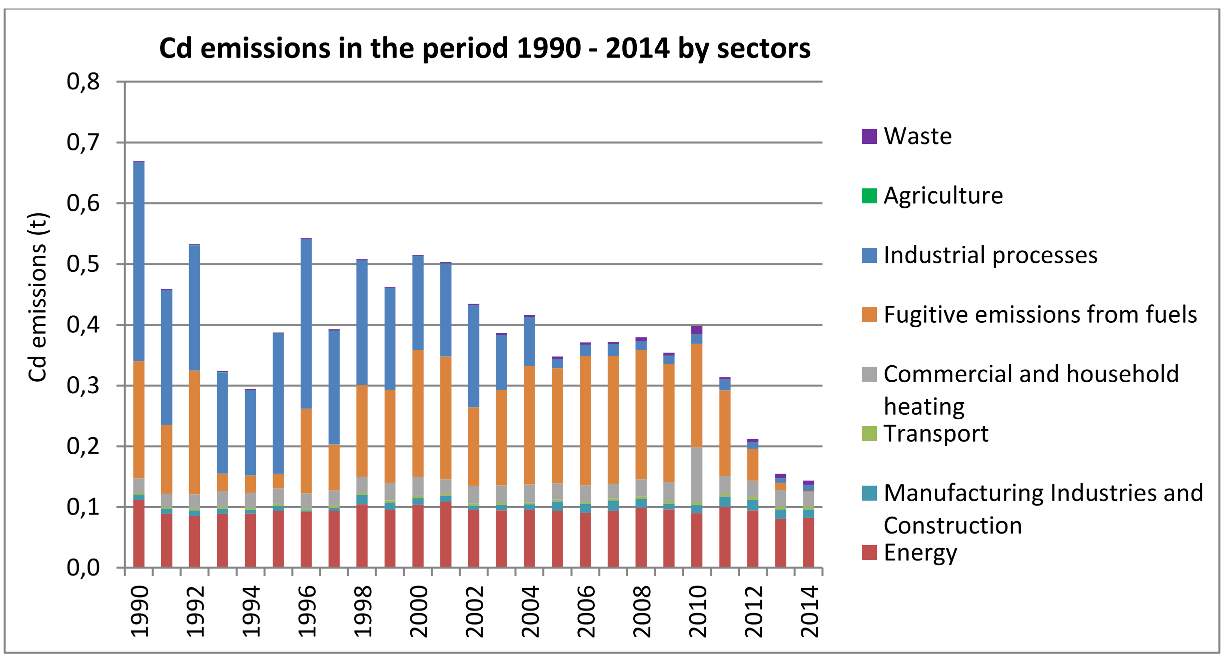

Figure 35. National cadmium emissions 1990-2014 by emission sector (MEPP, 2016).

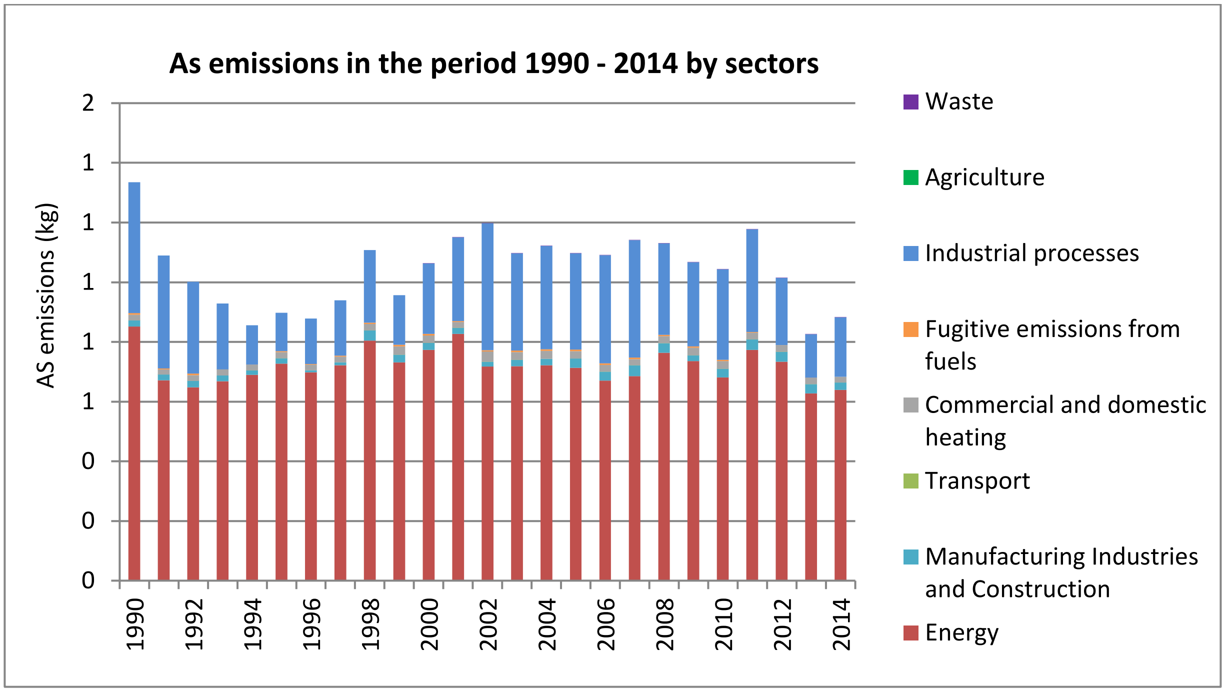

Figure 36. National arsenic emissions 1990-2014 by emission sector (MEPP, 2016).

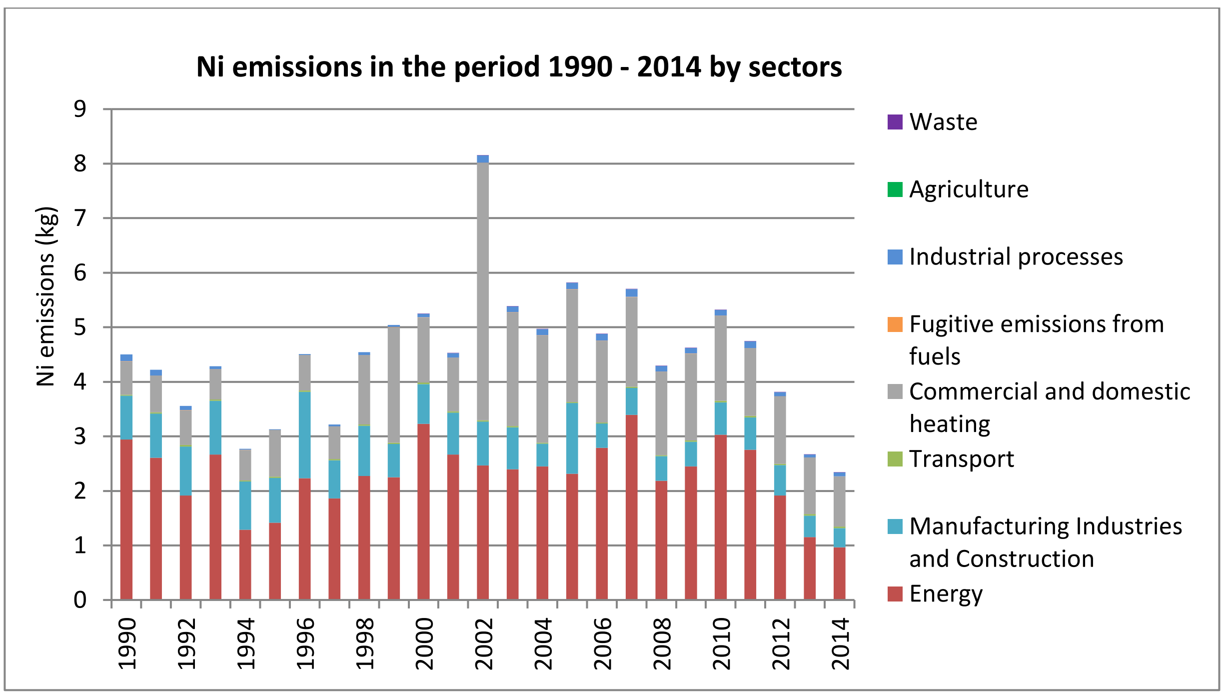

Figure 37. National nickel emissions 1990-2014 by emission sector (MEPP, 2016).

12.2. Air quality standards for heavy metals

National legislation as well as the European Union legislation defines (Directive 2004/107/EC) annual target values for following heavy metals: arsenic (As), cadmium (Cd), Nickel (Ni) and annual limit value (Directive 2008/50/EC) for lead (Pb).

Table 7. Air quality standards for heavy metals (EU, 2004 and 2008)

Pollutant	Objective and legal nature	Averaging time	Target and limit values
Arsenic	Human health target value	Calendar year	6 ng/m^3 *
Cadmium	Human health target value	Calendar year	5 ng/m^3*
Nickel	Human health target value	Calendar year	20 ng/m^3*
Lead	Human health limit value	Calendar year	0.5 μg/m^3*

*For the total content in the PM_{10} fraction averaged over a calendar year.

12.3. Heavy metal concentrations

Heavy metal concentrations are not monitored on a regular basis in the country, but number of measurement campaigns have been organized. The most recent campaigns were organized in Karpos, Skopje during August 2015 – March 2016 and in Tetovo during October 2015 – February 2016. Based on these results the heavy metal concentrations were well below the target values set for the protection of human health in Skopje and Tetovo.

The results of some of the older campaigns have indicated possible exceedance of target values for arsenic, cadmium and nickel. Limit value exceedances for lead the have not been reported. In Skopje, high cadmium concentrations have been measured in campaigns in 2014–2015 in contrast with the results from the latest study. Nickel has been found to exceed the target value only in the vicinity of Feni Industries in Kavadarci, which is the largest ferro-nickel producer in the country. For arsenic, the target value exceedances have been reported in Skopje and in Jegunovce in the vicinity of Jugohrom Alzar DOOEL which is a large ferroalloys production plant near Tetovo. All of the campaigns have been conducted for a period of less than one year, and therefore the results are only indicative when compared to target values which are set for annual average concentrations.

Based on the results of the indicative measurement campaigns and emission inventories of heavy metals it can be assessed that the highest concentrations of heavy metals can be found close to the metal industry. Based on the most recent measurement data, it seems that concentrations of heavy metals remain below the limit and target values in Skopje area.

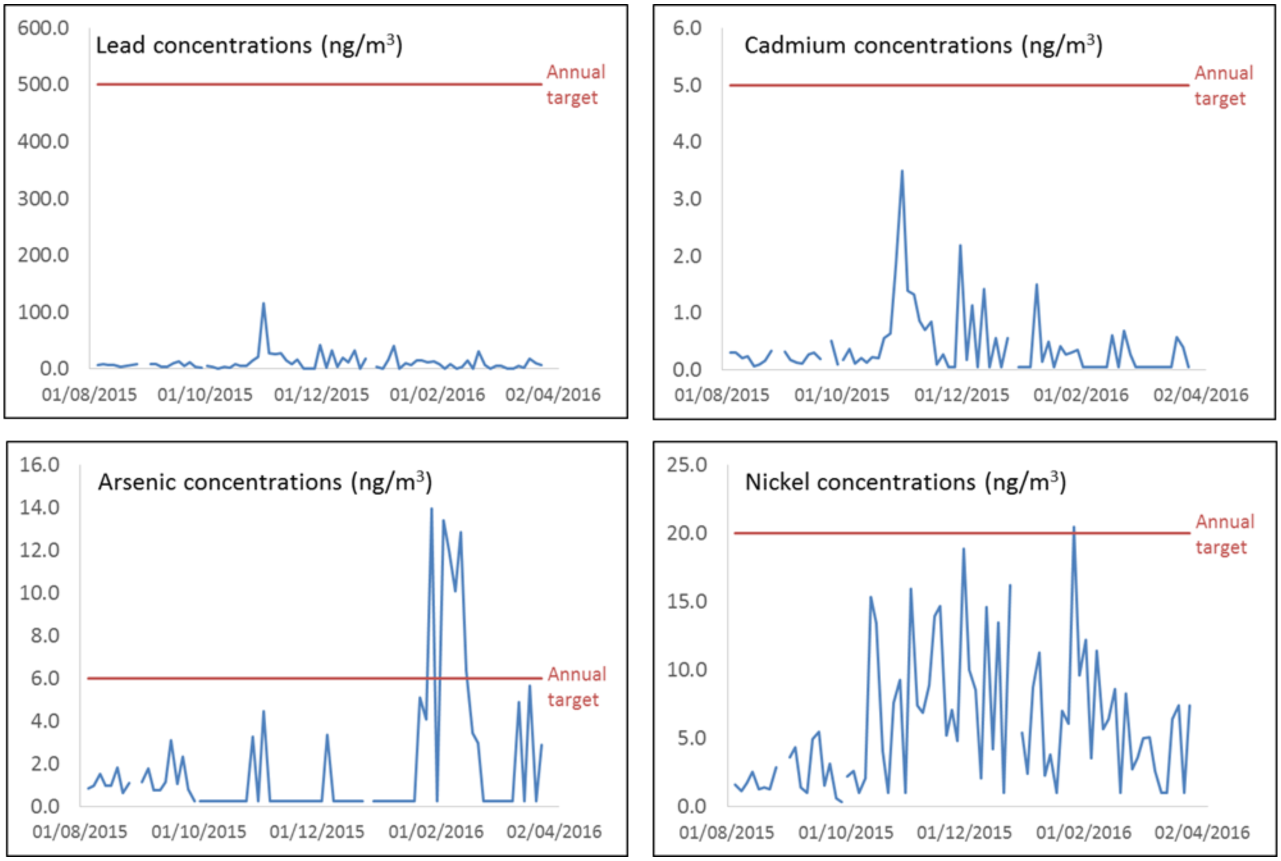

Figure 38. Concentrations of heavy metals (As, Cd, Ni and Pb) in Karpos, Skopje during the monitoring campaign in August 2015-March 2016.

Source apportionment study carried out for Karpos urban background air quality monitoring station in Skopje (August 2015-February 2016) included also an assessment of the contribution of the different emission sources to heavy metal concentrations. According to the results, the industry has largest contribution to cadmium and lead concentrations whereas nickel originates also from traffic, biomass burning and soil. Arsenic concentrations originate mostly from soil (Figure 39).

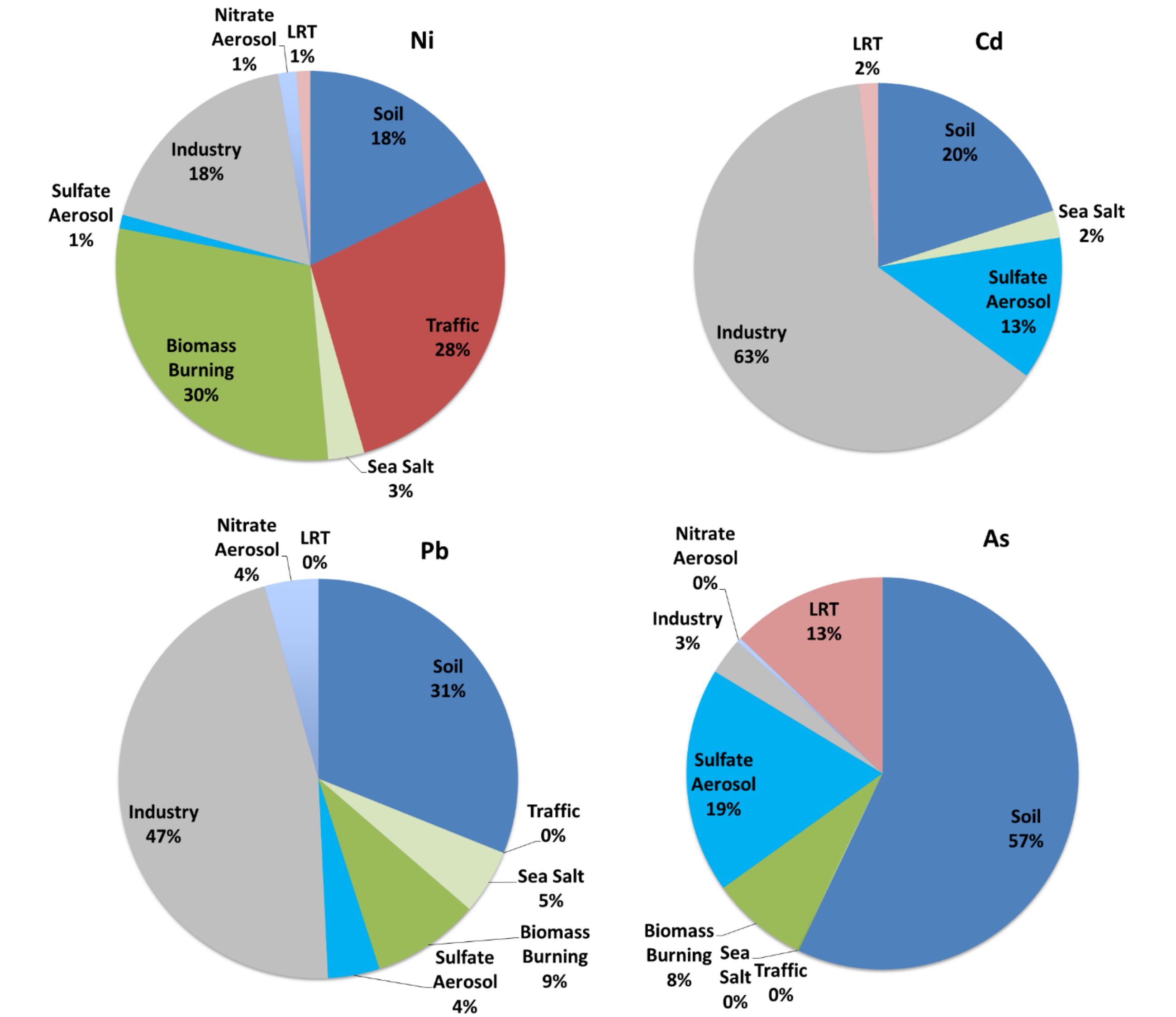

Figure 39. Contribution of different emission sources to cadmium, nickel, lead and arsenic concentrations at Karpos site, Skopje in August 2015-February 2016.

13. VOLATILE HYDROCARBONS

VOCs play a significant role in the formation of ozone and particulates in the atmosphere. Organic component in smog episodes is still poorly known. In addition, some of these VOCs, like benzene, are human carcinogens.

Volatile hydrocarbons have a key role in atmospheric ozone and secondary organic aerosol (SOA) formation. SOA forms a major part of atmospheric fine particles, especially in urban smog episodes. In urban environments, the traffic emissions are predominant man-made source of these reactive precursors of SOA. This important component that can significantly contribute to the fine particulate burden is still not well studied.

Non-methane volatile organic compounds (NMVOCs) are a group of chemically different compounds, such as benzene, ethanol, formaldehyde and cyclohexane. Air quality legislation includes a limit value for benzene concentrations in the ambient air.

13.1. Sources and emissions of benzene

The main emission sources for NMVOCs are mainly residential heating and solvent use, which contributed in 2014 with 31% and 21% respectively of the national total NMVOC emissions. Other sources of emissions are transport sector and agriculture (Figure 40). NMVOC emissions have decreased steadily mainly due to declining emissions from transport and solvent use. From 2013 to 2014 the emissions decreased by 16% also due to a reduced use of solvents as well as slightly lower emissions from the residential sector.

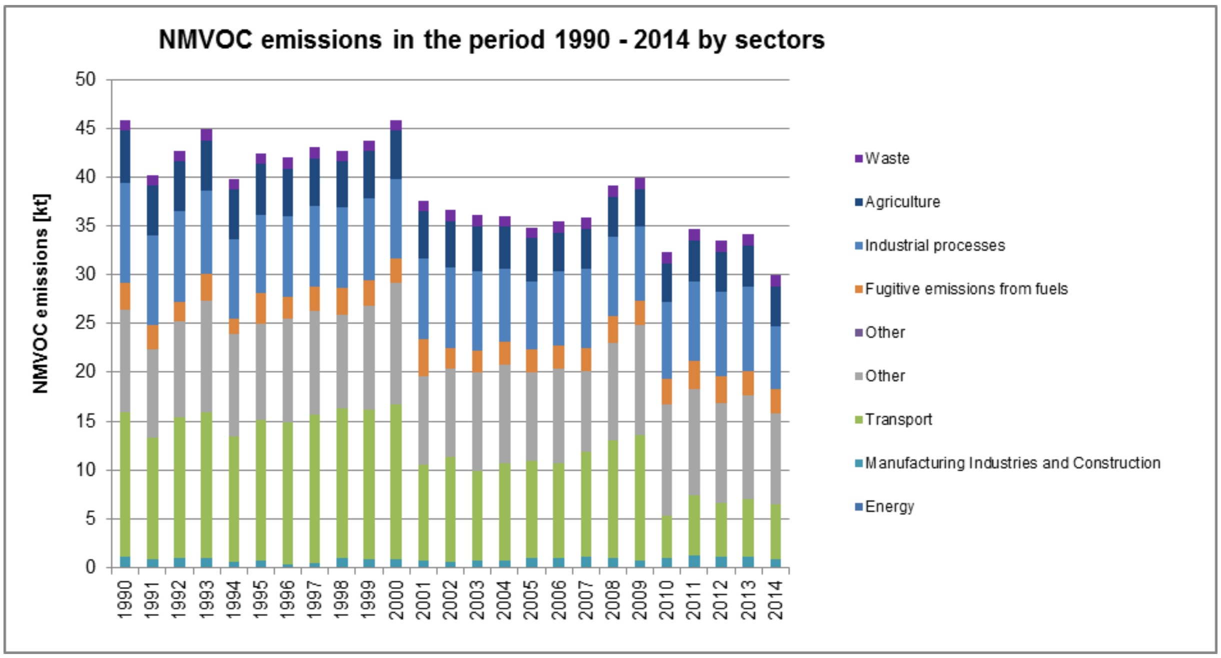

Figure 40. National NMVOC emissions 1990-2014 by emission sector (MEPP, 2016).

13.2. Air quality standards for benzene

National legislation as well as the European Union legislation defines (Directive 2008/85/EC) annual limit value for benzene (Table 8).

Table 8. Air quality standard for benzene

Objective	Averaging period	Target value
Human health protection	Calendar year	5 µg/m^3

13.3. Benzene concentrations

Benzene has been continuously measurement in Skopje on two sites in 2012-2014 (Figure 41). The limit value for benzene was exceeded in Centar station in 2013, while in 2012 both the stations recorded values very close to the limit value.

In 2011-2012 benzene was measured in two locations in Skopje, near a busy crossroad (Rektorat) close to city centre and in the vicinity of oil refinery Okta (Miladinovci) situated approximately 15 km from the city. The annual mean benzene concentrations were 3.7 µg/m^3 in Rektorat and 3.2 µg/m^3 in Miladinovci. High benzene concentrations (9-14 µg/m^3) were measured in Rektorat in November and December 2011.

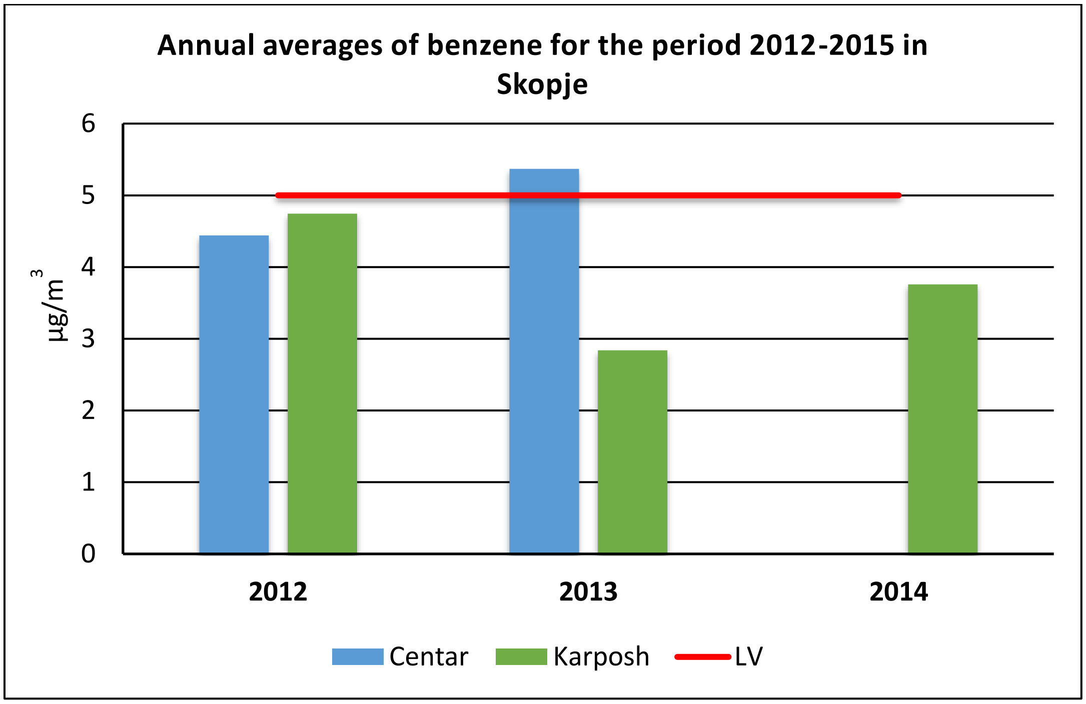

Figure 41. Benzene concentrations measured in on Skopje in 2012-2014.

14. HEALTH IMPACTS OF AIR QUALITY

Air pollution is a major environmental health problem affecting everyone in both developed and developing countries. Even relatively low concentrations of the air pollutants can cause health effects especially for vulnerable population groups.

The knowledge on the health effects caused by the exposure to different pollutants in the air is continuously increasing due to the research studies, and rising interest and awareness of the health aspect of pollutants in the air. Even relatively low concentrations of the pollutants can cause health effects especially for the vulnerable groups. Improved air quality can reduce the exposure to the pollutants and the negative health effect caused by the pollutants. Table 9 presents the main health effects of different pollutants (EEA, 2014).

Table 9. Main health effects of different air pollutants (EEA, 2014).

Pollutant	Health effects
Particulate matter (PM)	Can cause or aggravate cardiovascular and lung diseases, heart attracts and arrhythmias. Can cause cancer. May lead to atherosclerosis, adverse birth outcomes and childhood respiratory diseases. The outcome may be premature death.
Ozone (O_3)	Can decrease lung function. Can aggravate asthma and lung diseases. Can lead to premature mortality.
Nitrogen dioxide (NO_2)	Increased cardiovascular and respiratory mortality and respiratory morbidity.
Sulphur dioxide (SO_2)	Aggravates asthma and can reduce lung function and inflame respiratory tract. Can cause headaches, general discomfort and anxiety.
PAHs, especially benzo(a)pyrene	Carcinogenic
Carbon monoxide (CO)	May lead to heart disease and damage to nervous system. Can cause headache and fatigue.
Arsenic (As)	Carcinogenic. May cause lung cancer.
Cadmium (Cd)	Carcinogenic
Lead (Pb)	Can affect almost every organ and system, especially the nervous and cardiovascular systems. May have adverse cognitive effects in children and lead to increased blood pressure in adults.
Mercury (Hg)	Can affect the liver, kidneys and digestive and respiratory systems. May affect also the central nervous system.
Nickel (Ni)	Carcinogenic
Benzene (C_6H_6)	Carcinogenic

Particulates attribute to the most severe health risk from the air pollutants. A threshold for particulate concentrations below which no damage to health is observed has not been identified. The effects of PM on health occur at levels of exposure currently being experienced by most urban and rural populations in both developed and developing countries. Both short-term and long-term exposure for the particulates can cause health effects. The health effects of PM are caused after inhaling the particles. Depending on their size, particles can penetrate into lungs and blood streams causing adverse effects in the respiratory, cardiovascular, immune and neural systems. The smaller the particles are, deeper they penetrate into the lungs. Particulates mortality effects are clearly associated to the $PM_{2.5}$ fraction, which typically presents 40–80 % of the PM_{10} concentration in Europe. The mortality in cities with high levels of pollution exceeds that observed in relatively cleaner cities by 15–20%. Even in the EU, average life expectancy is 8.6 months lower due to exposure to the $PM_{2.5}$ produced by human activities (WHO, 2016; EEA, 2013).

15. AIR QUALITY IMPROVEMENT

According to the legislation, measures to improve the air quality must be implemented when limit values for protection of human health are exceeded. As described in the previous chapters, the limit values especially for particulate matter are widely exceeded in the country. In order to successfully decrease the emissions to air, efforts by central and local authorities, business sector and citizens alike are needed.

Due to the severe situation related to the particulate matter concentrations in the country and the significant contribution from domestic heating, measures should be implemented to reduce the emissions from wood burning. This reduction could be achieved with measures related to renewal of the old stoves used for heating and restrictions for wood burning. Also information measures aiming for better maintenance and use of the wood stoves are of importance together with energy saving actions. Furthermore, wider introduction of district heating can effectively decrease the use of wood for domestic heating and therefore the impacts to the air quality in urban areas.

In addition to the domestic heating, an important emission sector is road traffic, which contributes primarily to NO_2 concentrations but also to the PM_{10} and $PM_{2.5}$ concentrations. There are numerous different possibilities to decrease the traffic emissions, many of which require major investments to improve traffic networks and infrastructure in the local level. Improvement of public transportation in major urban areas, promotion of utilization of low-emission vehicles and cycling, and creation of pedestrian and low emission zones can be effective for pollution control in urban areas. The impact of the road dust can be reduced by improving the cleaning the roads especially during the dry periods. In addition, national regulations to control vehicle emissions by introducing measures to renew the vehicle fleet and regulate fuel quality are needed.

Figure 42. With long-term measures it is possible to improve the air quality and decrease the risks for human health (Photo: Aleksandar Ristovski).

Industry and energy production sectors can have a local effect for PM_{10} concentrations and contribute to the NOx, SO_2 and VOC emissions. Furthermore, the emissions from industry and energy production contribute to the formation of secondary particulates. For the industrial sector, efforts have been made to adopt and apply the pollution control regulations for the installations according to the legislation. For decreasing the emissions from industry and energy production, implementing the requirements from the legislation and environmental permitting are the most efficient way to decrease the air quality impacts.

Other emission sectors have a minor impact to the air quality, but nevertheless measures to improve waste management including enforcement or banning illegal waste burning and improvement of agricultural practices can contribute to the improvement of air quality in local level.

For successful improvement of air quality the improvement plans need to be harmonized with other policies, such as energy, climate and transport policies in national and local level. In addition urban planning can have a major role in the air quality improvement.

16. CONCLUSION

Air quality has been measured for more than 10 year period, currently in 17 locations in different parts of the country. Air quality monitoring data for the period 2005-2015 was used to analyse the trends in the air quality. The trend analysis was supported with information of the emissions from all polluting sectors and case studies prepared to analyse the contribution of different sources to air pollution.

The trend analysis was done for main pollutants which are measured continuously (particulate matter, NO_2, SO_2, O_3 and CO). For some pollutants, the statistical trend analysis was not possible due to the lack of good quality monitoring data and lack of required data coverage for the reporting period.

According to the analysis, a significant decreasing trend can be seen in the concentrations of sulphur dioxide during the 10 year period. This is due to the change of fuels used in a number of heating plants and utilization of fuels with low content of sulphur. Similar clear trend cannot be seen for the other pollutants. Particulate matter concentrations have remained in the same level during the ten year period, exceeding the limit values very significantly in all urban locations of the country. The high particulate matter concentrations pose a serious risk to the health of the population. Therefore, air quality improvement measures should be urgently implemented targeting the main emission sectors including domestic heating, road traffic and industry.

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ANNEX I EUROPEAN POLICIES FOR AIR QUALITY PROTECTION (EEA, 2014)

Policies		Pollutants							
		PM	**O_3**	**NO_2 NOx NH_3**	**SO_2 SOx**	**CO**	**Heavy Metals**	**BaP PAHs**	**VOC**
Directives regulating ambient air quality	2008/50/EC	PM	O_3	NO_2	SO_2	CO	Pb		Benzene
	2004/107/EC						As, Cd, Hg, Ni	BaP	
Directives regulating emissions of air pollutants	2001/81/EC	(a)	(b)	NOx, NH_3	SO_2				MNVOC
	2010/75/EU	PM	(b)	NOx, NH_3	SO_2	CO	Cd, TI, Hg, Sb, As, Pb, Cr, Co, Cu, Mn, Ni, V		VOC
	Euro standards on road vehicle emissions	PM	(b)	NOx		CO			VOC, NMVOC
	94/63/EC	(a)	(b)						VOC
	2009/126/EC	(a)	(b)						VOC
	1999/12/EC	(a)	(b)						VOC
	91/676/EEC			NH_3					
Directives regulating fuel quality	199/32/EC	(a)			S				
	2003/17/EC	(a)	(b)		S		Pb	PAHs	Benzene, VOC
International conventions	LRTAP	PM (a)	(b)	NO_2 NH_3	SO_2	CO	Cd, Hg, Pb	BaP	NMVOC

(a) Directives and conventions limiting emissions of PM precursors, such as SO_2, NOx, NH_3 and VOC, indirectly aim to reduce particulate matter ambient air concentrations

(b) Directives and conventions limiting emissions of O_3 precursors, such as NOx, VOC and CO indirectly aim to reduce troposphere O_3 concentrations

ANNEX II NATIONAL LEGISLATION REGARDING AIR QUALITY AND EMISSIONS

FRAMEWORK LAWS	
Law on environment	Official Gazette of RM no. 53/2005, 81/2005, 24/2007, 159/2008, 83/09, 48/10, 124/10, 51/11,123/12, 93/13, 44/15
Law on Ambient Air Quality	Official Gazette of RM no. 67/2004, 92/2007, 83/2009, 35/10, 47/11, 100/12, 163/13, 10/15, 146/15
SUBLAWS FOR AIR EMISSIONS	
Rulebook on the methodology for inventory and establishment of the levels of polluting substances emission into the atmosphere in tons per year concerning all types of activities, as well as other data to be submitted to the European Monitoring and Evaluation Program (EMEP)	Official Gazette of RM no. 142/07
Rulebook on the form, methodology and manner of keeping cadaster of polluters and pollutants	Official Gazette of RM no. 92/10
Rulebook on the limit values of permissible levels of emissions and types of polluting substances in waste gases and vapors released from stationary sources into the air	Official Gazette of RM no. 141/10
Decree on determination of large combustion capacities that should undertake measures for ambient air quality protection	Official Gazette of RM no. 112/11
Rulebook on the format and content of the forms for submission of data on ambient air emissions from stationary sources, manner and time interval of submission based on the capacity of the installation, content and manner of keeping the journal of emissions into the ambient air	Official Gazette of RM no. 79/11
Rulebook on the methods, manners and methodology of measuring the air emissions from stationary sources	Official Gazette of RM no. 11/12
SUBLAWS FOR AMBIENT AIR QUALITY	
Decree on the limit values of the levels and types of polluting substances in the ambient air and alert thresholds, deadlines for limit values achievement, margins of tolerance for the limit values, target values and long-term targets	Official Gazette of RM no. 50/05, 4/13
Rulebook on criteria, methods and procedures for ambient air quality assessment	Official Gazette of RM no. 169/13
Rulebook on the content and manner of delivery of data and information on the status of ambient air quality management	Official Gazette of RM no. 138/09
Rulebook on the methodology for ambient air quality monitoring	Official Gazette of RM no. 138/09
Rulebook on detailed conditions for performance of certain types of technical activities with regard to equipment, devices, instruments and appropriate business premises to be met by entities performing certain technical activities in the area of ambient air quality monitoring	Official Gazette of RM no. 69/11
SUBLAWS ON PLANS AND PROGRAMMES	
Rulebook on the detailed content and manner of preparation of the National Plan for Ambient Air Protection	Official Gazette of RM no. 108/9
Rulebook on detailed content and manner of preparation of the Plan for ambient air quality improvement	Official Gazette of RM no. 148/14
Rulebook on detailed content and manner of preparation of short-term action plans for ambient air protection	Official Gazette of RM no. 148/14

ANNEX III SUMMARY OF THE CONCENTRATIONS AND NUMBER OF EXCEEDANCES OF THE LIMIT AND TARGET VALUES

In all tables the annual data coverages below 70% are marked in red.

Table A1. SO_2 annual averages

Monitoring station	2005	2006	2007	2008	2009	2010	2011	2012	2013	2014	2015
Karpos	16.8	21.4	19.1	11.7				8.3	12.3		1.6
Centar	28.8	25.4	20.8	13.1				9.5	6.2	6.9	8.7
Gazi Baba			11.1	9.6	10.3	4.3	6.3	5.3	2.9	2.2	2.4
Lisice	15.5	20.7	13.9	7.5	6.0	8.8	6.0	2.6	1.4	2.0	2.9
Miladinovci					7.0	9.8	7.6	9.5	2.8	6.5	7.2
Mrsevci					5.0	0.6	5.9	4.0	1.9		
Kocani	4.9	10.0	11.9	3.9	3.7	2.3	3.1	2.9	5.1	13.5	16.0
Kumanovo	14.3	9.1	9.5	6.1	5.9						
Veles-1	29.2	27.7	17.9		12.2						
Veles-2	14.4	9.9	12.8	10.0	6.9	3.7	4.6	4.8	4.4	2.9	2.9
Kavadarci	7.7	8.2	10.3	9.2	3.5	1.2	3.6	3.4	5.7	1.6	1.0
Bitola-1	15.7	13.7	11.4	12.5	9.4	3.4	7.2		3.2	5.5	5.5
Bitola-2	4.2	2.7	3.1	3.1	1.6	0.9	1.2	2.3			
Tetovo	10.3	8.9	8.3	6.4	5.4	3.7	4.3	3.9	4.3	1.8	2.7
Kicevo	4.5	6.5	6.6	7.1	9.6	1.3			0.7	0.7	1.2
Lazaropole		3.4	3.3	2.7	2.4	3.7	4.0	5.9		1.5	1.2

Table A2. SO_2 number of exceedances of the hourly limit value within a calendar year

Monitoring station	2005	2006	2007	2008	2009	2010	2011	2012	2013	2014	2015
Karpos	10	33	14	0				0	0		0
Centar	11	13	21	0				0	0	0	0
Gazi Baba			0	0	0	0	0	0	0	0	0
Lisice	2	1	0	0	0	0	0	0	0	0	0
Miladinovci					0	0	0	0	0	0	0
Mrsevci					0	0	0	0	0		
Kocani	0	0	0	0	0	0	0	0	0	0	0
Kumanovo	0	0	0	0	0						
Veles-1	0	0	0		0						
Veles-2	0	0	0	0	0	0	0	0	0	0	0
Kavadarci	0	0	0	0	0	0	0	0	0	0	0
Bitola-1	0	0	0	0	0	0	0		0	0	0
Bitola-2	0	0	0	0	0	0		0			
Tetovo	0	0	0	0	0	0	0		0	0	0
Kicevo	0	0	0	0	0	0	0	0	0	0	0
Lazaropole		0	0	0	0	0	0	0		0	0

Table A3. SO_2 number of exceedances of the daily average limit value within a calendar year

Monitoring station	2005	2006	2007	2008	2009	2010	2011	2012	2013	2014	2015
Karpos	0	5	0	0				0	0		0
Centar	2	5	3	0				0	0	0	0
Gazi Baba			0	0	0	0	0	0	0	0	0
Lisice	0	1	0	0	0	0	0	0	0	0	0
Miladinovci					0	0	0	0	0	0	0
Mrsevci					0	0	0	0	0		
Kocani	0	0	0	0	0	0	0	0	0	0	0
Kumanovo	0	0	0	0	0						
Veles-1	0	0	0		0						
Veles-2	0	0	0	0	0	0	0	0	0	0	0
Kavadarci	0	0	0	0	0	0	0	0	0	0	0
Bitola-1	0	0	0	0	0	0	0		0	0	0
Bitola-2	0	0	0	0	0	0		0			
Tetovo	0	0	0	0	0	0	0		0	0	0
Kicevo	0	0	0	0	0	0	0	0	0	0	0
Lazaropole		0	0	0	0	0	0	0		0	0

Table A4. NO_2 annual averages

Monitoring station	2005	2006	2007	2008	2009	2010	2011	2012	2013	2014	2015
Karpos	50.8	46.2	36.3	34.2				30.0	24.7	20.5	22.4
Centar	52.1	52.8	50.3	56.7				43.1	36.9	26.3	26.8
Gazi Baba		52.8	35.8	27.4	15.1	21.6		29.4	30.4	24.7	25.0
Lisice	52.1	46.5	39.8	37.7			41.9	50.6			
Miladinovci					18.7	19.2	9.5		18.3	22.6	
Mrsevci					9.4	9.6	7.3		2.1		
Rektorat	59.2	55.1	42.8	36.7	32.0	11.0		36.5	25.5	15.4	
Kocani	18.5	15.9	15.1	12.9	11.7	12.7	30.6				
Kumanovo	28.7	23.1	25.5	22.4	18.0	12.9	8.2				
Veles-1	13.9	9.0	14.3		15.8	12.0	12.2	7.8			
Veles-2	28.3	25.6	19.6	16.6	18.5	21.5	21.3	42.4	19.5		
Kavadarci	30.3	24.6	25.9	20.4	19.5	4.4	6.0		5.4		
Bitola-1	19.8	25.4	18.7	16.8	20.4	18.1	13.7	28.0	5.6		
Bitola-2	34.3	36.8	22.6	29.9	27.1	20.4	19.5	14.0			
Tetovo	27.7	29.4	24.7	21.0	26.5	20.9	17.2		20.2	39.2	28.8
Kicevo	45.3		12.0	27.6	30.0	28.7	43.8	31.7			
Lazaropole	5.0	1.0	1.0	1.0	1.0	0.7	1.5	0.3			

Table A5. NO_2 number of exceedances of the hourly limit value within a calendar year

Monitoring station	2005	2006	2007	2008	2009	2010	2011	2012	2013	2014	2015
Karpos	0	8	1	0				1	0	0	0
Centar	7	6	8	2				13	0	0	0
Gazi Baba		0	0	0	0	0		0	7	3	2
Lisice	10	5	0	0			44	26			
Miladinovci					0	0	0		0	0	
Mrsevci					0	0	0		0		
Rektorat	8	62	1	0	2	0		1	0	0	
Kocani	0	0	0	0	0	0	0				
Kumanovo	0	3	17	3	0	0	0				
Veles-1	0	0	0		10	0	0	0			
Veles-2	4	0	0	0	0	0	0	0	0		
Kavadarci	0	0	0	0	0	0	0		0		
Bitola-1	0	0	0	0	0	0	0	0	0		
Bitola-2	1	15	0	0	0	0	0	0			
Tetovo	0	0	0	0	0	0	0		15	0	5
Kicevo	45		0	0	0	0	2	2			
Lazaropole	0	0	0	0	0	0	0	0			

Table A6. CO maximum daily 8 – hour mean within a calendar year

Monitoring station	2005	2006	2007	2008	2009	2010	2011	2012	2013	2014	2015
Karpos	4.59	5.97	6.99	4.13				5.64	7.29	5.29	5.77
Centar	8.04	9.90	7.81	7.58				6.47	9.11	6.54	6.48
Gazi Baba		5.27	6.17	5.60	4.04	3.15	4.99	6.86	8.28	7.22	8.65
Lisice	8.76	12.87	10.72	7.43	7.14	8.12	16.17	10.16	13.00	21.68	11.24
Miladinovci					2.72	2.55	5.01	4.39	2.08	2.60	2.76
Mrsevci					2.53	4.58	4.90	5.25	1.97		
Rektorat	3.97	8.40	6.77	4.81	5.14	4.33	6.25	7.43	3.88	6.62	8.81
Kocani	4.12	4.29	5.13	5.87	7.24				1.34	3.63	4.81
Kumanovo	4.28	6.20	4.73	3.09	5.30	3.66					
Veles-1	1.39	1.49	2.20		1.99	2.80	2.44	1.61			
Veles-2	3.99	4.58	3.92	3.83	3.42	4.32			1.66	4.09	5.47
Kavadarci	4.49	5.22	5.01	4.11	4.39	5.69	6.18	4.14	1.56	5.19	6.29
Bitola-1	8.88	6.91	6.62	4.38	3.71	5.92	7.10	8.45	8.06	8.53	6.04
Bitola-2	8.27	9.01	9.27	5.85	5.26	4.70					12.47
Tetovo	9.71	7.16	6.20	6.77	4.89	6.24	10.45	10.67	6.14	9.03	14.81
Kicevo	6.20	9.46	4.85	6.97	7.25	7.99	6.96		6.44	7.34	6.70
Lazaropole											

Table A7. CO number of exceedances of the maximum daily 8 – hour moving average limit value within a calendar year

Monitoring station	2005	2006	2007	2008	2009	2010	2011	2012	2013	2014	2015
Karpos	0	0	0	0				0	0	0	0
Centar	0	0	0	0				0	0	0	0
Gazi Baba		0	0	0	0	0	0	0	0	0	0
Lisice	0	9	1	0	0	0	7	1	11	8	4
Miladinovci					0	0	0	0	0	0	0
Mrsevci					0	0	0	0	0		
Rektorat	0	0	0	0	0	0	0	0	0	0	0
Kocani	0	0	0	0	0				0	0	0
Kumanovo	0	0	0	0	0	0					
Veles-1	0	0	0		0	0	0	0			
Veles-2	0	0	0	0	0	0			0	0	0
Kavadarci	0	0	0	0	0	0	0	0	0	0	0
Bitola-1	0	0	0	0	0	0	0	0	0	0	0
Bitola-2	0	0	0	0	0	0					2
Tetovo	0	0	0	0	0	0	1	1	0	0	7
Kicevo	0	0	0	0	0	0	0		0	0	0
Lazaropole											

Table A8. O_3 maximum daily 8 – hour moving average within a calendar year

Monitoring station	2005	2006	2007	2008	2009	2010	2011	2012	2013	2014	2015
Karpos	150.0	135.0	156.1	109.0				159.0	150.0	120.0	128.0
Centar								189.0	142.0	126.0	135.0
Gazi Baba											
Lisice	140.0	121.0	179.4	139.0	142.0	74.0	141.0	93.0	99.0	123.0	123.0
Miladinovci					129.0	130.0	142.0		144.0	118.0	136.0
Mrsevci											
Rektorat	97.0	146.0	164.0	153.0	138.0	120.0	108.0	112.0	102.0	116.0	109.0
Kocani	121.0	113.0	138.5	122.0	108.0	114.0	107.0	85.0	151.0	138.0	112.0
Kumanovo	169.0	166.0	197.1	177.0	166.0	149.0	156.0				
Veles-1	143.0	150.0	177.5		211.0	145.0	136.0	125.0			
Veles-2	184.0	147.0	220.6	197.0	146.0	130.0	118.0	107.0	127.0	94.0	95.0
Kavadarci	133.0	148.0	164.7	144.0	127.0	131.0	100.0	141.0			
Bitola-1	154.0	159.0	194.6	172.0	144.0	154.0	133.0	141.0	140.0	86.0	131.0
Bitola-2	152.0	156.0	157.9	172.0	153.0	161.0	144.0				150.0
Tetovo	154.0	160.0	187.9	186.0	142.0	150.0	113.0	109.0	145.0	101.0	146.0
Kicevo	126.0	136.0	184.4	155.0	138.0				100.0	99.0	104.0
Lazaropole	201.0	209.0	252.6	207.0	193.0	203.0					

Table A9. O_3 number of exceedances of the maximum daily 8 – hour moving average limit value within a calendar year

Monitoring station	2005	2006	2007	2008	2009	2010	2011	2012	2013	2014	2015
Karpos	13	5	11	0				48	32	0	11
Centar								17	6	2	2
Gazi Baba											
Lisice	8	1	26	17	10	0	19	0	0	2	2
Miladinovci					12	10	25		11	0	19
Mrsevci											
Rektorat	0	1	17	24	9	0	0	0	0	0	0
Kocani	1	0	8	1	0	0	0	0	40	7	0
Kumanovo	45	98	119	125	94	42	44				
Veles-1	45	51	51		29	20	29	1			
Veles-2	85	58	96	72	26	6	0	0	2	0	0
Kavadarci	13	23	33	29	5	4	0	6			
Bitola-1	80	60	56	54	12	23	9	14	14	0	17
Bitola-2	95	69	17	99	51	66	42				75
Tetovo	83	45	45	66	34	23	0	0	42	0	17
Kicevo	3	9	37	57	20		0		0	0	0
Lazaropole	257	276	240	186	175	110					

Table A10. PM_{10} annual averages

Monitoring station	2005	2006	2007	2008	2009	2010	2011	2012	2013	2014	2015
Karpos	91.8	89.5	78.1	74.0				71.3	65.1	60.9	57.1
Centar	104.0		65.1	77.3				82.1	76.3	65.1	72.8
Gazi Baba					84.4	65.8	133.7	99.6	67.9	84.7	80.7
Lisice			83.5	95.0	110.6	78.7	124.7	114.4	85.5	89.9	88.5
Miladinovci					56.5	50.7	61.4	57.7	42.8	53.0	41.3
Mrsevci					69.8	65.9	77.2				
Rektorat	211.0	173.2	101.9	91.5	96.0	69.1	103.1	75.0	68.3	68.7	57.5
Kocani	89.7	57.3	54.1	59.7	62.1	52.7	68.1	117.4	81.8	45.3	49.6
Kumanovo	97.0	93.7	94.2	68.1	79.4	70.5	81.9				
Veles-1	54.6	57.0	57.2		67.5	60.5	80.0				
Veles-2	80.0	79.3	88.9	64.5	63.2	57.9	70.7	61.2	51.1	60.4	52.2
Kavadarci	129.8	102.6	90.6	94.2	77.4	69.0	101.0		112.2	79.8	56.3
Bitola-1	65.9	92.7	63.0	63.3	59.7	49.9	70.7	55.8	69.2	55.9	51.2
Bitola-2	71.3	79.8	70.3	66.5	64.0	54.9	62.1	172.1			68.1
Tetovo	119.5	111.8	85.5	84.2	82.6	68.7	320.4	112.0	139.6	137.6	146.7
Kicevo	99.3	96.0	84.8	80.7	76.9	75.3	96.8	194.3	80.8	76.6	79.1
Lazaropole	15.9	13.7	16.8	18.5	16.6	17.0	14.1	12.9	21.4	15.5	13.1

Table A11. PM_{10} number of exceedances of the daily average limit value within a calendar year

Monitoring station	2005	2006	2007	2008	2009	2010	2011	2012	2013	2014	2015
Karpos	163	237	206	141				167	132	138	105
Centar	157		67	32				194	169	157	125
Gazi Baba					132	123	11	159	123	126	158
Lisice			172	162	192	190	255	263	189	213	162
Miladinovci					67	101	90	68	45	84	65
Mrsevci					143	157	132				
Rektorat	75	128	259	209	175	98	57	188	145	155	95
Kocani	185	164	126	77	113	127	183	11	19	51	133
Kumanovo	126	225	182	120	179	108	79				
Veles-1	125	176	108		126	150	60				
Veles-2	252	224	119	124	110	142	142	119	113	137	121
Kavadarci	63	311	234	242	151	217	94		124	74	147
Bitola-1	138	73	154	98	72	102	159	44	142	133	104
Bitola-2	189	191	180	87	117	140	147	69			140
Tetovo	293	303	241	174	142	162	33	193	306	299	347
Kicevo	233	230	215	167	175	173	56	4	243	217	185
Lazaropole	0	4	14	5	0	5	0	0	2	0	0

Table A12. $PM_{2.5}$ annual averages

Monitoring station	2012	2013	2014	2015
Centar	51.93	44.6	37.39	40.14
Karpos	50.53	24.03	41.58	50.51

Table A13. Benzene annual averages

Monitoring station	2012	2013	2014	2015
Centar	4.4	5.4	11.1	
Karpos	4.7	2.8	3.8	0.4