Source apportionment with receptor and MATCH modelling in Bosnia and Herzegovina

Report prepared by SMHI.

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EXECUTIVE SUMMARY

The Swedish Environmental Protection Agency (SwEPA) conducted a source apportionment study in Bosnia and Herzegovina together with the Swedish Meteorological and Hydrological Institute (SMHI) and the Institute for Public Health (IPH) in Belgrade as part of the IMPAQ project, which was funded by the Swedish Agency for International Development (Sida). This study was conducted over the course of two years (2020-2022) and included several methodological approaches.

Methodology

The purpose of a source apportionment study is to identify sources of air pollution. This particular source apportionment study was conducted using both air quality measurements and emissions inventory data. In a measurement-based source apportionment study, particulate matter (PM) samples are taken from ambient air and undergo chemical analysis. Computer modelling is then performed to attribute the particulate matter to specific emission sources. This type of study was conducted in Sarajevo, Tuzla, Zenica, Banja Luka, Bijeljina and Brod during the winter of 2020-2021 and again in Sarajevo and Banja Luka during the winter of 2021-2022. Receptor modelling with positive matrix factorization was used to determine the source apportionment.
An additional source apportionment analysis was conducted for Sarajevo and Banja Luka using emission data. This study also looked at how pollutants move within defined geographic areas around these two cities by using the MATCH model.

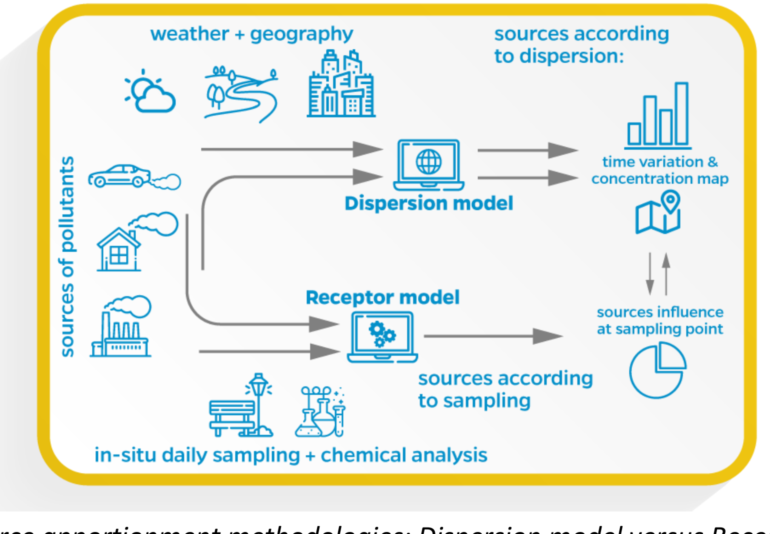
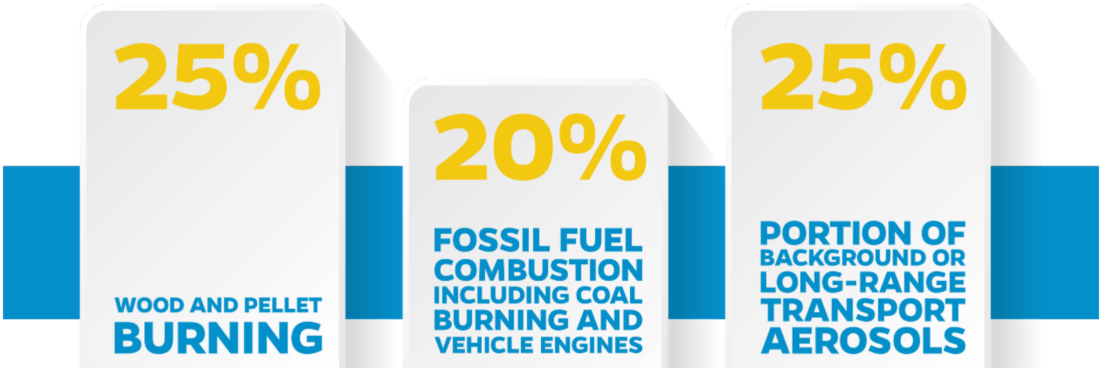


Figure 1: Source apportionment methodologies: Dispersion model versus Receptor model

Results

Positive matrix factorization with receptor modelling

The results of the winter 2020-2021 measurement campaign suggest that around 25% of PM2.5 pollution is emitted from wood and pellet burning, which are used for heating purposes. Furthermore, 20% of PM2.5 could be attributed to fossil fuel combustion including coal burning and vehicle engines. The portion of background or long-range transport aerosols represented more than 25% of the total PM2.5.



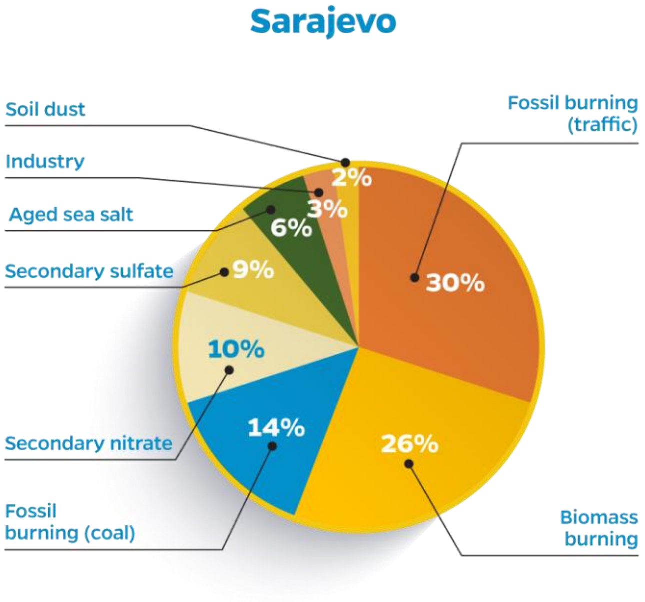
The results were then further investigated using local meteorological data, which in some cases increased the understanding of where and when different pollution sources originate from.

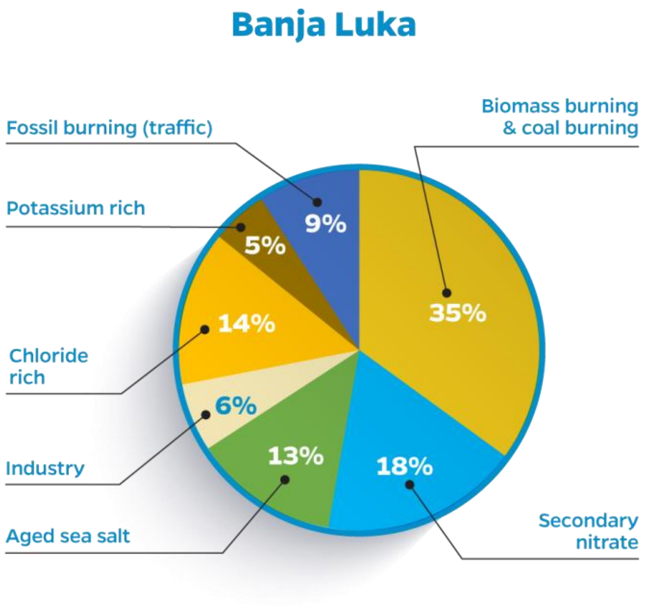
Source	Banja Luka	Bijeljina	Brod	Sarajevo	Tuzla	Zenica
Secondary Sulphate aerosols	9%	18%	5%	16%	23%*	21%
Secondary nitrate aerosols	24%*	8%	24%	14%	10%	14%
Biomass burning	19%	36%	44%*	27%	34%	54%*
Fossil fuel burning and traffic		36%*		23%	20%	
Soil dust	23%	2%	6%	8%	13%	6%
Other	25%**		21%***	12%		5%

Table 1: Source apportionment 2020-2021based on receptor modelling

*including coal burning
** 20% of this is explained by a potassium rich source and 5% by industry
***Heavy oil primary sulfate

The winter 2021-2022 round of receptor modelling for Sarajevo mainly confirmed the results of the 2020-2021 round of measurements and provided more detailed information regarding traffic and household burning. However, the level of particle pollution resulting from traffic exhaust was higher in the second round of measurements than it was in the first round of measurements. For Banja Luka, the second round of results showed higher levels of particle pollution resulting from non-combustion sources. These are not entirely explained and require further investigation.





Source	Banja Luka	Sarajevo
Fossil burning (traffic)	9%	30%
Biomass burning	35%*	26%
Fossil burning (coal)	See biomass burning	14%
Secondary nitrate	18%	10%
Secondary sulfate		9%
Aged sea salt	13%**	6%
Industry	6%	3%
Soil dust	See aged sea salt	2%
Chloride rich	14%	
Potassium rich	5%	

Table 2: Source apportionment 2021-2022based on receptor modelling

*including coal burning

**including soil dust

Some of the differences in the results from the 2020-2021 campaign versus the 2021-2022 campaign can be explained by variations in levels of air pollutants from one year to the next. The chemical analysis that was done to the filters in the 2021-2022 campaign was able to disaggregate sources more accurately than the chemical analysis from the first measurement campaign, which led to more nuanced results.

MATCH model

The source apportionment analysis based on emissions inventories suggests that the transport sector dominated the NO_2 levels, while individual residential heating dominated particle levels. This was apparent in both Sarajevo and Banja Luka. For SO_2, domestic heating contributed to a large proportion of the pollution, but a large proportion of the modelled concentrations also originated from the other sectors, mainly industry. For particles, the waste and agriculture sectors also contributed significantly to emission totals.

Sector	NOx	SO_2	PM_{10}	$PM_{2.5}$
Public Power	639.7	396.7	12.2	6.6
Other Stationary Combustion	326.3	617.1	1689.8	1601.5
Industrial combustion & processes*	89.6	911.4	431.8	160.6
Fugitives*	0	0	83.7	9.6
Solvents*	0	0	0	0
Transporst	3458.1	2.2	1418.8	1163.4
Other mobile sources*	0	0	0	0
Waste*	102.5	6.1	325.3	304.4
Agriculture*	0	0	38.1	8.4

Table 3: Total emissions (tonnes/year) of the main air pollutants for the Sarajevo model domain from local emission data set together with the CAMS regional emissions database (*).

Sector	NOx	SO_2	PM_{10}	$PM_{2.5}$
Public Power	87.4	61.4	181.1	89.1
Other Stationary Combustion	89.7	61.8	2371.8	1170.5
Industrial combustion & processes*	8.1	49	76.4	70.4
Fugitives*	0	0	0	0
Solvents*	0	0	0	0
Transporst	760.3	1.5	181.1	78
Other mobile sources*	0	0	0	0
Waste*	47.6	2.9	292.3	141.3
Agriculture*	0.5	0.1	125.6	23.4

Table 4: Total emissions (tonnes/year) of the main air pollutants for the Banja Luka model domain from local emission data set together with the CAMS regional emissions database (*).

Comparison of the results

It is useful to analyze source apportionment using both PM measurements and emissions inventories as a way of verifying and triangulating the results of the respective studies.

Source apportionment through PMF receptor modelling is a reconstruction of the most likely types of sources that influence the air quality experienced at the place where the sampling was made, and during the time when measurements were taken at that location. The result is an apportionment of type of sources and their time-variation along the measurement period.

Source apportionment through MATCH dispersion modelling is the result of a reconstruction of the dispersion and atmospheric chemical reactions of the pollutants emitted from the sources previously inventoried. The result is a map of the potential contribution to pollution levels of each type of source inventoried. The source apportionment is also shown as a time series for a specific location throughout the modelled period. This analysis provides information about the variation of source contributions on a day-to-day basis throughout the year.

A comparison of the apportionment conducted through the two different methods has been conducted for the period of the 16th of November to the 10th of March at the following locations:

- Sarajevo, in the garden of the FHMZ at Bjelave
- Banja Luka, next to the RHMZ Air Quality monitoring station Vrtić Kolibri (funded by the IMPAQ project

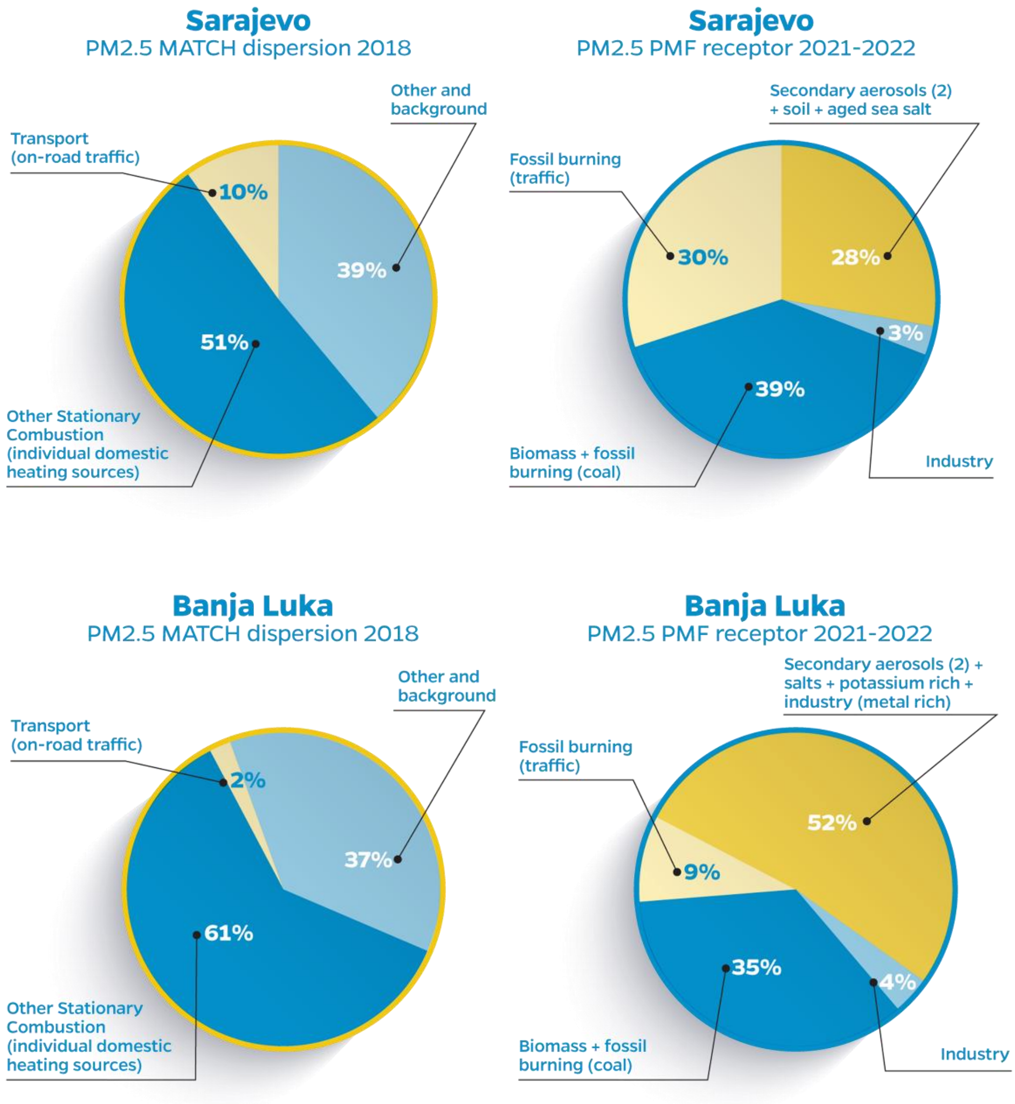


Figure 2: Comparison of dispersion versus receptor modelling

The comparison confirms that the contribution of traffic in the MATCH dispersion model is underestimated when compared with the results of the PMF receptor model. This underestimation happens in both Sarajevo and in Banja Luka and reaches a factor 3 for the PM2.5 during winter months. This means better traffic-related emissions inventories need to be compiled in relation to traffic flow and the quality of the fleet of vehicles.

For Sarajevo, there is a traffic underestimation even without the contribution of the two major other group of sources. However, the ratio for the sources "stationary combustion" and "other and background" from the MATCH Model and the equivalents from PMF model are similar for Sarajevo (51/39 ~ 39/28). This is an indication that the effort undertaken in Sarajevo to inventory the individual heating point sources was adequate.

For Banja Luka, the same ratios are not comparable. This is an indication that some important sources are missing in the overall understanding of the emission in that town. A large panel of non-burning source of particles was observed in Banja Luka in both measurement campaigns of 2020-2021 and 2021-2022. The chemical and temporal signatures of these emissions suggest that the type of source might be unusual and maybe isolated. Therefore, a field survey might help to better identify emissions sources and an abatement strategy should be feasible if confirmed that these sources of non-burning particles are only a few.

Most of the industry-related emissions are missing in the newly inventoried emissions data used for the MATCH dispersion modelling. However, the portion of particles related to industry represent only 3-4%.

Next steps

This study has provided a scientific basis for policy making. The results can be used to determine focus sectors for policy implementation, and to determine how specific actions could affect overall emission totals and their impact on air pollution levels.

INTRODUCTION

The following report is the analysis of a 2-year source apportionment study in Bosnia and Herzegovina. The study examined which pollutants are in the air, where they come from, and how they move. This report was financed by the Swedish Embassy in Sarajevo and was carried out as a part of the Swedish Environmental Protection Agency's (SwEPA) IMPAQ project.

The purpose of a source apportionment study is to identify sources of air pollution. This particular source apportionment study was conducted using both air quality measurements and emissions inventory data. These different sources of information were then analyzed using different modelling approaches. This comprehensive form of analysis has enabled the project to both verify and triangulate the source apportionment results. The emissions inventory data was also used to model how pollutants typically move within defined geographic areas around Banja Luka and Sarajevo during different times of the year.

This report is broken up into two parts, part 1 is the source apportionment study based on measurements, part 2 is the source apportionment and dispersion modeling study based on emissions inventories. The Swedish Meteorological and Hydrological Institute (SMHI) conducted the modelling for both studies, which were based on data received from measurements made by the Institute of Public Health in Belgrade (IPH), and emissions data collected by municipalities, cantons and consultants in Bosnia and Herzegovina.

PART 1: SOURCE APPORTIONMENTS RESULT FROM RECEPTOR MODELLING PMF 5.0 ON CHEMICAL ANALYSIS OF DAILY PM2.5 SAMPLES DURING WINTERS 2020–2021 AND 2021-2022 IN BOSNIA AND HERCEGOVINA

Contributors to fine particulate matter (PM2.5) have been examined in six cities in Bosnia and Herzegovina (Sarajevo, Tuzla, Zenica, Banja Luka, Bijeljina, and Brod). The analysis was carried out by a measurement campaign during the winter 2020-2021 followed by receptor-modelling from EPA's Positive Matrix Factorization (PMF) model. The same types of measurements were conducted during winter 2021-2022 in Sarajevo and Banja Luka.

Background

Ambient air pollution is a global health problem and WHO estimates that it causes around 7 million premature deaths every year. In a recent report they also concluded that the global health risk from air pollution is equally big as other factors such as unhealthy diet and tobacco smoking. WHO has set up guidelines about thresholds for clean air and according to these thresholds 99% of the world population lives in areas with poor air quality.
One among many air pollutants is fine particulate matter (PM) which are problematic since they affect the lung capacity, and lead to other negative health consequences. PM is often divided into the categories PM2.5 and PM10, which can be complex combinations of many pollution sources, for example combustion, sea salt and soil sources.

Bosnia and Herzegovina (BiH), and many other countries, struggle to mitigate high concentrations of PM pollution in ambient air. It is believed that heating of homes explains a major part of PM2.5 as many households use wood or coal as heat sources. There is a challenge in describing exactly how much of PM2.5 comes from these sources. One way of examining this problem is to chemically analyze samples of PM2.5 to determine its shares of different chemical species. Many of the species can in fact be connected to different emission sources, but the task is rather complex. Receptor modelling from EPA's Positive Matrix Factorization (PMF) can be used to discriminate between the different emission sources.

In this study, a daily collection of particles using filters for PM2.5 was done in six cities in BiH during three winter months (November, December and January). Subsequent gravimetric and chemical analyses have been conducted on each filter for many specific elements, molecules and ions. This data was then able to be used as an input for the PMF model. From the model outputs attempts have been made in order to identify partitioned emission sources.

In order to better discriminate sources in the two bigger and more complex cities of Sarajevo and Banja Luka, a second campaign was conducted during the winter 2021-2022. Some elements of the methodology have been changed in order to enhance the results as well.

Method

Sampling campaigns 2020-2021

The sampling of PM2.5 was conducted at six sampling sites in Bosnia and Herzegovina for three winter months from November 2020 to January 2021. The position of each site can be seen in Figure 3. All of them are situated in cities. The sites in Sarajevo, Zenica, Tuzla, Bijeljina and Brod are classified as urban background while the site in Banja Luka is classified as urban traffic.

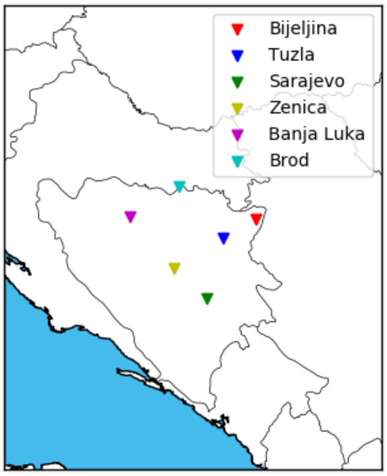


Figure 3: Sampling sites in Bosnia and Herzegovina during winter 2020-2021

Low volume samplers, Sven Leckel SEQ47/50-RV, were used in accordance with SRPS EN12341:2015 standard reference method. Maintenance, installation, and uninstallation of the samplers was provided by the official Sven Leckel distributor for Serbia.

Figure 4: Sven Leckel SEQ47/50-RV sampler and transport case

Whatman QM-A quartz filters, 47mm were used for the sampling campaign and 92 daily samples were collected during the measurement campaign.
Each sampler was equipped with two sets of filter magazines so that they could be filled with new set of unexposed filters in the controlled environment of the laboratory. At the sampling site the sampler was prepared and refilled with an interval of 14 days by an experimented team from the Institute of Public Health of Belgrade. Each time the samplers were refilled, at least one filter was not sampled and remained in the magazine to serve as a field blank. Hence the reloading induced an interruption to the sampling series and a daily sample was therefore missing for each round of sampling.

The scheduled settings during the visits were:

- Exchange of the filter magazine of the sampler done in laboratory. During transportation the magazines were covered and put into insulated containers to avoid external contamination and excessive heating.
- Change of the cleaned and pre-greased impaction plate from the laboratory.
- Change of the nozzle for a clean one.
- Check of the sampler flow rate using a regularly calibrated ORIWLOW-reference flowmeter for samplers and leak check of the sampling system.

Back at the laboratory the sampled filters were stored at a suitable temperature of ca 4°C so that loss of volatile and semi-volatile materials was minimized over the storage period.

	Sarajevo Bjelave meteorological site	Tuzla meteorological site	Zenica Brist meteorological site	Banja Luka Lazarevo meteorological site	Bijeljina meteorological site	Brod refinery meteorological site
Operator of station	Federalni hidrometeorološki zavod (FHMZ)	FHMZ	FHMZ	Republički hidrometeorološki zavod (RHMZ)	RHMZ	Optima Rafinerija nafte
Latitude, longitude	43.867743, 18.422950	44.542060, 18.685136	44.202056, 17.900428	44.793801, 17.205743	44.753659, 19.192754	45.135325, 17.982985
Type	Urban background	Urban background	Urban background	Urban Traffic	Urban background	Urban background
View of the sampler at sites						
Sampling period	2020-10-30 to 2021-02-03	2020-10-29 to 2021-02-02	2020-10-30 to 2021-02-03	2020-10-31 to 2021-02-04	2020-10-29 to 2021-02-02	2020-10-31 to 2021-02-04
Missing days (samplers reloading days)	2020-11-17 2020-12-03 2020-12-22 2020-01-11 2020-01-28	2020-11-16 2020-12-02 2020-12-21 2020-01-10 2020-01-27	2020-11-17 2020-12-03 2020-12-22 2020-01-11 2020-01-28	2020-11-18 2020-12-04 2020-12-23 2020-01-12 2020-01-29	2020-11-16 2020-12-02 2020-12-21 2020-01-10 2020-01-27	2020-11-16 2020-12-04 2020-12-23 2020-01-10 2020-01-29

Table 5: Characteristics of the sampling sites

It should be emphasized that all activities related to sampling were conducted in extraordinary circumstances during the pandemic of COVID-19 virus, with the epidemiological measures in force in Serbia and Bosnia and Herzegovina constantly changing, including tightening measures for crossing the state border (e.g. PCR testing when entering Serbia). All activities related to sampling were conducted during the winter period accompanied with a significant amount of snowfall.

Sampling campaigns 2021-2022

Additional samplings of PM2.5 were only done at two sampling sites, Sarajevo and Banja Luka, during nearly four full winter months from November 2021 to March 2022. The position of the site in Sarajevo was exactly the same. The position of the site in Banja Luka was moved 400 meters north of the 2020-2021 measuring site, beside the newly installed air quality measuring station next to the Vrtić Kolibri school. The reason for this move was that the direct vicinity of important emitters - buss parking, gas station, and pellet boiler - may have affected the representativeness of the sampling and provided results that were difficult to interpret.

In order to lower the method detection limit and get a larger range of usable species in the model, it was decided to sample simultaneously with two low volume samplers at both sites. That sampling method allowed the conduction of the analysis with larger surface of filters and thus with more substance to measure.

Gravimetric analysis

Gravimetric analysis of total mass concentration of PM2.5 was performed by standard reference method SRPS EN 12341:2015, identical with EN 12341:2014 which guarantees that all of the requirements for the method performance and quality control are met.

Filter conditioning, sampling and weighing procedures included:

- Filter conditioning and weighing prior to sampling.
- Sampling procedure.
- Filter conditioning and weighing after sampling.
- Weighing room procedures.
- Filter blanks for quality control.

Uncertainty budget for the gravimetric analysis of total mass concentration was calculated considering all individual sources of uncertainty in accordance with SRPS EN 12341:2015
The result for expanded uncertainty for gravimetric analysis was: U=(0,5+0,07*x), with x being the calculated mass concentration of PM2.5.
The Method Detection Limit was 1 µg/m^3.
Regarding the measurement campaign of 2021-2022, the gravimetric analysis has been conducted on both filters when using the same methodology as for the year before.

Filter partitioning before further analysis

After gravimetric analysis of total mass concentration was completed, further chemical and elemental analysis was performed. Each filter was split in half. From one half of the filter a punch for organic carbon (OC) and elemental carbon (EC) analysis was extracted and the rest of the filter was used for elemental analysis. From the second half of the filter a punch for ion analysis was extracted and the rest of the filter was used for the anhydrosugar analysis.

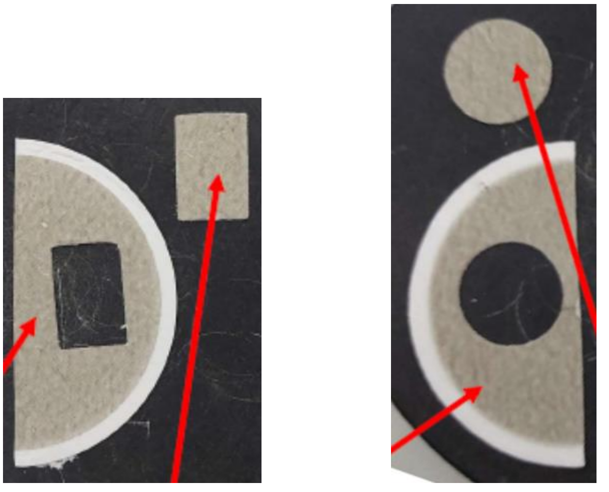


Figure 5: 2020-2021 campaign - filter cutting for analysis; 1/2 filter for elemental + punch for OC/EC (left),and 1/2 for anydrosugar + punch for ion chromatograhy (right)

During the sampling campaign 2021-2022, one of the two daily filters were used for the elemental analysis in order to get more material to analyze and to get a lower Method Detection Limit (MDL).

The second daily filter was partitioned in order to get two disc-punches for the ion chromatography, and thus a lower MDL, one rectangular punch for the OC/EC analysis and the remaining for the anhydrosugar analysis.

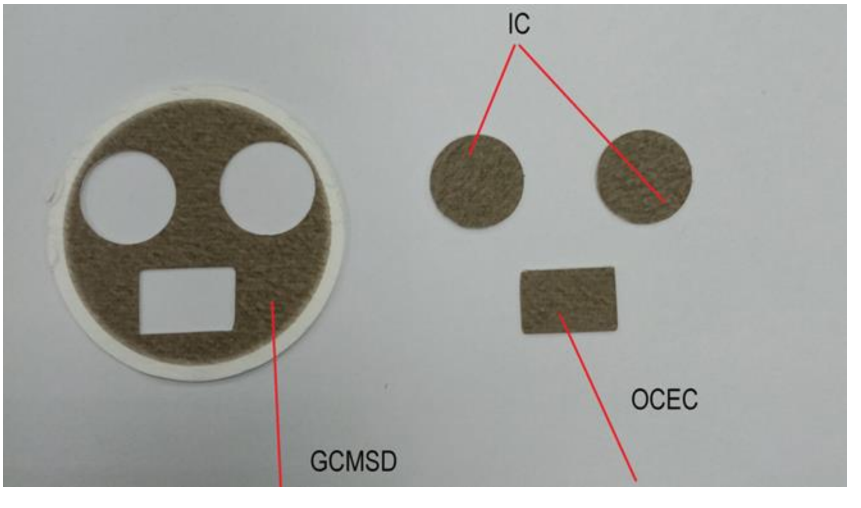


Figure 6: 2021-2022 campaign - filter cutting for analysis; remaining filter for elemental anydrosugar + 2 disc-punches for ion chromatograhy + rectangular punch for OC/EC.

Elemental analysis conducted in 2020-2021

Elemental analysis of As, Cd, Cr, Mn, Ni, Pb was based on the standard reference method SRPS EN 14902: 2008/AC:2013 Ambient air quality - Standard method for the measurement of the Pb,Cd, As and Ni in the PM10 fraction of suspended particulate matter for which the Institute of Public health of Belgrade is accredited.
The method of analysis included:

- Microwave digestion using Anton Paar equipment.
- ICP-MS analysis using Agilent ICP-MS, Series 7500, device for:
  As, Cd, Cr, Mn, Ni, Pb.
- ICP-OES analysis using Agilent ICP-OES model 5110SVDV for:
  Al, Co, Cu, Fe, V, Zn.
- Quality controls.

The uncertainties have been calculated considering random uncertainty, uncertainty of the sampling volume, non-random uncertainty of the analysis process. The results for the expanded uncertainty for elemental analysis is presented in the Table 6.

Element	U	Element	U	Element	U
As	0,00002+0,075**x*	**Cd**	0,00002+0,071**x*	**Cr**	0,00001+0,094**x*
Mn	0,112**x*	**Ni**	0,00003+0,117**x*	**Pb**	0,00005+0,082**x*
Al	0,257**x*	**Co**	0,12**x*	**Cu**	0,21**x*
Fe	0,155**x*	**V**	0,116**x*	**Zn**	0,116**x*

Table 6: 2020-2021 campaign - expanded uncertainties for elemental analysis, with x being the calculated mass concentration of each element

The method detection limits in µg/m³ is presented in the Table 7:

Element	MDL (µg/m³)	Element	MDL (µg/m³)	Element	MDL(µg/m³)
As	0,001	**Cd**	0,0001	**Cr**	0,005
Mn	0,0024	**Ni**	0,003	**Pb**	0,005
Al	23,7	**Co**	23,7	**Cu**	23,7
Fe	23,7	**V**	11,8	**Zn**	2,4

Table 7: 2020-2021 campaign - Method detection limit for the elemental analysis

Elemental analysis conducted in 2021-2022

The two same types of elemental analysis have been conducted but on one whole filter instead of less than a half and with a preference for the ICP-MS analysis much more accurate than the ICP-OES.

However, two species were not possible to analyse using the ICP-MS because of their high level in the background that affects the calibration. The Al, Fe and Zn were therefore analysed using the ICP-OES with the improvement of using a larger surface of filter in order to reach a lower detection limit, and maybe enough to effectively measure these concentrations often enough for the PMF modelling.

The method of analysis included:

- ICP-MS analysis using Agilent ICP-MS, Series 7500, device for: As, Cd, Co, Cr, Cu, Mn, Ni, Pb, V.
- ICP-OES analysis using Agilent ICP-OES model 5110SVDV for: Al, Fe, Zn.

The uncertainties have been calculated like previously. The results for the expanded uncertainty for elemental analysis is presented in the Table 8.

Element	U	Element	U	Element	U
As	0,002+0,075**x*	**Cd**	0,00002+0,071**x*	**Cr**	0,001+0,094**x*
Mn	0,112**x*	**Ni**	0,003+0,117**x*	**Pb**	0,005+0,082**x*
Al	0,257**x*	**Co**	0,12**x*	**Cu**	0,21**x*
Fe	0,155**x*	**V**	0,116**x*	**Zn**	0,116**x*

Table 8: 2021-2022 campaign - expanded uncertainties for elemental analysis, with x being the calculated mass concentration of each element

The method detection limits in µg/m³ is presented in the Table 9:

Element	MDL (µg/m³)	Element	MDL (µg/m³)	Element	MDL(µg/m³)
As	0,0005	**Cd**	0,00005	**Cr**	0,0015
Mn	0,0005	**Ni**	0,0005	**Pb**	0,0005

Element	MDL (µg/m³)	Element	MDL (µg/m³)	Element	MDL(µg/m³)
Al	6,7	**Co**	0,0005	**Cu**	0,0005
Fe	19,2	**V**	0,0005	**Zn**	16,2

Table 9: 2021-2022 campaign - Method detection limit for the elemental analysis

Ion Chromatography analysis

Ion chromatography of: SO_4^{2-}, NO_3^-, NH_4^+, Cl^-, Na^+, Mg^{2+}, K^+, Ca^{2+} was performed by an in-house method, based on the standard reference method SRPS EN 16913:2017 for which the Institute of Public health of Belgrade is accredited.

The method of analysis included:

- Ion chromatography using Methrom, model IC 930 Flex.
- Quality controls.

The uncertainties have been calculated considering the uncertainty of the sampling volume, calculated recovery based on matrix spike sample, calibration of IC equipment, and reference material on daily measurement. The results for the expanded uncertainty for ion chromatography are presented in Table 10.

Ion	SO_4^{2-}	NO_3^-	NH_4^+	Cl^-	Na^+	Mg^{2+}	K^+	Ca^{2+}
U	0,08*x	0,08*x	0,13*x	0,08*x	0,06*x	0,073*x	0,07*x	0,15*x

Table 10: Expanded *uncertainties for Ion Chromatography analysis, with x being the calculated mass concentration of each ion*

The method detection limits in µg/m³ is presented in the Table 11:

Ion	SO_4^{2-}	NO_3^-	NH_4^+	Cl^-	Na^+	Mg^{2+}	K^+	Ca^{2+}
MDL (µg/m³)	0,8	0,8	0,08	0,8	0,8	0,4	0,8	3,1

Table 11: 2020-2021 campaign - Method detection limit for the Ion Chromatography

Regarding the measurement campaign 2021-2022, the analysis was conducted with twice as many filter materials (two punches instead of one) and the method detection limit has been lowered as the same for most of the measured species.

The method detection limits in µg/m³ is presented in the Table 12:

Ion	SO_4^{2-}	NO_3^-	NH_4^+	Cl^-	Na^+	Mg^{2+}	K^+	Ca^{2+}
MDL (µg/m³)	0,4	0,4	0,04	0,4	0,04	0,4	0,4	1,6

Table 12: 2021-2022 campaign - Method detection limit for the Ion Chromatography

Analysis of the organic markers levoglucosan, manosan and galactosan

Analysis of the organic markers levoglucosan, manosan and galactosan was performed using a method that is based on the standard method VDI 2444, Ambient Measurements of Levoglucosan, Chromatographic Method, March 2020.

The method of analysis included:

- Ultrasonic extraction.
- derivatization and GCMS quantification, using Agilent GCMS – single quad 5975T.
- Quality controls.

Quality control was done according to the standard VDI 2444 and combined with quality control from the standard reference method SRPS EN 15549:2010, Air quality — Standard method for the measurement of the concentration of benzo[a]pyrene in ambient air.

It is relevant to highlight that the supervisor in charge for the Institute of Public Health of Belgrade is a member of the CEN working group CEN/TC 264/WG 21 for the development of standard method: Ambient air – Determination of the concentration of Levoglucosan – Chromatographic method, upon the call from JRC Ispra, AQUILA group.

In order to improve the quality of the data obtained for organic markers, IPH has participated in the Levoglucosan interlaboratory comparison study in the working group, which is a final step before applying for accreditation of the method.

The uncertainties have been calculated considering uncertainty of the sampling volume, calculated recovery based on matrix spike sample, mass of sampled organic marker (sampling efficiency and stability, selectivity), mass of organic marker in blank sample.

The result for expanded uncertainty, with x being the calculated mass concentration of respective hydrocarbon, was:

- U=0,1448*x for Levoglucosan,
- U=0,162*x for Manosan,
- U=0,1448*x for Galactosan,

For both measurement campaigns, the related Method Detection Limit was:

- 0,001 µg/m³ for Levoglucosan
- 0,0009 µg/m³ for Manosan
- 0,0009 µg/m³ for Galactosan

Analysis of the organic markers OC and EC

EC (Elemental Carbon) is a fraction of pure carbon usually emitted from combustion process. OC (Organic Carbon) is a fraction of carbon blended with organic components either emitted from combustion process, or as the result of atmospheric oxidation and/or condensation process.

Analysis of the organic markers OC and EC was performed by an in-house method based on the standard reference method SRPS EN 16909:2017 using the EUSAAR 2 thermal optical protocol as stated in standard reference method SRPS EN 16909 Ambient air — Measurement of elemental carbon (EC) and organic carbon (OC) deposited on filters, for which Institute of Public health of Belgrade is accredited. The in-house method is nearly exactly as the same as the standard reference method but was set before the standard method was published in Serbia. The IPH is currently in a process to update the accreditation. The laboratory inter-comparison exercises have nevertheless shown very good results of the method.

The method of analysis included:

- Lab OC-EC Aerosol Analyzer, Sunset Laboratory Inc.
- Quality controls.

The uncertainties have been calculated considering the uncertainty of the sampling volume, peak area for the relevant carbon fraction (OC or EC) measured on the loaded filter sub-sample thermogram, peak area for the calibration gas measured on the loaded filter sub-sample thermogram as well as on the external calibration standard thermogram, and the volume of external calibration standard solution analyzed.

Regarding the measurements from the 2020-2021 campaign, the result for expanded uncertainty, with x being the calculated mass concentration of respective carbon fraction, was:

- U=(0,5+0,1*x) for OC,
- U=(0,3+0,11*x) for EC.

The related Method Detection Limit was:

- 0,5 µg/m³ for OC
- 0,5 µg/m³ for EC

In order to improve the quality of the data obtained for organic markers, IPH has participated in the OC an EC interlaboratory comparison in 2021 with very good results. Following the interlaboratory comparison, the measurement get a lower method detection limit the year after. Regarding the 2021-2022 measurements campaign, the result for expanded uncertainty, with x being the calculated mass concentration of respective carbon fraction, was therefore:

- U=(0,05+0,1*x) for OC,
- U=(0,1+0,11*x) for EC.

The related Method Detection Limit was:

- 0,04 µg/m³ for OC
- 0,33µg/m³ for EC

Analytical results

Table 13 shows the average concentration in µg/m³ and associated standard deviation (SD) for all the measured species.
Depending on the chosen analytical method, many species have not been able to be measured above the detection limit (ADL) often enough to be relevant for the PMF model and nor for the calculation of an average concentration. These species are identified in the Table 13 as:

- A minus sign (-) when all concentration data was below the Method Detection Limit (MDL),
- A plus sign (+) when the number of samples ADL was 29% or less.

Regarding the Elemental analysis, nearly all of the concentration levels were below MDL when ICP-OES method was used and therefore these results could not be included in this investigation.

In addition, Zn (the only element that was enough represented when ICP-OES method was used) showed unusual high levels. The average levels of Zn from the different sites were indeed 200 to 700 times the one recently measured in the region in similar urban areas. This high level together with other weak results lead to a suspected contamination. For these reasons none of the results issued from the ICP-OES method have thus been used here after.

Regarding the Ion Chromatography, the numerous results detected under the MDL highlighted the inadequacy of the analytical method for that type of low volume PM 2.5 sampling. Na+ and Cl- were among the missing ions and they are important tracers for sea salt, which usually is a small but significant part of PM2.5.

Regarding the anhydrosugars on the other side, the analysis originally limited to the Levoglucosan has been successfully expanded in order to measure Manosan and Galactosan as well. All the results for anhydrosugar have been detected over the MDL.

	Sarajevo		Tuzla		Zenica		Banja Luka		Bijeljina		Brod	
	Average (µg/m³)	SD	Average (µg/m³)	SD	Average (µg/m³)	SD	Average (µg/m³)	SD	Average (µg/m³)	SD	Average (µg/m³)	SD
Al	-		-		-		-		-		-	
As	0,00699	0,00563	0,00686	0,00387	0,02257	0,01284	0,00211	0,00077	0,01283	0,00749	0,00686	0,00387
Cd	0,00085	0,00072	0,00044	0,00030	0,00108	0,00074	0,00061	0,00025	0,00040	0,00022	0,00044	0,00030
Co	-		-		-		-		-		-	
Cr	-		+		+		+		+		+	
Cu	-		-		-		+		-		-	
Fe	-		-		-		-		-		-	
Mn	0,00453	0,00247	0,00375	0,00183	0,03610	0,03244	0,01133	0,01224	0,00291	0,00027	0,00375	0,00183
Ni	+		+		+		+		+		+	
Pb	0,01453	0,01026	0,00960	0,00478	0,02334	0,01180	0,00929	0,00340	0,00812	0,00330	0,00960	0,00478
V	-		-		-		+		-		-	
Zn	*11,35647	8,11133	*14,72071	7,96330	*9,80347	5,13138	*7,45335	3,77896	*13,68638	7,39706	*3,83592	1,10229
SO_4^{2-}	6,72088	5,66253	8,75496	6,37894	12,97391	7,09257	6,52068	3,35438	7,18864	3,77389	5,18687	4,08349
NO_3^-	4,85086	4,07375	3,46188	2,10774	2,89716	1,57844	4,36488	2,25072	4,28666	2,32440	3,94316	2,51337
NH_4^+	3,12683	3,04969	3,42089	2,76581	4,32165	2,83087	2,24009	1,41273	3,36945	1,99995	2,52341	1,77741
Cl^-	+		+		+		+		+		+	
Na^+	+		+		+		+		+		+	
Mg^{2+}	-		-		-		-		-		-	
K^+	2,06985	1,45119	1,22536	0,42793	1,74805	0,79234	2,49399	1,62470	1,28594	0,48228	1,32356	0,57653
Ca^{2+}	-		-		-		-		+		-	
OC	16,56153	15,69400	16,98529	9,32303	20,45732	10,77313	18,84537	8,76522	15,98242	8,21045	10,50078	6,36503
EC	2,58778	1,27603	2,11548	1,06134	2,84121	0,99552	2,78565	1,00173	1,83905	0,68836	1,32261	0,81706

	Sarajevo		Tuzla		Zenica		Banja Luka		Bijeljina		Brod	
	Average (µg/m³)	SD	Average (µg/m³)	SD	Average (µg/m³)	SD	Average (µg/m³)	SD	Average (µg/m³)	SD	Average (µg/m³)	SD
Levoglucosan	1,30186	0,95600	0,98490	0,53000	1,64989	0,87665	1,62815	0,80455	1,49462	0,83845	1,52928	1,05892
Mannosan	0,25671	0,21383	0,16144	0,09212	0,31444	0,15573	0,31444	0,15573	0,19356	0,10744	0,17801	0,14014
Galactosan	0,09554	0,08524	0,06257	0,03687	0,11429	0,06338	0,11429	0,06338	0,09395	0,05295	0,07711	0,06046
PM 2.5	60,56475	53,72316	60,76448	32,20991	75,99298	36,70262	73,01956	37,40793	61,72345	26,31241	41,74085	17,57572

Table 13: 2020-2021 campaign - Averaged concentration, and standard deviation (SD), of PM 2.5 and chemical species

**Species excluded due to an overestimation artefact probably due to the measurement method.*

As for the 2021-2022 campaign, the three species measured using the ICP-OES method (Al, Fe and Zn) were nearly always below MDL and these results could not be included in this investigation. It is important to note that these three species are very common in all background including laboratories and are therefore very difficult to measure at the required low level of detection.

The Co have not been detected either despite the use of the ICP-MS method and probably because of the very level of Co in the PM 2.5. Therefore, the Co which is a tracer element of road traffic couldn't be used in this investigation.

	Sarajevo		Banja Luka	
	Average (µg/m³)	SD	Average (µg/m³)	SD
Al	+		-	
As	0,00343	0,00291	0,0012	0,00061
Cd	0,00049	0,00043	0,00048	0,00025
Co	-		+	
Cr	0,00252	0,00124	0,00578	0,00772
Cu	0,00459	0,006	0,00826	0,00735
Fe	-		+	
Mn	0,00264	0,00198	0,0087	0,00827
Ni	0,00274	0,00649	0,00366	0,00516
Pb	0,00946	0,00759	0,00837	0,00464
V	0,00141	0,00099	0,00141	0,00101

	Sarajevo		Banja Luka	
	Average (µg/m³)	SD	Average (µg/m³)	SD
Zn	+		-	
SO_4^{2-}	5,6919	4,1963	4,76817	2,66704
NO_3^-	4,83253	3,31816	4,81249	2,43852
NH_4^+	2,17844	2,49869	0,94866	0,95079
Cl^-	0,83526	0,49588	2,44878	2,48198
Na^+	0,58568	0,2104	0,64703	0,27637
Mg^{2+}	+		0,74243	0,53759
K^+	1,96936	1,82231	1,62602	1,3644
Ca^{2+}	2,54301	0,68859	7,71129	4,23865
OC	20,5448	16,4091	36,4499	18,5935
EC	3,84588	1,9912	5,91295	2,37683
Levoglucosan	1,81023	1,3573	2,82139	1,33991
Mannosan	0,22021	0,18549	0,2838	0,14972
Galactosan	0,09815	0,09007	0,13711	0,07412
PM 2.5	53,5125	41,2993	86,3229	38,7478

Table 14: 2021-2022 campaign - Averaged concentration, and standard deviation (SD), of PM 2.5 and chemical species

Analysis of total mass

Among the measured species, OC is by far the most abundant and explains a large part of the whole PM2.5. The ratio between average OC and average EC (Table 14) is always high, from 6:1 for the two biggest cities of Sarajevo and Banja Luka to over 8:1 for the smaller cities. The traffic tends to even out this ratio and is probably most significant in the biggest cities. In the same way the coal and brown coal burnings that emit more EC than OC tends to even out this ratio as well. It is possible that the burning of such calorific solid fuel, which is easier to handle and store than wood, is more common in the biggest cities. In addition, burning oil for heating is quite common depending on the city, and natural gas is used in Sarajevo as well.

The mass of OC is highly correlated with the mass of PM 2.5, except in Banja Luka. This last disconnection suggests that an important non-burning source of particles has influenced the results in Banja Luka. Furthermore, it is important to note that the percentage of average OC compared to the mass of PM 2.5 (Table 13) is quite constant, between 25 to

27%, for all the cities, including Banja Luka. This means that the non-correlation found in Banja Luka is more related to a time-series divergence between OC and PM 2.5 than the total amount of OC. In other words, there are a significant number of peaks of PM2.5 that are disconnected to the peaks of OC. A similar anomaly had been noticed at the same place in 2015 (Almeida, 2020).

	Sarajevo	Tuzla	Zenica	Banja Luka	Bijeljina	Brod
Population	642 000	80 000	75 000	250 000	50 000	71000 – incl. Slavonski-Brod
Ratio OC / EC	6,4	8,1	7,3	6,7	8,9	8,1
Correlation OC / PM 2.5	0,99	0,94	0,98	0,25	0,88	0,90
Correlation EC / PM 2.5	0,80	0,76	0,81	0,13	0,86	0,68
Σ of species [µg/m3]	37,3	37,0	46,8	38,8	35,5	26,2
% of mass explained	62%	61%	62%	53%	58%	63%

Table 15: Campaign 2020-2021 - parameters for the appreciation of total mass of PM 2.5

When comparing to a similar analysis from (Perrone, 2017), the results in BiH show 3:1 more OC among the PM 2.5 mass than in Zagreb in 2013. The same trend is noticed with the Levoglucosan and K^+ which are 4:1 to 8:1 over the levels observed in Zagreb. There is therefore a good reason to suspect that the wide range of individual household stoves with generally low temperature firebox and a high PM 2.5 emission ratio, are the most common source of particulate matters when burning wood, as well as coal and brown coal.
The sum of all the measured species reaches about 60% of the total mass of PM 2.5. In Peronne et al. 2017 66% of the total mass of PM2.5 was explained using the same method.

The total mass reconstruction might however be underestimated here since: (1) The OC mass should be converted in Organic Matter (OM) mass by using a factor between 1.4 to 1.8 (Chow, 2015) even if the anhydrosugar should be then subtracted from the OC; (2) As mentioned, some species that are used to identify soil sources are not among the detected ones; (3) Other important species that have not been detected in this study are those that are included in sea salt, especially Na and Cl, which together make up almost 80% of the composition of sea salt. According to Peronne et al. 2017, soil dust and sea salt reached a total of about 2 µg / m3 in Zagreb in 2013. This level can be assumed to be at least equivalent in BiH in 2020-2021.

The lacking species are most importantly those that have a significant part of the of the overall PM2.5 weight such as calcium, silicon, iron, and aluminum, and that have not been measured. Measuring these crustal species is difficult as they are common in the background and can interfere with the measurement. For the purpose of this study they have been considered uniformly distributed in all apportioned sources.

	Sarajevo	Banja Luka
Population	642 000	250 000
Ratio OC / EC	5,3	6,2
Correlation OC / PM 2.5	0,97	0,96
Correlation EC / PM 2.5	0,85	0,85
Σ of species [µg/m3]	45,7	69,3
% of mass explained	85%	80%

Table 16: Campaign 2020-2021 - parameters for the appreciation of total mass of PM 2.5

The total mass analysis from the 2021-2022 measurement campaign shows similar results as for the previous campaign. The OC is still the most abundant measured specie. The mass of OC is highly correlated with the mass of PM 2.5 for Sarajevo, and now also for Banja Luka.

Since the analytical methods were more accurate for the second campaign, the sum of all measured species reaches 80% to 85%. As explained before, the reconstructed total mass might be even higher since the OC mass should be converted in Organic Matter (OM) mass by using a factor between 1.4 to 1.8.

That level of total mass analytical measurement, comparable to other recent studies, makes the results of the modelling event more accurate.

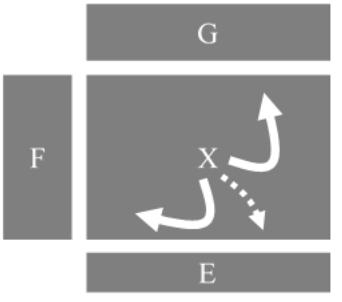
PM2.5 source apportionment was estimated by conducting receptor modelling PMF (Paatero, 1994) with the USEPA PMF v3.0 software.

Positive Matrix Factorization (PMF) modelling

The underlying principle of the receptor model is that mass conservation can be assumed, and a mass balance analysis can be used to identify and apportion sources of airborne particulate matter in the atmosphere. This PMF is a multivariate factor analysis tool that uses two matrices of the measured concentrations and of the related uncertainties, and provides families of solution that solve the mass balance equation.

$X = G \times F + E$. where:

- X is the original matrix of measurements and uncertainty,
- F is a matrix whose vectors represent the profiles of *p* sources,
- G is a matrix whose columns represent the contributions of the *p* sources,
- E is the residual matrix.



The PMF factor analysis generally produces a batch of solutions with different G and F matrices. Each solution is unique, and this is called the rotational ambiguity of the model. In order to be able to find the best-fit solution within the batch it is important to run about 100 random calculations.

Then the model assists in choosing the best solution using the objective function Q that aims to minimize the difference between the real measurements and the modelled values. This difference is represented by the residual matrix. The residual matrix is typically influenced by the outliers which are extreme values that differ from the mean trend of all the data. These outliers can either be unwanted data-contaminations or true outliers.

Choosing a solution with numerous factors leads to a quite sure solution, with a low Q. But the goal is also to connect the solution provided by the model to the reality of the environment where a few families of factors are expected or can be explained (in this study a few families of air pollutant emitters).

A useful tool for determining the best-fit solution among all the calculated solutions is then to compare the the Q expected (Q_{exp}), calculated using all the data, with the Q robust (Q_{robust}), calculated excluding the points beyond a decided uncertainty-scaled residual. The

best-fit solution is when the difference is minimal, when there were a very few points that need to be excluded (Q_{robust} shall be less than 2:1 the Q_{exp}).
After deciding for the best-fit solution, the error estimation methods included in the PMF 5.0 software should be used to confirm or reject the chosen solution.

The Bootstrap (BS) helps to detect and estimate possible random errors due to disproportionate effects of a small set of observations on the solution. It literally shows how strongly defined the factors are. An acceptable solution must have 80% of the iterative calculations providing the same mapping of factors.

The Displacement (Disp) defines the span of rotationally accessible space for the solution. The strong species have their value one by one “displaced,” a little in the profile of each factor calculated. The effect on the other factors is then observed. The idea is to see how often factors change enough to exchange identities depending on the size of the displacement. An acceptable solution shall have no swap of identity for the minimal displacement.

Within this study the result was a number of factors (5 to 6) defined by the contribution of the factor to the weight of each species (and *vice versa* the concentration of the different measured species in the factor), and the related times-series defined by the contribution of each daily measurement to the overall weight of the factor during the period.

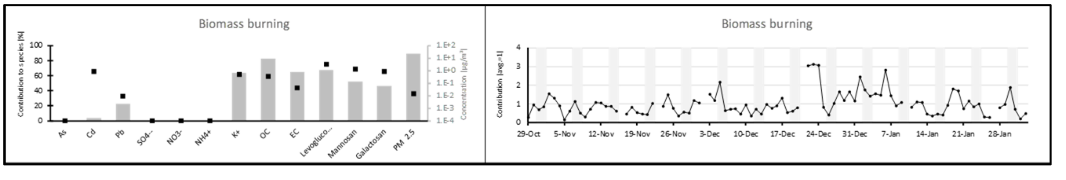


Figure 7: Typical result with factor definition (left) and times-series of the factor (right)

Scales from the left to the right: Contribution to species (%) / Concentration (µg/m³) / Contribution (average =1)

Input data and settings in PMF analysis in 2020-2021

The input data pre-processing and settings when following the PMF US-EPA guidelines are summarized in Table 17.

PMF analysis was run separately for each city. The amount of PM 2.5 samples was 92 for each of the six sampling locations. Few samples were excluded due to some very unusual and isolated events, in most cases only one isolated sample was excluded but in one occasion 5 consecutive samples were excluded. These last might have been related to a Sahara dust incursion episode that started during that period in early February 2021. The

range of modelled samples was therefore between 93% to 100%, depending on the sampling location.
The values below method detection limit (MDL) were replaced by half of the detection limit (DL/2) accordingly to the guidelines.
The number of species actually used was between 12 and 13. The species were classified as "weak" when the number of samples ADL was < 55% and "bad" when the number of samples ADL was < 35%. However, only K^+ was set as "weak" until 17% due to its importance for the discrimination of the biomass burning.
The ratio signal to noise (S/N) was also used in order to classify the species. If S/N for one specie was below 1, then it was classified as weak or bad if S/N<0.5.

The uncertainty has been provided by the IPH of Belgrade together with the measurement data and both concentrations and observation-based uncertainties were considered. The missing uncertainties related to the missing values were replaced by 5/6*DL as recommended by the guidelines.

In order to account for unknown sources of uncertainty, the analytical uncertainty provided was incremented by an extra-modelling uncertainty of 7% for all species. This is understood as making the final PMF solution stronger. For Sarajevo however, no extra uncertainty was added in order for the model to be able to converge to a solution.

A first estimation of the number of factors was accomplished by step-wise analysis of the Q value of multiple runs with increasing number of factors. The quality of the fit led to the decision of the best number of factors to be used (scaled residuals, fit of the observed versus predicted plots and histograms, $Q/Q_{expected}$ for the species). The interpretability of the results led to the decision of the number of factors (in terms of chemical profile and time series) as well as the decision to run a constrained model or not.

The best number of factors was either 5 (Bijeljina, Brod, Zenica, and Tuzla) or 6 (Banja-Luka and Sarajevo). As seen in Table 17, the PMF solution from Sarajevo did not quite reach the criteria from EPA's user guide. Though the results were improved by implementing constraints there is still one swap present from the DISP-analysis. Therefore, the results should be interpreted with this in mind.

	Sarajevo	Tuzla	Zenica	Banja Luka	Bijeljina	Brod
Period	2020-10-30 to 2021-02-03	2020-10-29 to 2021-02-02	2020-10-30 to 2021-02-03	2020-10-31 to 2021-02-04	2020-10-29 to 2021-02-02	2020-10-31 to 2021-02-04

	Sarajevo	Tuzla	Zenica	Banja Luka	Bijeljina	Brod
Missing days (samplers reloading days)	2020-11-17 2020-12-03 2020-12-22 2020-01-11 2020-01-28	2020-11-16 2020-12-02 2020-12-21 2020-01-10 2020-01-27	2020-11-17 2020-12-03 2020-12-22 2020-01-11 2020-01-28	2020-11-18 2020-12-04 2020-12-23 2020-01-12 2020-01-29	2020-11-16 2020-12-02 2020-12-21 2020-01-10 2020-01-27	2020-11-16 2020-12-04 2020-12-23 2020-01-10 2020-01-29
Nb. of samples	92	92	92	92	92	92
% ADL | S/N | chosen category (Strong, Weak, Bad)						
As	72% | 5,4 | Strong	89% | 7,1 | Strong	99% | 9,7 | Strong	**37% | 1,8 | Weak**	99% | 9 | Strong	89% | 7,1 | Strong
Cd	53% | 4,5 | Strong	**50% | 3,3 | Weak**	91% | 8,5 | Strong	59% | 4,9 | Strong	**51% | 3,5 | Weak**	**50% | 3,3 | Weak**
Mn	50% | 4 | Strong	**24% | 1,8 | Bad**	95% | 7,5 | Strong	85% | 6,8 | Strong	12% | 0,9 | Bad	**24% | 1,8 | Bad**
Pb	**66% | 4,7 | Weak**	74% | 4,5 | Strong	99% | 8,2 | Strong	**72% | 4,5 | Weak**	**79% | 4,6 | Weak**	**74% | 4,5 | Weak**
SO_4^{2-}	96% | 9,5 | Strong	99% | 9,9 | Strong	96% | 9,7 | Strong	96% | 9,7 | Strong	100% | 10 | Strong	98% | 9,8 | Strong
NO_3^-	79% | 7,9 | Strong	86% | 8,6 | Strong	87% | 8,7 | Strong	93% | 9,5 | Strong	99% | 9,9 | Strong	87% | 8,7 | Strong
NH_4^+	78% | 5,4 | Strong	90% | 6 | Strong	78% | 5,2 | Strong	74% | 5 | Strong	95% | 6,3 | Strong	71% | 4,7 | Strong
K^+	49% | 5,1 | Strong	41% | 4,1 | Strong	66% | 6,7 | Strong	78% | 7,9 | Strong	65% | 6,5 | Strong	**27% | 2,6 | Weak**
OC	100% | 5,7 | Strong	100% | 6,4 | Strong	100% | 6,7 | Strong	100% | 6,6 | Strong	100% | 6,3 | Strong	100% | 5,3 | Strong
EC	100% | 3,1 | Strong	100% | 2,7 | Strong	100% | 3,5 | Strong	100% | 3,5 | Strong	100% | 2,5 | Strong	100% | 1,8 | Strong
Levoglucosan	100% | 5,9 | Strong	100% | 5,9 | Strong	100% | 5,9 | Strong	100% | 5,9 | Strong	100% | 5,9 | Strong	100% | 5,9 | Strong
Mannosan	100% | 5,2 | Strong	100% | 5,2 | Strong	100% | 5,2 | Strong	100% | 5,2 | Strong	100% | 5,2 | Strong	100% | 5,2 | Strong
Galactosan	100% | 5,9 | Strong	100% | 5,9 | Strong	100% | 5,9 | Strong	100% | 5,9 | Strong	100% | 5,9 | Strong	100% | 5,9 | Strong
PM 2.5	**100% | 9,7 | Weak**	**100% | 10 | Weak**	**100% | 10 | Weak**	**100% | 10 | Weak**	**100% | 10 | Weak**	**100% | 9,9 | Weak**

	Sarajevo	Tuzla	Zenica	Banja Luka	Bijeljina	Brod
Reason for unusual category if any	Pb as Weak since the specie was driving the high Q/Qexp trend which prevent the model to find a stable solution.	-	-	Pb as Weak since the specie was driving the high Q/Qexp trend which prevent the model to find a stable solution.	Pb as Weak since the specie was driving the high Q/Qexp trend which prevent the model to find a stable solution.	K+ as Weak instead of Bad even if only 27% ADL, but acceptable S/N and important role in biomass burning identification. Pb as Weak for the same reason as for the other sites.
Excluded from modelling	2020-11-24 2020-01-30 2020-01-31 2020-02-01 2020-02-02 2020-02-03	2020-11-23	2020-11-21	2021-01-22	-	2020-12-05
Reasons of exclusion	24/11suspicious Mn outlier + last 6 days disturbed by Sahara dust incursion	Isolated event that prevent the model to converge to an acceptable solution.	Inconsistent level of NO3 and a high Q/Qexp trend which prevent the model to converge to an acceptable solution.	High level of anhydrosugar inconsistent with very low OC and EC that prevent the model to converge to an acceptable solution.	-	Isolated event with very isolated source of EC that prevent the model to converge to an acceptable solution.
% of tot. samples modelled	93,5%	98,9%	98,9%	98,9%	100%	98,9%

Table 17: Input data and PMF 5.0 settings

	Sarajevo	Tuzla	Zenica	Banja Luka	Bijeljina	Brod
Base runs						
Nb. of runs	100	100	100	100	100	100

	Sarajevo	Tuzla	Zenica	Banja Luka	Bijeljina	Brod
Seed number	22	22	22	22	22	22
Nb. of factors – tests solutions	4 to 7	4 to 6	4 to 7	4 to 7	4 to 6	4 to 6
Nb. of factors – final solution	6	5	6	6	5	5
Extra modelling Uncertainty	0%	7%	7%	7%	7%	7%
Choose of base run	86 instead of 80	13 instead of 22	27 as suggested	84 as suggested	81 as suggested	17 as suggested
Reason if not the suggested one	Better discrimination of the Mannosan.	Otherwise the SO4 influenced factor display a very high and isolated peak in November	-	-	-	-
Bootstrap (BS) analysis for base run						
min. nb. of BS mapped / total	**79** / 100	**66** / 100	94 / 100	85 / 100	84 / 100	99 / 100
max nb. of swap on one other factor	6	21	6	11	6	1
Displacement (DISP) analysis for base run						
Error code | 2nd valour | first raw of swaps	0 | -1,780 | 0 0 0 0 0 0	Not ran	0 | -1,788 | 0 0 0 0 0 0	0 | -0,003 | 0 0 0 0 0 0	0 | -0,053 | **5** 0 **6 3 14**	0 | -0,014 | 0 0 0 0 0
Comment on error estimation for Base run	One BS under 80%	One BS was far under 80% with 21% of swap toward one other factor	Both BS and DISP are acceptable	Both BS and DISP are acceptable	A lot of swaps (14) toward a mixed burning source	Both BS and DISP are acceptable

Table 18: Base runs diagnostics

	Sarajevo	Tuzla	Zenica	Banja Luka	Bijeljina	Brod
Rotational tools – Fpeak						
Fpeak	0	0	0	0	0	0
Rotational tools – Constraints						
Applied constraints	Mannosan pulled down (soft pulling) for all factors other than Biomass burning	Mannosan pulled down (soft pulling) for all factors other than Biomass burning	Mannosan pulled down (soft pulling) for all factors other than Biomass burning	Mannosan pulled down (soft pulling) for all factors other than Biomass burning + a mixed burning	Mannosan pulled down (soft pulling) for all factors other than Biomass burning	-
Reason for constraints	Mannosan as a reliable indicator of biomass burning should not be over-represented in other factors.	Same reason as on the left	Same reason as on the left	Same reason as on the left	Same reason as on the left	-
% of dQ [should be close as possible than 1%]	1,07%	1,11%	0,70%	0,74%	0,98%	-
Bootstrap (BS) analysis for constrained run						
min. nb. of BS mapped / total	87 / 100	93 / 100	91 / 100	99 / 100	91 / 100	-
max nb. of swap on one other factor	6	6	8	1	8	-
Displacement (DISP) analysis for constrained run						

	Sarajevo	Tuzla	Zenica	Banja Luka	Bijeljina	Brod
Error code | 2nd valour | first raw of swaps	0 | -27,478 | **1** 0 0 0 **1** 0	0 | -0,905 | 0 0 0 0 0	0 | -5,670 | 0 0 0 0 0 0	0 | 0,000 | 0 0 0 0 0 0	0 | 0,000 | 0 0 0 0 0	-
Comment on error estimation for constraint run	Only one swap between a Cd related factor and the crustal factor.	Both BS and DISP are acceptable	Both BS and DISP are acceptable	Both BS and DISP are acceptable	Both BS and DISP are acceptable	-

Table 19: Constrained runs settings and validations

Input data and settings in PMF analysis in 2021-2022

The input data pre-processing and settings when following the PMF US-EPA guidelines are summarized in Table 20.

The PMF analysis was run separately for the cities of Sarajevo and Banja Luka, as it was for the previous campaign. Only the differences from the method outlined above are here after described.

The number of PM 2.5 daily samples (as a reminder, one daily sample include two filters since two samplers operated simultaneously at each place), was 108 for each site. Few daily samples were excluded due to some very unusual and isolated events, as was the case for 2 samples in Sarajevo and 4 in Banja Luka. The proportion of modelled samples was therefore 98% and 96% respectively.

The number of species actually considered was 19 for Sarajevo and 20 for Banja Luka. The species were classified as "weak" when the number of samples ADL was < 55% and "bad" when the number of samples ADL was < 35%. Exception was granted for two species in Sarajevo because of their importance to the discrimination of some sources. The NH_4^+ has been set as "strong" even though the ADL was 54,6%, and the Cl^- was set to "weak" even though the ADL was 31,5%.

To account for unknown sources of uncertainty, the analytical uncertainty provided was incremented by an extra-modelling uncertainty of 4% for all species.

The best number of factors for a reliable solution was 8 for both Sarajevo and Banja-Luka.

	Sarajevo	Banja Luka
Period	2021-11-16 to 2022-03-10	2021-11-16 to 2022-03-10
Missing days (samplers reloading days)	2021-12-03 2021-12-04 2021-12-22 2021-12-23 2022-01-12 2022-02-01 2022-02-21 2022-02-22	2021-12-03 2021-12-21 2021-12-22 2022-01-11 2022-01-31 2022-02-20 2022-02-21
Nb. of samples	108	108
% ADL | S/N | chosen category (Strong, Weak, Bad)		
As	92% | 5,0 | Strong	93% | 2,9 | Strong
Cd	93% | 6,3 | Strong	99% | 7,5 | Strong
Cr	42% | 2,6 | Weak	65% | 4,7 | Strong
Mn	87% | 6,9 | Strong	99% | 7,9 | Strong
Ni	44% | 0,9 | Weak	82% | 2,5 | Strong
Pb	98% | 5,4 | Strong	**99% | 5,7 | Weak**
Co	0% | 0 | Bad	1% | 0,1 | Strong
Cu	97% | 3,7 | Strong	**96% | 3,6 | Weak**
V	43% | 3,2 | Weak	88% | 6,7 | Strong
Al	2% | 0 | Bad	0% | 0 | Bad
Fe	0% | 0 | Bad	3% | 0,1 | Bad
Zn	5% | 0,3 | Bad	0% | 0 | Bad
Cl^-	**32% | 3,1 | Weak**	71% | 7,1 | Good
NO_3^-	89% | 9,0 | Strong	99% | 9,9 | Strong
SO_4^{2-}	100% | 10,0 | Strong	100% | 10,0 | Strong

	Sarajevo	Banja Luka
Na^+	99% | 9,9 | Strong	100% | 10,0 | Strong
NH_4^+	**55% | 3,6 | Strong**	30% | 1,8 | Weak
K^+	48% | 4,8 | Weak	63% | 6,2 | Strong
Mg^{2+}	20% | 2,1 | Bad	54% | 5,2 | Weak
Ca^{2+}	31% | 1,8 | Bad	**85% | 4,8 | Weak**
OC	100% | 8,6 | Strong	100% | 8,8 | Strong
EC	100% | 6,0 | Strong	100% | 6,8 | Strong
Levoglucosan	100% | 5,9 | Strong	100% | 5,9 | Strong
Mannosan	100% | 5,2 | Strong	100% | 5,2 | Strong
Galactosan	100% | 5,9 | Strong	100% | 5,9 | Strong
PM 2.5	**100% | 9,7 | Weak**	**100% | 9,9 | Weak**
Reason for unusual category if any	NH_4^+ moved from Weak to Strong, Cl- moved from Bad to Weak since these species are important to discriminate respectively some secondary aerosols and aged see salt or soil dust.	Cu moved from Strong to Weak, Pb moved from Strong to Weak Ca moved from Strong to Weak since these three species were poorly predicted by the model and had their Q/Qexp over the wanted threshold of 2.
Excluded from modelling	2022-03-02 2022-01-01	2021-11-16 to 2021-11-19
Reasons of exclusion	03/02 suspected Mn outlier 01/01 suspected Cu outlier	19/11 Extremely high isolated Na+ peak 16 to 18/11 Many unmapped $SO4^{2+}$ during the beginning of the measurement which is not extremely significant for the study.
% of tot. samples modelled	98%	96%

Table 20: Input data and PMF 5.0 settings

	Sarajevo	Banja Luka
Base runs		
Nb. of runs	100	100
Seed number	22	22
Nb. of factors – tests solutions	6 to 10	6 to 10
Nb. of factors – final solution	8	8
Extra modelling Uncertainty	4%	4%
Choose of base run number	88 as suggested by the model	40 as suggested by the model
Reason if not the suggested one	-	-
Bootstrap (BS) analysis for base run		
min. nb. of BS mapped / total	86 / 100	99 / 100
max nb. of swap on one other factor	5	1
Displacement (DISP) analysis for base run		
Error code | 2nd valour | first raw of swaps	0 | -0,002 | 0 0 0 0 0 0 0 0	0 | -0,018 | 0 0 0 0 0 0 0 0
Comment on error estimation for Base run	Both BS and DISP are good	Both BS and DISP are good

Table 21: Base runs diagnostics

Optimisation of model from “base solution” to “constrained solution”

In the Positive Matrix factorization model, the best solution obtained was not unique. Because of the free rotation of matrices there was a family of solutions that were equally fit because of the so-called rotational ambiguity.

In this work all solutions from the 2020-2021 sampling campaign (except Brod) have been “constrained”, considering that manosan is an accurate tracer of biomass burning even in a region were brown coal (lignite) is a common fuel. In the chosen constrained solutions manosan was pulled down maximally (“soft pulling”, dQ = 0,50%) in factors where the contribution from biomass burning shouldn’t appear.

Thanks to a wider panel of measured species, the two solutions from the 2021-2022 sampling campaigns didn’t need any constrains to be optimized since the base runs successfully passed the BS and DISP quality tests. Hence, they are both labelled “Base solution”, as Brod was in the first winter.

Results

Display of the results

The result for each sampling site was a number of factors (5 to 6) defined by the contribution of the factor to the weight of each species (and *vice versa* the concentration of the different measured species in the factor), and the related times-series defined by the contribution of each daily measurement to the overall weight of the factor during the period.

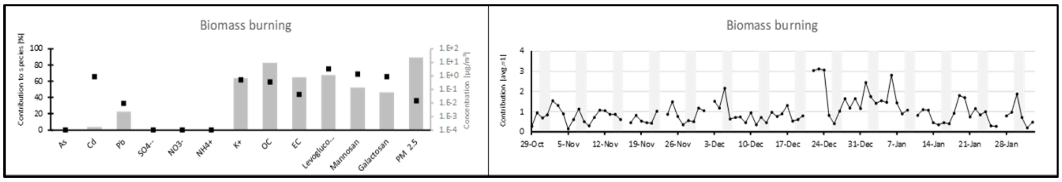


Figure 8: Example of result with factor definition (left) and times-series of the factor (right)

Scales from the left to the right: Contribution to species (%) / Concentration (µg/m³) / Contribution (average =1)

On the left side of Figure 8 the black dots show the contribution of the factor (here biomass burning) to the total weight of each species. In this example 60% of the OC was found in the biomass factor. This mean that the OC is meaningful to explain the sources that form the biomass burning factor.

On the left side of Figure 8 the grey bars show the concentration of each species in the apportioned factor. In this example the concentration of levoglucosan is about 1 µg/m^3 while the concentration of Mannosan is about 0,1 µg/m^3.

On the right side of Figure 8 the dots show how the part of the factor modelled that day compares to its cumulative weight during the whole period. In this so-called times-series example, while the daily portion of factor is 1% in average, a few days around the 24th of December show 3% of the whole biomass burning factor calculated for the 91 accumulated days. Since the scale of that graphic is adaptative, it means that when the scale is lower (up to 4%) one can say that the modelled factor should be related to a rather continuous source, while if the scale is higher (about 8%) the modelled factor should be related to a discontinuous source.

The right side of Figure 8 also shows grey bars representing the weekends. A factor which emissions normally would be more substantial during business days is expected to show a decrease during these days. As an example, the emissions from traffic usually show a weekly decrease during at least one day of the weekend.

The shows the daily relative contribution figured in a colored scale with similar boundaries for the times-series itself. Each day is represented by a point where the position depends on the wind direction origin (north is up) and the wind speed (from the centre). The points are then merged in a single pattern. These so-called polar-plots were produced while using the Openair R package developed for the purpose of analyzing air quality data (Carslaw & Ropkins, 2012).

The range of colors is the result of a Nonparametric Wind Regression (NWR) gaussian smoothing that weight concentrations on a surface according to their proximity to defined wind speed and direction intervals. The NWR have been chosen since the number of measurements was limited. Because of the smoothing effect on extreme values, only the pattern of color should be interpreted and not the intensity of the red and blue spots.

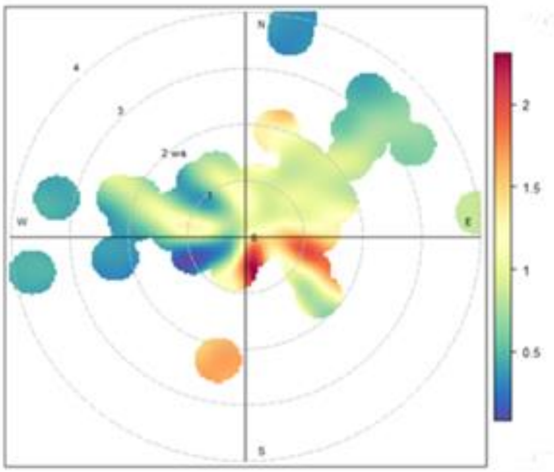


Figure 9: Polar plot of the times-series

Identification of different sources – campaign 2020-2021

The identification and naming of the likely sources for each factor provided by the model solution has been made using the range of relative mass observed for these species in the chemical profiles of Specieeurope (Pernigotti, 2016) and various literature related to similar socio-geographic environments (see REFERENCES, p.125).

The discrimination between the factors was made by isolating known fingerprints from different sources. When it comes to regional sources, secondary aerosols are common and often in form of SO_4^-, NO_3^- and NH_4^+. See more in the section about secondary aerosols.

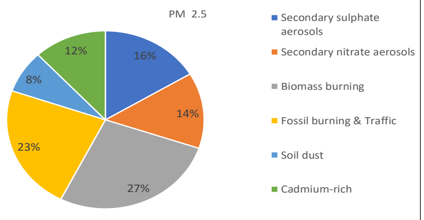
Biomass burning is one factor that used to be obvious in many cities. This factor is well identified through the high contribution of OC, usually higher than the EC, and which typically occurs together with a clear contribution of K^+. As illustrated in the literature the best marker for the biomass burning factor is Manosan together with Galactosan. Another factor that is always present is soil dust. It is well identified by the large contribution of As. In the Balkan region, As is quite common in mines and slope cuttings, and topsoil and is therefore a good marker of a crustal source. Other factors show more variability in their fingerprints in our study, probably because of a blend of sources that wasn't possible to discriminate further with the set of species that have been successfully measured.

The signal for fossil fuel burning is quite often a blend of different sources such as coal burning, possibly oil burning and probably traffic exhaust. The set of species makes it hard to discriminate traffic from other fossil fuel burning sources. Coal burning, however, typically has a higher contribution of EC than OC together with a high contribution SO_4^{2-}. It is however important to note that brown coal (lignite) emits less SO_4^{2-} than coal and that both types are burned in BiH.

Occasionally there are fingerprints with a high a contribution from Mn and these factors have so far been attributed to industry. On one occasion (Banja-Luka) there was a fingerprint with a high K+ contribution that couldn't be attributed to a known source so far.

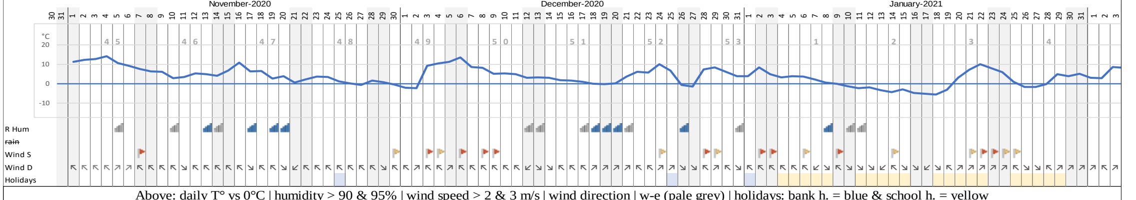
Sarajevo – constrained solution, winter 2020/2021

Sarajevo - Constrained solution



Above: distribution of the sources of PM 2.5 mapped by the model

Above: position of the sampler during winter 2020-2021

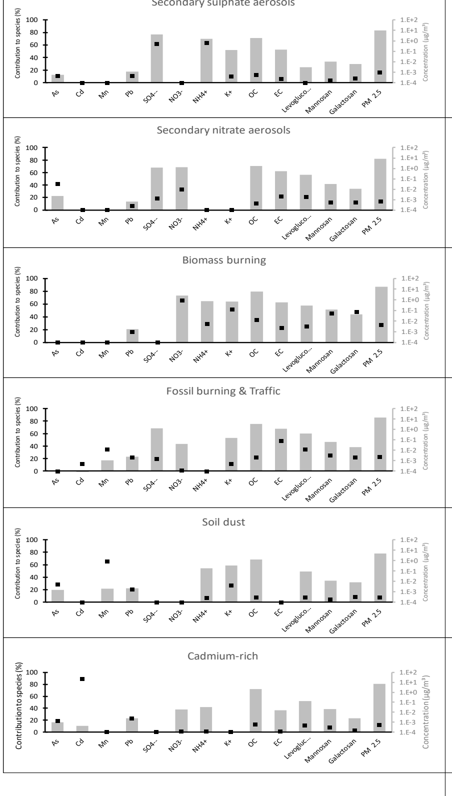


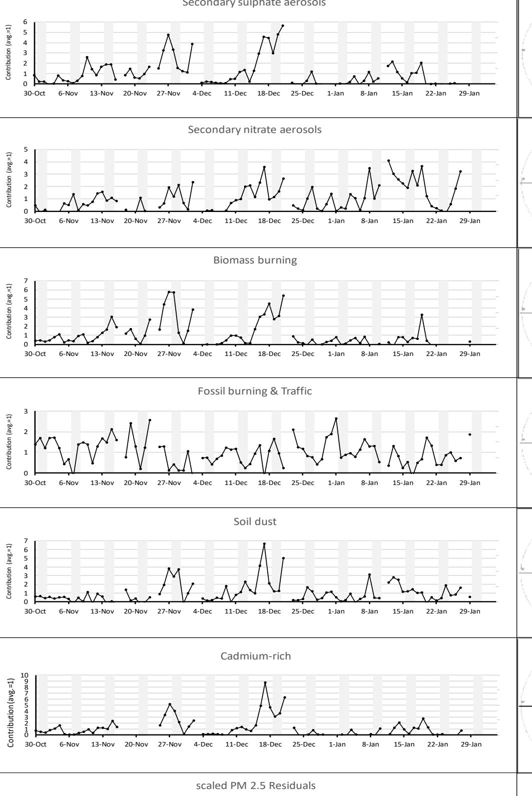
Above: daily T° vs 0°C | humidity > 90 & 95% | wind speed > 2 & 3 m/s | wind direction | w-e (pale grey) | holidays: bank h. = blue & school h. = yellow

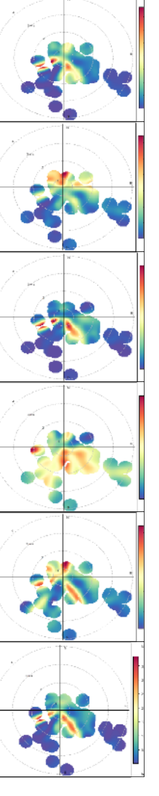
Below: source profile with percentage contribution to species (black dots) & concentration (grey bars)

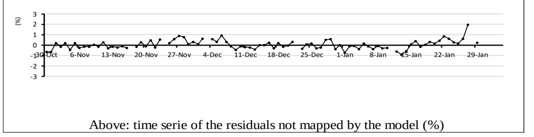
Below: time series of the source concentration (adaptative scale)

Below: concentration (colour) of factor depending on wind direction & speed





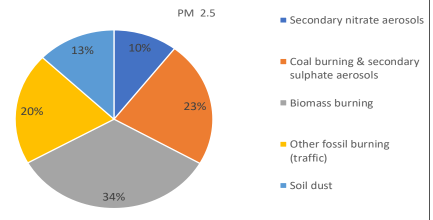




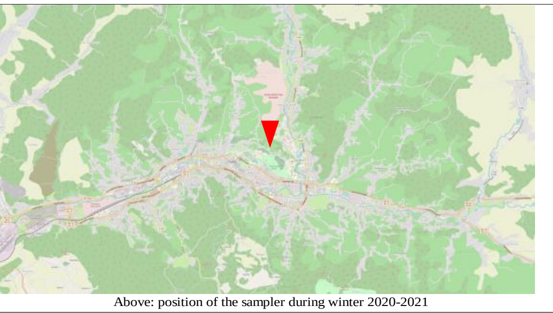
Above: time serie of the residuals not mapped by the model (%)

Tuzla – constrained solution, winter 2020/2021

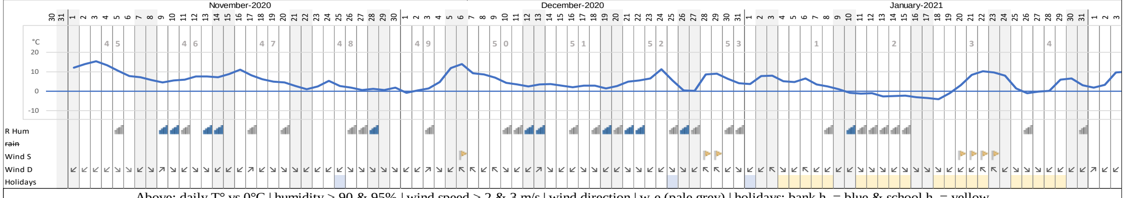
Tuzla - Constrained solution



Above: distribution of the sources of PM 2.5 mapped by the model



Above: position of the sampler during winter 2020-2021

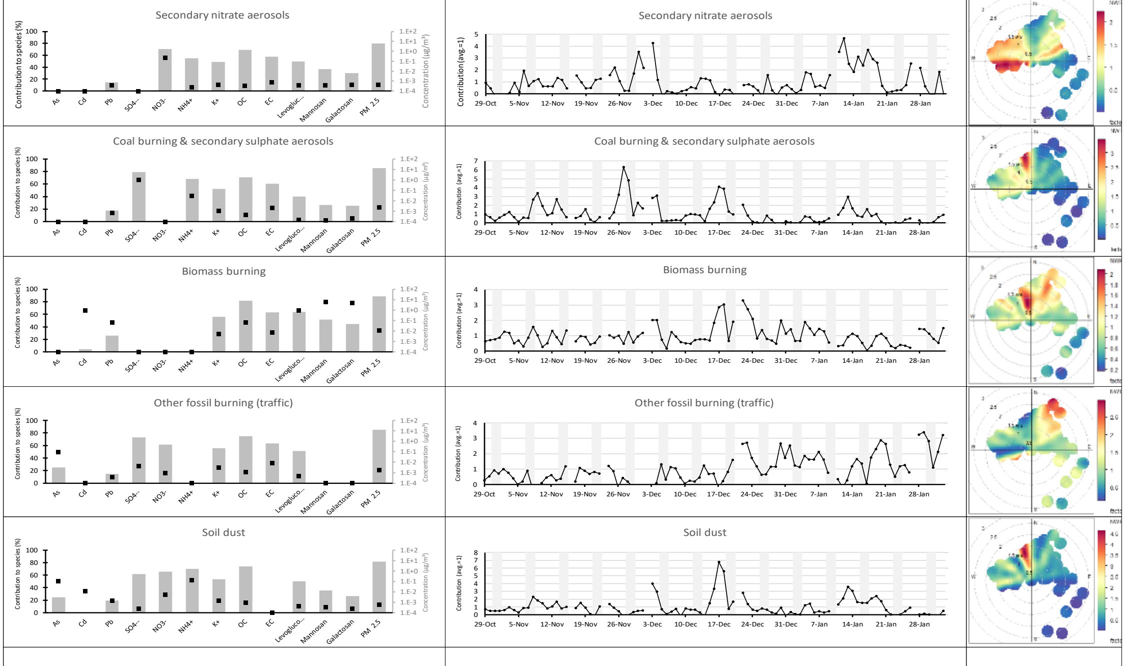


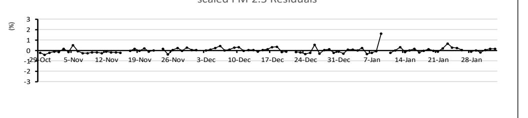
Above: daily T° vs 0°C | humidity > 90 & 95% | wind speed > 2 & 3 m/s | wind direction | w-e (pale grey) | holidays: bank h. = blue & school h. = yellow

Below: source profile with percentage contribution to species (black dots) & concentration (grey bars)

Below: time series of the source concentration (adaptative scale)

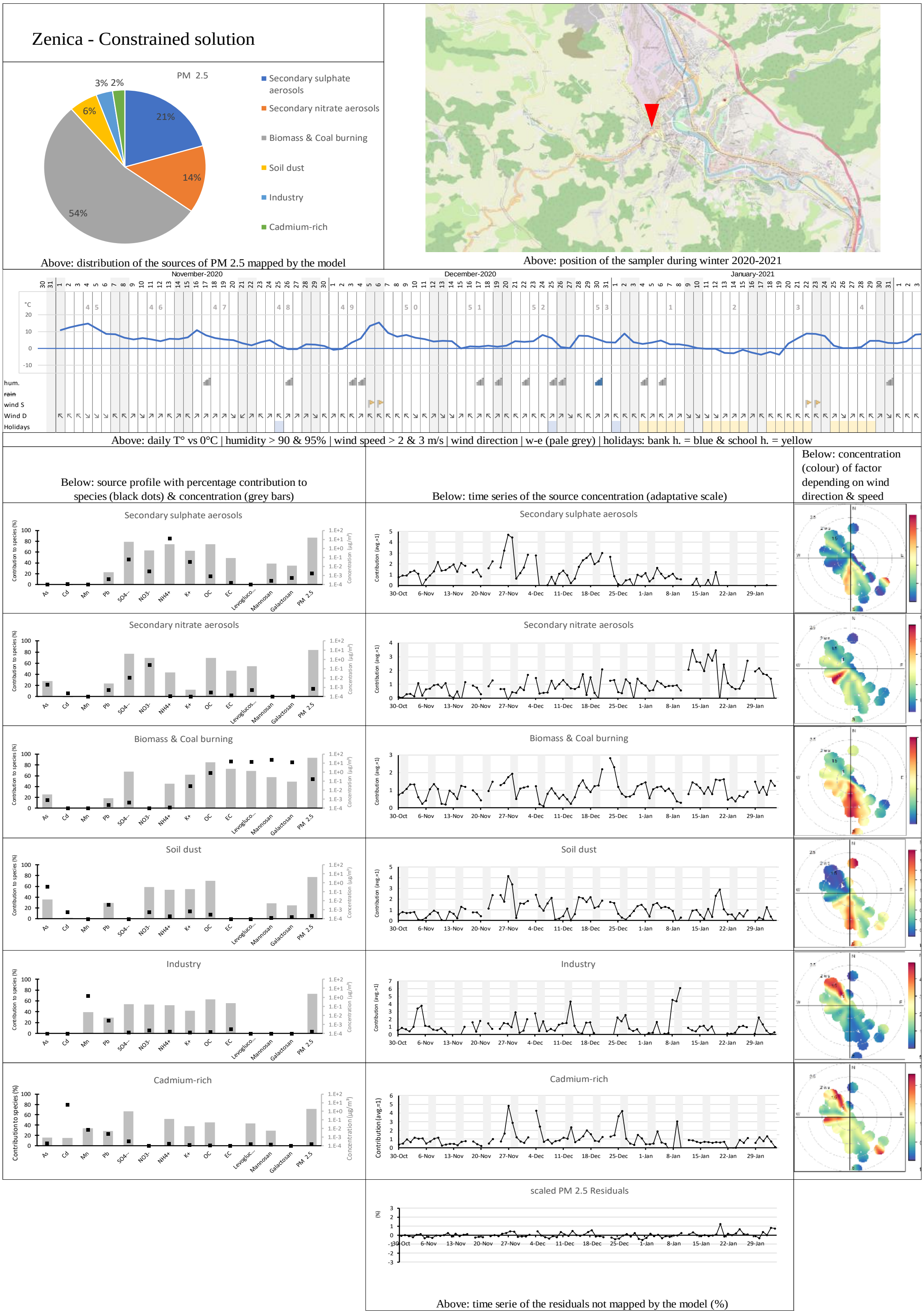
Below: concentration (colour) of factor depending on wind direction & speed





Above: time serie of the residuals not mapped by the model (%)

Zenica – constrained solution, winter 2020/2021



Above: distribution of the sources of PM 2.5 mapped by the model

Above: position of the sampler during winter 2020-2021

Above: daily T° vs 0°C | humidity > 90 & 95% | wind speed > 2 & 3 m/s | wind direction | w-e (pale grey) | holidays: bank h. = blue & school h. = yellow

Below: source profile with percentage contribution to species (black dots) & concentration (grey bars)

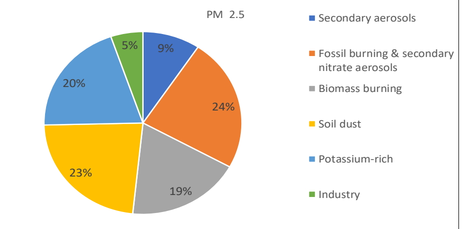
Below: time series of the source concentration (adaptative scale)

Below: concentration (colour) of factor depending on wind direction & speed

Above: time serie of the residuals not mapped by the model (%)

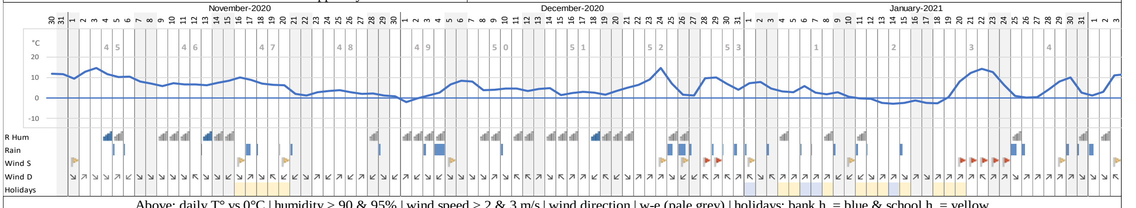
Banja Luka – constrained solution, winter 2020/2021

Banja Luka - Constrained solution



Above: distribution of the sources of PM 2.5 mapped by the model

Above: position of the sampler during winter 2020-2021

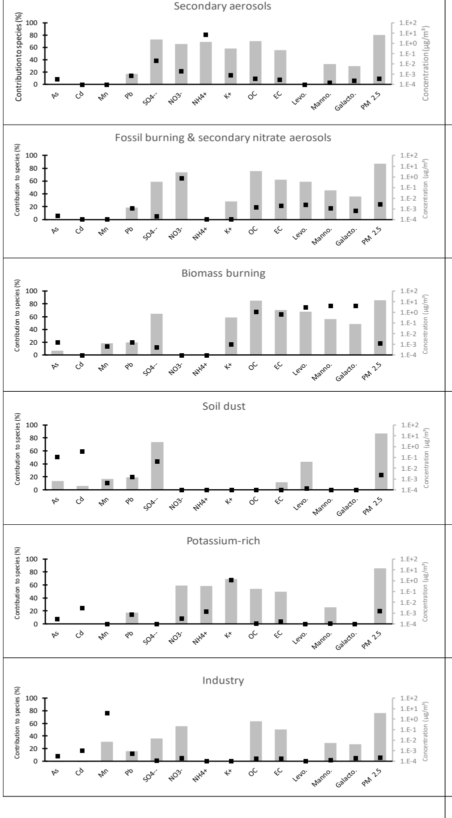


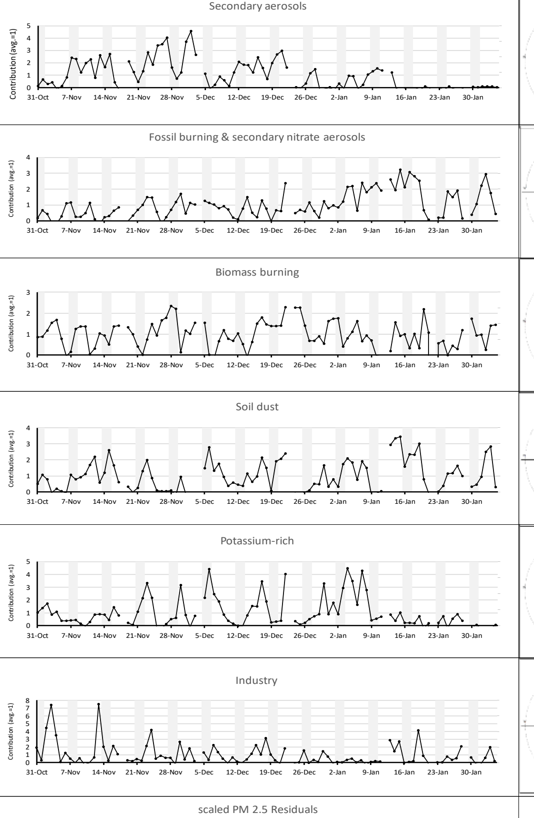
Above: daily T° vs 0°C | humidity > 90 & 95% | wind speed > 2 & 3 m/s | wind direction | w-e (pale grey) | holidays: bank h. = blue & school h. = yellow

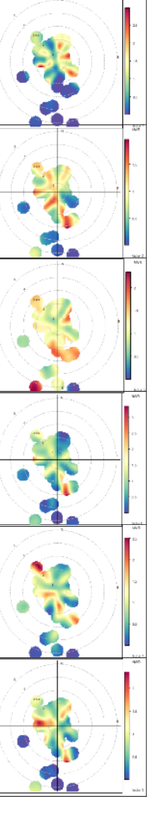
Below: source profile with percentage contribution to species (black dots) & concentration (grey bars)

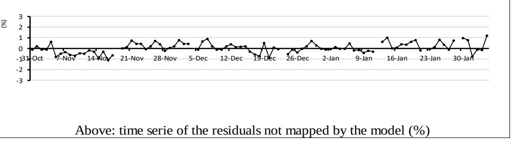
Below: time series of the source concentration (adaptative scale)

Below: concentration (colour) of factor depending on wind direction & speed





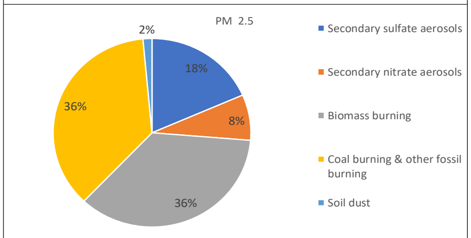




Above: time serie of the residuals not mapped by the model (%)

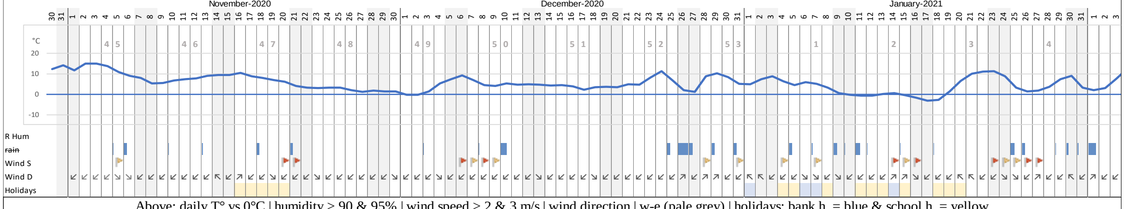
Bijeljina – constrained solution, winter 2020/2021

Bijeljina - Constrained solution



Above: distribution of the sources of PM 2.5 mapped by the model

Above: position of the sampler during winter 2020-2021

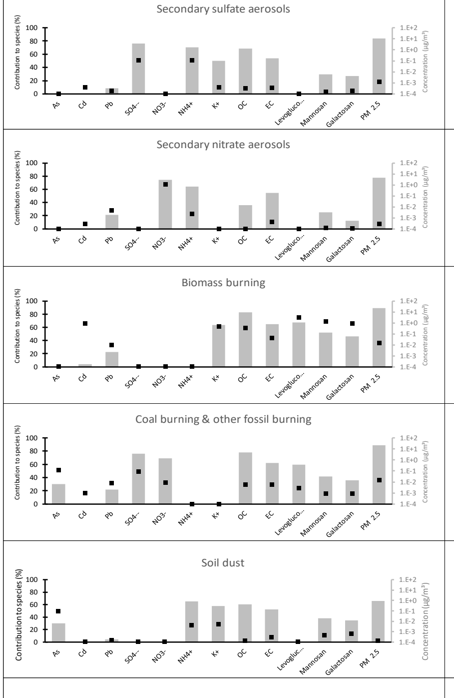


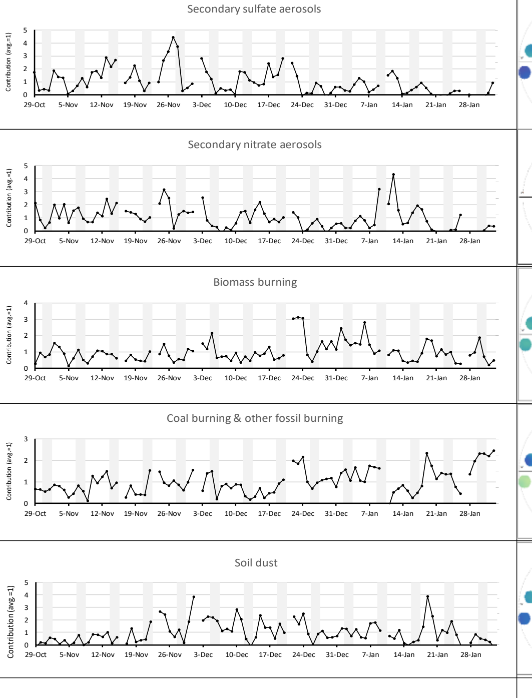
Above: daily T° vs 0°C | humidity > 90 & 95% | wind speed > 2 & 3 m/s | wind direction | w-e (pale grey) | holidays: bank h. = blue & school h. = yellow

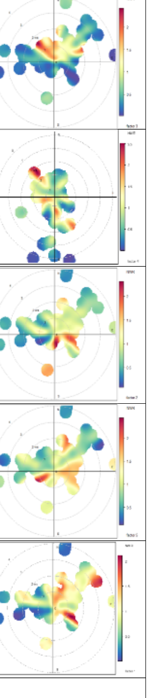
Below: source profile with percentage contribution to species (black dots) & concentration (grey bars)

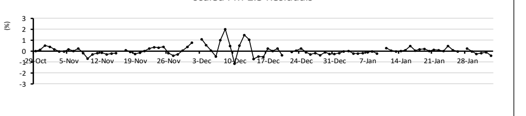
Below: time series of the source concentration (adaptative scale)

Below: concentration (colour) of factor depending on wind direction & speed





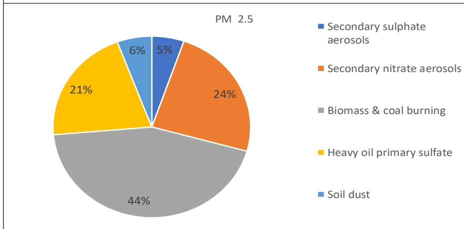




Above: time serie of the residuals not mapped by the model (%)

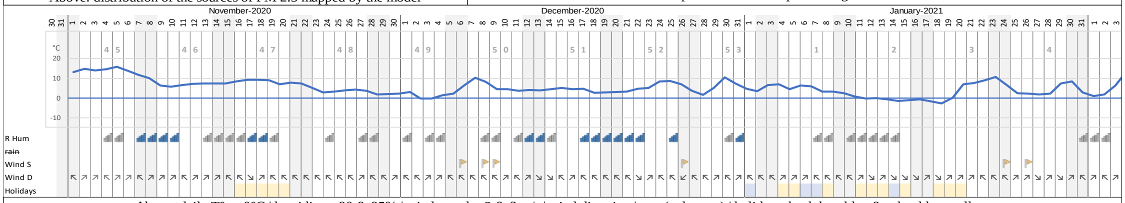
Brod – base solution, winter 2020/2021

Brod - Base solution



Above: distribution of the sources of PM 2.5 mapped by the model

Above: position of the sampler during winter 2020-2021

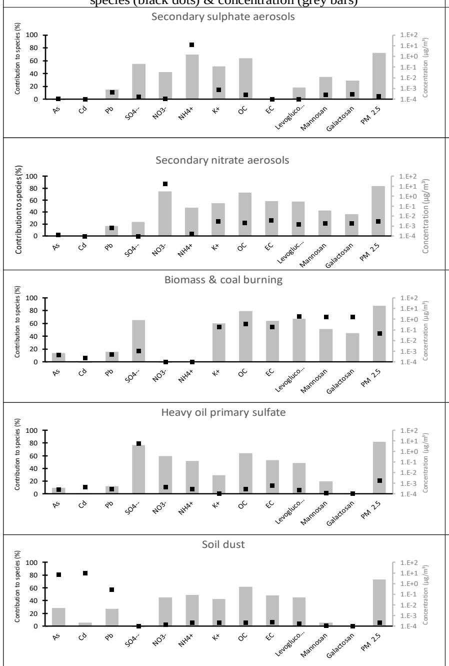


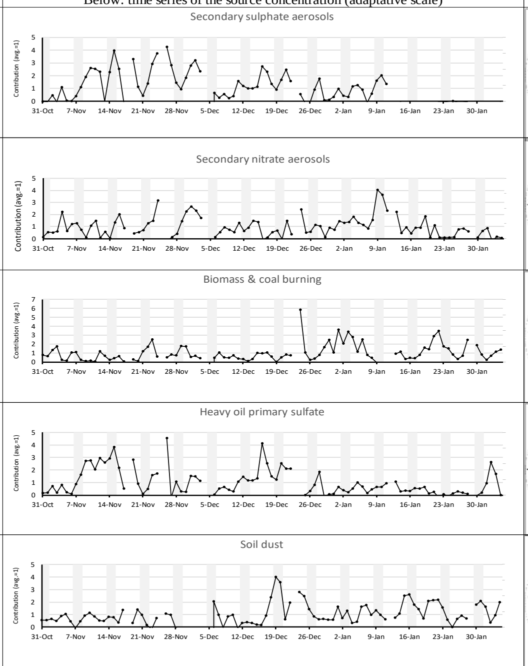
Above: daily T° vs 0°C | humidity > 90 & 95% | wind speed > 2 & 3 m/s | wind direction | w-e (pale grey) | holidays: bank h. = blue & school h. = yellow

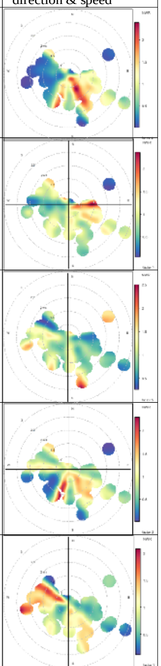
Below: source profile with percentage contribution to species (black dots) & concentration (grey bars)

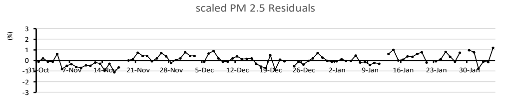
Below: time series of the source concentration (adaptative scale)

Below: concentration (colour) of factor depending on wind direction & speed









Above: time serie of the residuals not mapped by the model (%)

Secondary aerosols

Two ions, sulphate (SO_4^{2-}) and nitrate (NO_3^-), were analyzed from the filter samples and used as representatives for the secondary inorganic aerosols (SIA), principally existing together with ammonia ions. SIA is formed in the atmosphere through the transformation of gas phase precursors – principally NH_3, SO_2 and NOx - emitted both by anthropogenic and biogenic sources (Amato, 2016). The SIA gas-phase formation can take from a few hours to a few days depending on the weather conditions (e.g.; temperature, humidity), solar radiation and the concentration of different oxidants. With high aerosol water content (AWC), e.g.; during haze conditions, there are also highly effective aqueous-phase transformation processes producing sulphate and nitrate (Chunrong Chen, 2021). One could thus expect variations in the SIA components that respond to different meteorological conditions (Ogulei, 2006) (Prakash, 2017) and stable atmospheric inversion episodes (Srivastava, 2018).

The Hydrological and Meteorological Institute of the Federation of Bosnia and Herzegovina (FHMZ) and the Hydrological and Meteorological Institute of Republika Srpska (RHMZ) provided observations and oral communication about the weather situation that occurred during the monitoring campaign. November and December 2020 often exhibited a high relative humidity, fog and/or overcast weather conditions. January, on the other hand, showed more clear weather conditions with variable precipitation. Figure 10 shows that sulphate levels were significantly higher during November-December in all six cities. For nitrate the results were more diverse.

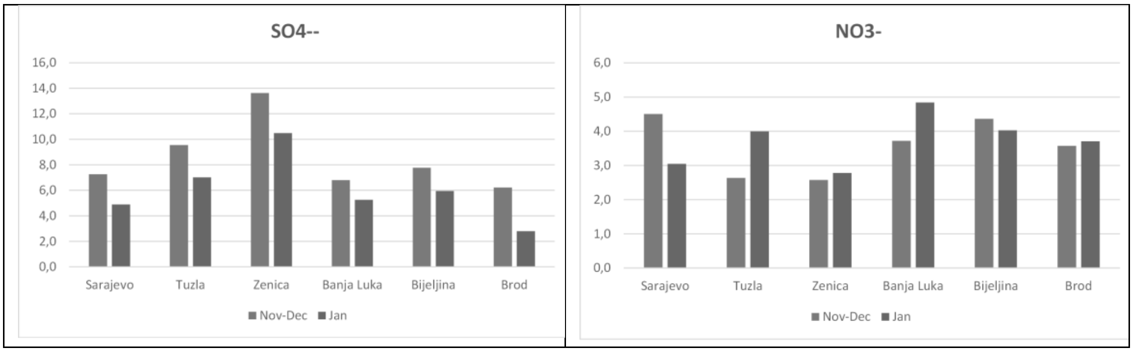


Figure 10: Concentrations (µg/m³) of sulphate and nitrate during November-December 2020 and January 2021

All combustion of sulphur-containing fuels emit SO_2, but also a fraction of sulphate. The latter is normally considered small. In dispersion modelling the Sulphur emissions from industrial ovens are typically set to 95% as SO_2 and 5% as SO_4^{2-}. However, a study from China (Dai Q, 2019) from an area with extensive residential coal combustion indicated that the primary emitted sulphate could raise up to 40-50% during wintertime. Since we have a similar emission pattern in BiH, we could expect that a significant part of the sulphates is directly emitted and not secondarily formed in the atmosphere. This is likely to produce PMF results with more overlap between sulphate and other locally emitted residential heating markers such as OC and EC.

	Sarajevo	Tuzla	Zenica	Banja Luka	Bijeljina	Brod
Factor: Secondary sulphate aerosol	16%	23% (shared with coal burning sources)	21%	9%	18%	5%
Factor: Secondary nitrate aerosol	14% (shared with fossil burning sources)	10%	14%	24% (shared with fossil burning sources)	8%	24%

Table 22: Secondary aerosols factors comparison

Brod showed a factor strongly pointing to ammonium which explained about 80% of all measured ammonium. This factor could be named spontaneous primary ammonium because of this strong and isolated signal. The contribution of that factor to the SO_4^{2-} seems to be negligible. However, the relative concentration of SO_4^{2-} seems to be like the ones within the fingerprints of secondary sulphate aerosols displayed for other places like Zenica. It is therefore possible that the strong emission of sulphate, specific to the Brod location, and related to the heavy oil has evened out the sulphate contribution in this first factor that would otherwise be much higher. That is why this first factor is called primary sulphate factor, even if the contribution of SO_4^{2-} seems to be negligible.

Biomass Burning

Biomass burning is often an important source of PM 2.5 particles in the Balkan region and is frequently related to old fashion stoves, low temperature fireboxes, moist wood and hard wood. The classical fingerprints of biomass burning when running PMF model are a significant contribution of OC and K^+ together with anhydro-sugar like levoglucosan, manosan and galactosan. Biomass burning often shows an EC contribution as well, but always with a lower contribution compared with OC.

It is meaningful to underline that the most common burnt wood in BiH is beech and that the combustion of that wood emits 10 times less levoglucosan per mass of burnt wood than other hard woods like the oak (Collet S, 2016). In addition, it has been shown in Poland that the combustion of brown coal (in particular lignite) can produce a high signal of Levoglucosan on the contrary for Manosan and Galactosan (Rybicki, 2020). Since there are several brown coal mines in operation in BiH and an active market for brown coal (Eurostat, 2021) these are common sources of energy even for individual household and particularly in towns. However, different policies and incentive programs conducted during the last decade have accounted for a progressive shift to biomass burning, in particular pellet burning.

Because of the reasons mentioned above, Levoglucosan wasn't chosen as a primary indicator of biomass burning but instead Manosan was chosen since it is more closely related to biomass burning.

In the table below the average contribution from biomass burning is listed, in addition to weeks of peak events. In some of the cases these peaks coincide with low temperatures, which should make sense since the need for heating increases. However, this is not always the case and there are other meteorological factors than temperature that can affect the factor contribution such as events of atmospheric inversions, wind patterns and precipitation.

	Sarajevo	Tuzla	Zenica	Banja Luka	Bijeljina	Brod
Factor: Biomass burning	27%	34%	54% (shared with coal burning source)	19%	36%	44% (shared with coal burning source)
Peak period (weeks number)	48, 49, 51, **52**	49, 51, **52**	48, **52**, 3	48, **52, 3**	**52, 53, 1**	**52, 53, 1**, 2
Period with temperature near or below 0°C (weeks number)	48, 49, 51, **2, 3, 4**	**48**, 49, 51, **2, 3, 4**	48, **49, 51, 2,** 3, **4**	48, **49, 2**	**48, 49, 2**	**49**, 2, **3**

	Sarajevo	Tuzla	Zenica	Banja Luka	Bijeljina	Brod
				The importance of a non-burning source specific to the site in Banja Luka has probably evened out the relative size of the biomass burning factor because of the unusual counter weight of the non-burning.		

Table 23: Biomass factor comparison

Fossil Fuel Burning

Fossil fuel burning is characterised by a higher contribution of EC compared with OC, and a generally low contribution of K^+. The major sources of fossil fuel burning particles in BiH are coal and brown-coal burning, in power-plant and district heating or individual household heating, together with diesel and gasoline engines. The range of measured species does not permit the clear discrimination of coal sources from liquid fuel sources unless their time series differ from each other. Coal burning sources are expected to have their peaks related to the coldest periods whereas traffic exhaust sources are expected to decrease during the weekend. Fuel burning for heating purposes is however common in BiH, which made even harder to isolate traffic exhaust.

For these reasons the model didn't often succeed in separating fossil fuel burning sources from other sources. Therefore, the analysis has been divided and distributed in the below table in order to easily compare the underlying principles.

When the factor has a significant contribution to the SO_4^{2-} then the reasonably suspected fuel is coal or brown coal (Prcanovic, 2018).

	Sarajevo	Tuzla	Zenica	Banja Luka	Bijeljina	Brod
Fossil burning (coal/brown coal)		23% (shared with Secondary sulphate aerosol)	54% (shared with Biomass burning)	24% (shared with secondary nitrate aerosol)	36%	
		High contribution to NH_4^+ and SO_4^{2-} that suggests a secondary sulphate aerosol source. But the time-series shows peaks correlated to the coldest weeks and the factor contribute to the EC as well. For these reasons this factor is suspected to be a blend of coal burning and sulphate aerosol.	Very high contribution to anhydrosugar and OC suggests Biomass burning source. But the very high contribution to EC shows that fossil-burning sources constitute a major part as well, probably about ½ of it. The time-series displays a quite constant source (there are no significant peaks, the max value was at 3%) Polar-plot shows a spreading from the direction of the university and up-hill settlements.	High contribution to NO_3^- that suggest a secondary nitrate aerosol source. But the significant contribution to EC combined with the lack of other possible factors suggest that a fossil-burning source is blended in this factor.	Significant contribution to EC and to SO_4^{2-} and As suggests a coal burning source. The time-series displays a quite constant source. Polar-plot shows a spreading mostly from the direction of the town.	

Table 24: Fossil burning (coal / brown coal) factor comparison

Fossil burning & traffic	23%	20%				
	High contribution to EC and significant to metals & anhydrosugars suggests both exhaust and non-exhaust traffic sources. Time-series show a recurrent decreasing trend during weekends. Polar-plot shows a large spreading of the sources.	High contribution to EC & significant to As suggests both exhaust and non-exhaust traffic sources. Time-series show a recurrent decreasing trend weekend. Polar-plot shows a spreading in the direction from the main settlements up-wind.				

Table 25: Fossil burning & traffic factor comparison

Heavy oil burning primary sulphate in Brod

The Heavy oil burning primary sulphate explains about 80% of SO_4^{2-} and a small portion of EC. That fingerprint, with very low K^+ and low OC is consistent with the "petrochemical and heavy oil combustion suburban" in the fingerprints database of SpecieEurope (JRC, 2017). The origin of that factor is probably to a large degree from the refinery situated south of the sampler but might also be from some heating facilities in Brod and the neighboring Croatian city of Slavonski Brod (Jeričević, 2019). It can also be seen from the polar plot that high concentrations of this factor often coincide with winds coming from the south where the refinery is. It is important to underline, however, that the refinery had a very low activity during the sampling period and the last two years. This factor has a significant contribution of 21% of the total PM 2.5.

Soil dust

The Soil dust factor is characterized by a recurrent significant contribution to As concentration. In this study between 30% to 60% of this element is usually explained by the soil dust factor. It is worth noting that the central part of the Balkan peninsula shows a complex geology and has some geological formations and ore deposits rich in As (Tarvainen T, 2013). In this context the mine tailings, infrastructure earthworks and road dust resuspension can be direct sources of particulate matters with a fair amount of As. Additionally, the metallurgical and electric-power plants that process such ore can be important sources of As, which are absorbed by the emitted particulate matter. Therefore, As is used in this study as an indicator of soil dust and soil resuspension.

It seems that this factor is often driven by some local and intermittent sources that can vary a lot depending on the sampling location and place. These puffs of soil dust factors are depicted in the time-series by some sudden peaks, and in the polar plots by some preferred wind direction with contrasted contributions.

	Sarajevo	Tuzla	Zenica	Banja Luka	Bijeljina	Brod
Factor: Soil dust	8%	13%	6%	23%	2%	6%
	Probably driven by two unusual but non-identified events in the vicinity and otherwise by the traffic resuspension.	Probably driven by one significant non-burning source in the vicinity of the sampling site.	Probably driven by the traffic resuspension.	Probably driven by several significant non-burning sources in the vicinity of the sampling site.		Probably driven by the nearby road.

Table 26: Soil dust factor comparison

Other factors

Industry factor in Zenica, Banja Luka and Tuzla

The Industry factor explains about 80% of Manganese, which used to be linked to the metal industry, at least when not together with a soil marker. The polar plot from Zenica shows a specific pattern with N-NW winds and the industrial steelwork area is situated 1,5 km upwind. For Banja Luka, the combination of the time series and the polar plot points to a general background enriched with a few concentrated events, with two single days explaining each 12% of that factor. However, it is not possible to point out a specific source. The portion of the industry metal factor for all PM 2.5 is only about 5% both in Banja Luka and Zenica. In Tuzla, none of the measured PM2.5 could be correlated with industry. This might sound surprising, since the Termoelektrana Power Plant southwest of the city is a large emitter. However, the sampler was situated quite far from the power plant (7 km), around 100 m above the power plant in altitude and most importantly, wind conditions were not favorable for transport of particles from the west-south-west during the campaign, with almost no wind recorded from that direction.

Cadmium-rich factor in Sarajevo:

One factor among the validated modelled solution explains about 90% of the Cadmium measured during the period. The portion of the Cadmium-rich factor among all PM 2.5 is rather high and reaches 12%. Both the time series and the polar plot indicate that the major sources should be linked to two events in late November and in mid-December, with one single day explaining 10% of that factor. Both isolated events seem closely related to the soil dust factor. A possible source may be some isolated fire of blended materials in the vicinity at that time or some more common sources such as fossil fuel combustion (Huremović, 2020), but the latter makes it difficult to explain the peaks.

Cadmium-rich factor in Zenica:

One factor among the validated modelled solution explains about 80% of the Cadmium measured during the period. The combination of the few peaks in the time series and the generally yellow polar plot allows to conclude for a general background enriched with a few concentrated events coming from two narrow places situated in the W-NW of the sampling place. It is not possible however to point out a specific source. The portion of the Cadmium factor among the whole PM 2.5 is only about 3% in Zenica.

Potassium-rich in Banja Luka:

The Potassium-rich factor explains about 60% of potassium whereas biomass burning explains only 20% of it. The combination of the time series and the polar plot indicates a quite continuous source in the background but that differs from the biomass burning factor. There is a significant correlation between the Potassium-rich factor and the Soil dust factor, probably related to the similar contribution time-series at least in November and December. It is worth considering merging these two factors as a single soil dust factor. However, the soil dust factor already represents 23% of modelled PM 2.5 and it may not be reasonable to add a Potassium-rich factor that represent 20% of the modelled PM 2.5. Additional information or measurements are needed to identify a specific sort of source for that amount of Potassium in Banja Luka.

Discussion regarding 2020-2021 campaign

The Secondary aerosols take a large part of the PM2.5 measured. Together the Secondary sulphate and Secondary nitrate aerosols represented between 20% to 35% of the measured PM2.5. These aerosols are the products of the natural atmospheric oxidation of the precursors that are emitted mostly by the anthropogenic activities (NH_4 by agriculture, NOx by all types of combustion, and SO_2 by coal combustion). Since this transformation takes between a few hours to a few days, Secondary aerosols could be considered to originate from areas and regions distant from the sampling place. However, the contribution of *local* Secondary aerosol emissions would also be possible during episodes of temperature inversions. These stable atmospheric conditions limit the air mass movement over the area and promote the accumulation of local emissions sources as well as their transformation *in-situ*.

The emissions by combustion of biomass like wood, pellet or shrub waste also represent a large part of the PM2.5. The portion of biomass burning group of sources seems to be larger in small towns, where it represents about 35% of the PM2.5, than in larger towns, where it represents between 19% to 25%. This might be related to a greater part of other local sources that reduces the relative portion of the Biomass burning. These can be heavier traffic load in the area of the sampler, more frequent use of coal combustion for residential heating in larger cities than in small cities, etc.

Due to a restricted number of measured species provided by the chemical analysis, the portion of PM2.5 emitted by the combustion of fossil fuel (coal, brown coal, diesel, gasoline) have been difficult to separate from some other groups of sources. In this study they are often blended in the results with other type of sources like Secondary aerosols and Biomass burning. It is possible however to say that the contribution of the combustion of coal and brown coal to the ambient PM2.5 is between 14% to about 25% and can in several cases be related to large district heating installations or industrial installations in neighbouring areas.

The portion of PM2.5 issued from vehicles have been isolated only for Tuzla and includes both exhaust and non-exhaust emissions. The contribution of traffic to the ambient PM2.5 is about 20% in these results.

The portion of PM2.5 related to Soil dust either naturally emitted, or resuspended by human activity, has always been identified. The soil contribution depended on the sampling location and varied between 2% to 13%. One particulate sampling location in Banja Luka has however a result largely over the range with a Soil dust contribution largely disturbed by a significant source of inorganic particles that haven’t been possible to explain after the 2020-2021 measurement campaign.

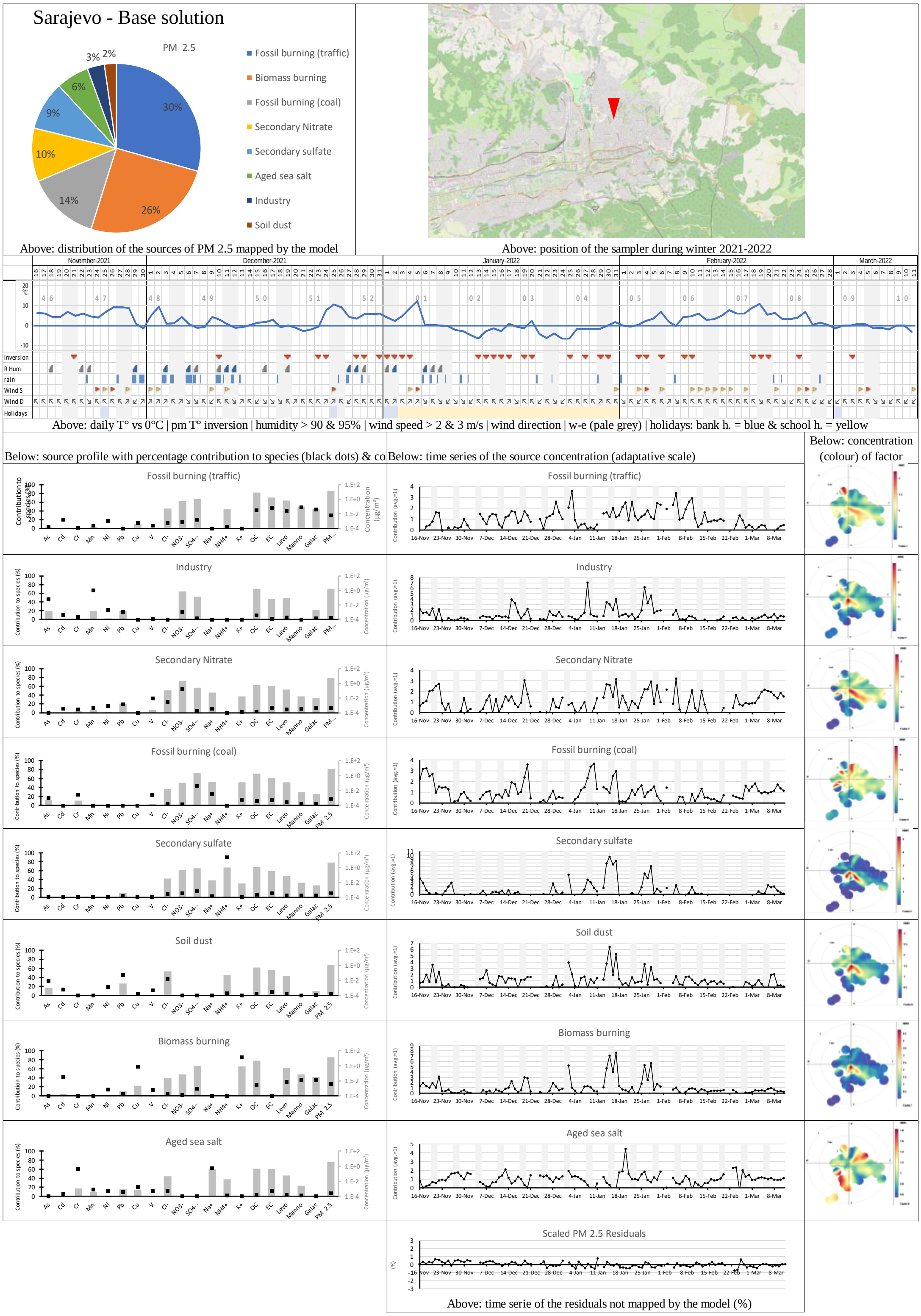
The total weight of all the measured species represents about 60% of the total mass of PM2.5 sampled. This ratio is common for the kind of analysis used. It means that some species that haven’t been measured but that used to have a significant part of the overall PM2.5 - like calcium, silicon, iron, aluminum - have been uniformly distributed in all apportioned sources. But since these species are more related to the Soil dust factor than the other factors, it is therefore likely that the Soil dust source has a larger share of the PM2.5 than calculated by the PMF. It probably can even double and consequently reduce the portions of the other sources. The results should be interpreted with this in mind.

Finally, with an aim to conclude with a satisfactory analytical measurement, it was agreed by partners to conduct a second sampling campaign during the winter of 2021-2022, with different technical arrangements than during the winter 2020-2021. These new arrangements aimed to get a wider range of measured species and thus to a more accurate result of the PMF model. This campaign has been conducted in Sarajevo and Banja Luka, where the receptor model had the most difficulty to resolve a source apportionment of high quality. Their very complex urban and topographical environments, especially in Sarajevo, require exceptionally accurate data for the PMF model to produce reliable results.

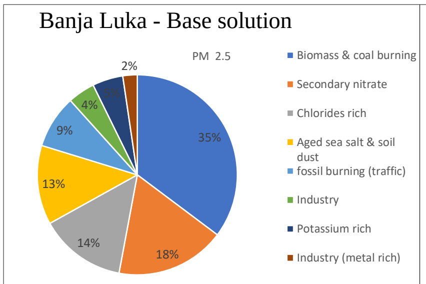
Identification of different sources – campaign 2021-2022

See figures below for Sarajevo and Banja Luka.

Sarajevo – base solution, winter 2021/2022

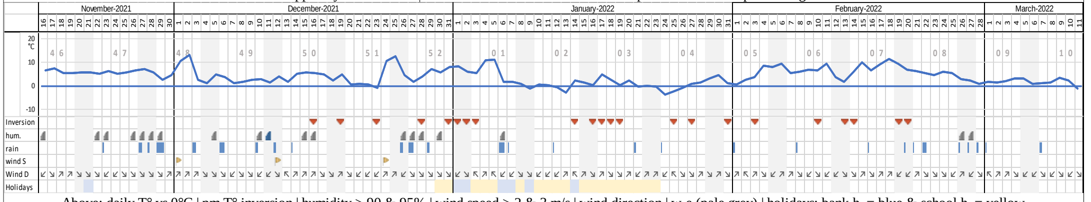


Banja Luka - base solution, winter 2021/2022



Above: distribution of the sources of PM 2.5 mapped by the model

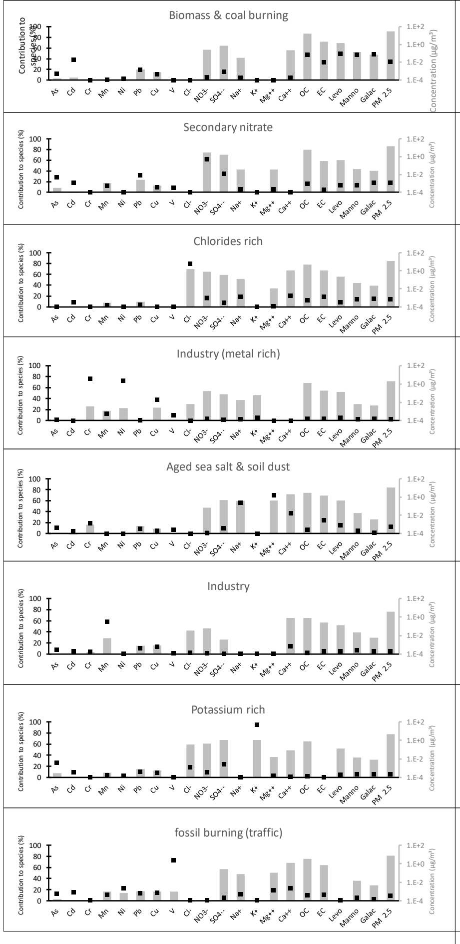
Above: position of the sampler during winter 2021-2022

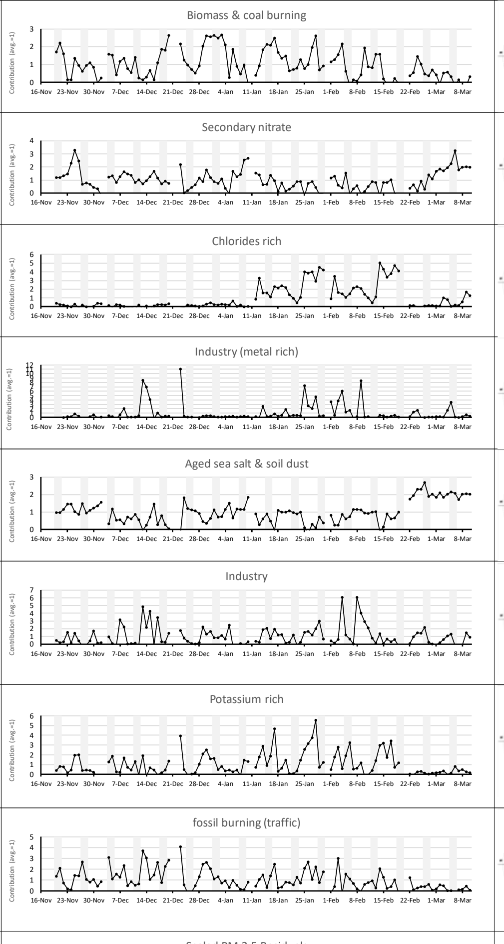


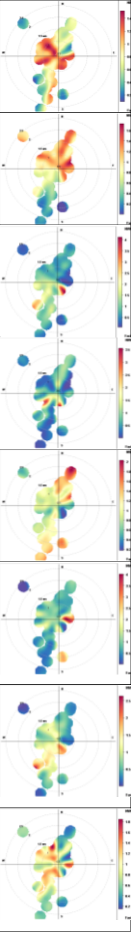
Above: daily T° vs 0°C | pm T° inversion | humidity > 90 & 95% | wind speed > 2 & 3 m/s | wind direction | w-e (pale grey) | holidays: bank h. = blue & school h. = yellow

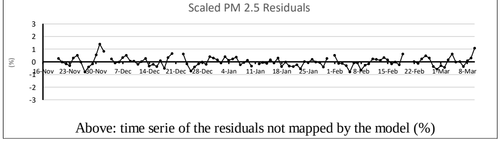
Below: source profile with percentage contribution to species (black dots) & co Below: time series of the source concentration (adaptative scale)

Below: concentration (colour) of factor









Above: time serie of the residuals not mapped by the model (%)

Fossil burning

The wider range of species measured in 2021-2022 does permit to discriminate two sources of fossil burning in Sarajevo. However only one source of fossil burning has been discriminated among others in Banja Luka.

The factor for fossil burning from traffic in Sarajevo is characterised by higher contribution to EC than to OC. The activity of that source provides additional information about the source with both the time series pointing out higher levels during weekdays than during weekends, and with the polar plot that suggesting two significant sources NW and SE of the sampler, which is in accordance with the position of the major roads in the vicinity.

The factor for fossil burning from coal in Sarajevo is characterised by a significant contribution to SO_4^{2-}. The temporal activity of that source shows a good agreement between its peaks and the three periods with negative temperatures during weeks 51, 2, 3, 9 and 10. The polar plot displays a yellow field in every direction with low wind speed that suggest that several sources are spread around the sampling point.

The factor for fossil burning from traffic in Banja Luka is characterised by a high Vanadium (V) contribution that is agreed to be an indication of Heavy oil and diesel burning (Pitiranggon, 2021). The temporal pattern however does not pinpoint a clear weekday activity. The polar plots show again that the emission sources of this factor are spread around the sampling site. Therefore, this factor is believed to be related to traffic with a fraction of it probably related to oil burning for heating purposes.

The amount of Fossil burning source in the total fine particles in Banja Luka is unexpectedly low (only 9%) whereas biomass burning is very high (35%) when comparing to the results of the 2020-2021 campaign (19%). It is therefore reasonable to believe that a part of the fossil burning source related to coal burning is actually blended in the biomass burning factor. The 35% slice in the diagram on the previous page has therefore been labelled “biomass and coal burning”.

The total amount of fossil burning modelled for Sarajevo is much higher for the winter 2021-22 than for the winter 2020-21. The major differences between the two winters were a much longer inversion period during the second winter that kept the pollution near ground level, and more normal anthropogenic activity during the second winter than earlier, when harsher restrictions related to the Covid-19 pandemic affected the community.

That type of trend is not possible to see for Banja Luka since the fossil burning factor was blended together with the secondary nitrate aerosols factor for the winter 2021-22. Therefore, it is not possible to say whereas the fossil burning source has increased or decreased between the two winters in Banja Luka.

Place:	Sarajevo		Banja Luka	
Winter:	2020-2021	2021-2022	2020-2021	2021-2022
Factors:	Fossil burning & traffic 23% [13,5 µg/m³]	Fossil burning (traffic) 30% [15,3 µg/m³] Fossil burning (coal) 14% [7,2 µg/m³]	Fossil burning & secondary nitrate aerosols 24% [16,8 µg/m³]	Fossil burning (traffic) 9% [7,7 µg/m³]
Reason according to Finger prints: – Times series: – Polar-plots:		<u>Fossil burning (traffic):</u> High contribution to EC and significant to metals & anhydrosugars suggests both exhaust and non-exhaust traffic sources. Time-series show a recurrent decreasing trend during weekends. Polar-plot pinpoint directions in agreement with the direction towards the major roads. <u>Fossil burning (coal):</u> High contribution to SO_4^{2-} together with significant contribution to EC suggests a coal burning source. The time-series displays higher peaks at the same time the temperatures where below zero °C. Polar-plot shows a general spreading of the sources all around.		High contribution to the V that is agreed to be the signature of heavy oil and diesel burning. Times-series shows a recurrent decreasing trend during weekends (pointing to a traffic source) but also some peaks at the same time as temperatures below zero °C (pointing towards a heating source). Polar-plot shows a general spreading of the sources all around.

Table 27: Fossil burning factor overall comparison

Biomass burning

It is important to note that for this second analysis an identification method of temperature inversions has been tested. The RHMZ has conducted a daily analysis from the 10th of December of SkewT diagrams (modelled temperatures in different altitudes according to meteorological models). The FHMZ used a simpler measurement approach to identify an inversion, comparing the ground level temperatures at different altitudes, in the city and up the hills. These methodologies helped to identify possible inversion episodes at the afternoon of each day which is represented on the graphs by a red triangle pointing down. It is remarkable how the level of PM 2.5 related to some local sources as Biomass burning seems to be increased when there is a succession of days with afternoon temperature inversion.
The biomass burning factor was always isolated, both for the winter 2020-21 and 2021-22. This was possible thanks to the combination of the tracers OC and K^+ together with anhydro-sugar like levoglucosan, manosan and galactosan. The difference for the winter 2021-22, is that the data set was sufficiently wide and long to lead to a stable and reliable solution of the model without any constraints on the manosan to pin-point the Biomass factor.
The factor Biomass burning in Sarajevo is characterised by the combination of OC, K^+ and anhydrosugars. The source is discontinuous and coincidental with cold events which is suggesting heating appliances sources. The polar plot displays a point pattern which mean that the sources were active in the area during periods with calm winds.

The factor Biomass burning in Banja Luka is characterised by the same type of fingerprint. However, another factor named “Potassium-rich” captured that particular K^+ species. That source is quite constant for Banja Luka and much higher (35%) in the last winter 2021-22 than previously estimated during the winter 2020-21 (19%). This difference together with the absence of K^+ in the signal suggests that the biomass factor in 2021-22 may be blended with another heating-purpose factor as the Fossil burning coal which is otherwise not present.

Place:	Sarajevo		Banja Luka	
Winter:	2020-2021 □ 2021-2022		2020-2021 □ 2021-2022	
Factors:	Biomass burning 26% [16,1 µg/m³]	Biomass burning 27% [13,3 µg/m³]	Biomass burning 19% [13,3 µg/m³]	Biomass burning & coal burning 35% [31,6 µg/m³]

Place:	Sarajevo		Banja Luka	
Winter:	2020-2021 ⍰ 2021-2022		2020-2021 ⍰ 2021-2022	
Reason according to Finger prints: – Times series: – Polar-plots:		Significant contribution to OC and to the K^+ together with significant contribution to anhydrosugars suggests biomass burning sources. The time-series displays higher peaks at the same time temperatures were below zero °C which suggest a heating source. Polar-plot with a central point major contribution suggest a local source that are active mostly under wind-still days.		High contribution to the OC together with significant contribution to anhydrosugers suggest biomass burning sources. The K^+ is missing in this fingerprint but Banja Luka is remarkable by another high potassium rich factor that capture the K^+ to the other's detriment. Times-series even if quite continuous (between 0 to 3%), shows an agreement of the peaks with the time when temperatures were below zero °C (pointing towards a heating source). Polar-plot shows a general spreading of the sources all around which agrees with the hypothesis of domestic heating.

Table 28:Biomass burning factor overall comparison

Secondary aerosols

The secondary aerosols have been identified using the same species as tracers when it was possible (The $NH4^+$ haven't been detected enough in Banja Luka for being used).
The factor for Secondary nitrate aerosols is characterised by a high contribution of that factor to NO_3^- and a decoupling with the inversion periods. It is visible in both places where peaks occur at the beginning and the end of the measurement period and generally when there were no inversion situations. Since secondary nitrates are a product of a chemical atmospheric transformation, its sources are considered to be situated in a significant distance of the sampling site. This is visible through the polar plots that display a background, mostly coming from the Northern sectors where most of the anthropogenic sources are located, both regarding Sarajevo (Tuzla, Brcko and Croatia) and Banja Luka (Croatia).

The factor Secondary sulphate aerosol is characterised by a high contribution of both SO_4^{2-} and NH_4^+. The literature and the results from the previous campaign have shown that this factor is the product of a chemical atmospheric transformation either through a long-range transport or through an accumulation from local coal burning sources under high humidity and temperature inversions. That factor was possible to discriminate only in Sarajevo and its time series show quite a good relationship of its peaks with the inversion episodes. The more or less local sources seem to be predominant here.

The total amount of Secondary Organic Aerosols (SOA) is similar between the two places for the winter 2020-21 as well as it is for the winter 2021-22. But there is a significant decrease between the two winters regarding the secondary nitrates. However, that decrease can be explained since secondary nitrate aerosols were believed to be blended with a fossil burning factor for the winter 2020-21 and that have been better discriminated after the second measurement campaign during the winter 2021-22.

Place:	Sarajevo		Banja Luka	
Winter:	2020-2021 ⍰ 2021-2022		2020-2021 ⍰ 2021-2022	
Factors:	Secondary nitrate aerosols 14% (shared with fossil burning sources) [8,1 µg/m³]	Secondary nitrate aerosol 10 % [5,3 µg/m³]	Secondary nitrate aerosols 24% (shared with fossil burning sources) [16,8 µg/m³]	Secondary nitrate aerosol 18% [15,8 µg/m³]
	Secondary sulphate aerosols 16% [9,7 µg/m³]	Secondary sulphate aerosols 9 % [4,8 µg/m³]	Secondary sulphate aerosols 9% [6,7 µg/m³]	– – % [– µg/m³]

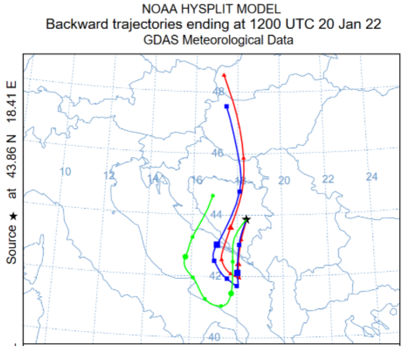
Place:	Sarajevo		Banja Luka	
Winter:	2020-2021 ⍰ 2021-2022		2020-2021 ⍰ 2021-2022	
Reason according to Finger prints: – Times series: – Polar-plots:		Significant contribution of the factors to NO_3^- regarding the Secondary nitrate otherwise to the couple NH_4^+ and SO_3^{2-} regarding the Secondary sulphate. The time-series displays stronger contribution of the Secondary nitrate when there wasn't significant temperature inversion. The time-series of the secondary sulphate display at the contrary stronger peak concurrently to the inversion episodes. Polar-plots related to Secondary nitrate displays a general background from the north sector compliant with a long-range dispersion. A the contrary the polar plot related to the Secondary sulphate display local source with low wind speed compliant to the atmospheric situation of an inversion.		Significant contribution of the factors to NO_3^- regarding the Secondary nitrate. The secondary Sulphate aerosols haven't been identified probably because the NH_4^+ haven't often enough been detected by the chemical analysis. The time-series displays stronger contribution of the Secondary nitrate when there wasn't significant temperature inversion (i.e. at the beginning and the end of the measurement period). Polar-plots related to Secondary nitrate displays a general background from the north sector compliant with a long-range dispersion.

Table 29: Secondary aerosols factor overall comparison

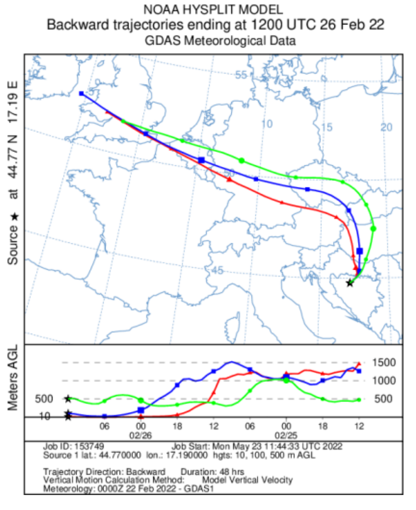
Aged sea salt

The aged sea salt factor wasn't possible to discriminate for the winter 2020-21 since neither of the Na$^+$, Cl$^-$ nor the Mg^{2+} were measured at a high enough resolution. During the winter of 2021-22, and after changes in the analytic method, these ions were sufficiently represented in the measurements to be able to discriminate that factor.

The aged sea salt factor is characterised by a significant contribution to Na$^+$ as well as Mg$^+$. Under the timeframe from its production at sea and its detection over land, most of the Cl$^-$ tends to combine with other species under atmospheric chemical reactions and is no longer significant in the measurement results (Xu, 2021).



To ensure the correct identification of the Aged sea salt factor, some backward trajectories have been calculated using the Hysplit Model from NOAA. Backtracking from dates of higher peaks for this factor shows that high peaks of Aged sea salt are often related to sequences of strong winds / storms happening over either the North Atlantic or the Mediterranean Sea.



The backtracking maps beside show a point every 6 hours of the position of the air-mass with the backward trajectories calculated from a position (black star) at 12:00 on Jan 20 in Sarajevo (upper picture) and in Banja Luka on Feb 26 (lower picture). Three different air masses are considered: at ground level (red), at 10 meters height (blue), and at 500 meters height (green).

It shows that the higher peak of Aged sea salt at Sarajevo on Jan 20 was related to an air-mass that has been transiting a whole day over the Adriatic-sea in stormy conditions (storm Elpis) only 10 hours before arriving over Sarajevo.

It shows otherwise that the peak of Aged sea salt at Banja Luka on Feb 26 was related to an air-mass that have travelled quite fast over northern Europe from the north Atlantic with a moderate vertical movement of 1000-1500m.The higher peak was then related to an air-mass that has been over the Atlantic surface, in stormy conditions (storm Eunice), 2 days before arriving over Banja Luka.

Other backtracking calculations have also been conducted as a negative control and have shown that days of lower contribution of Aged sea salt coincided with air-mass coming from continental areas.

Both the amount and the trends of this Aged sea salt factor are different between the two places. In Sarajevo, this factor represents 6% of the modelled factors which is high comparing to the yearly average in the region but is still possible during winter since stormy conditions at sea occur more often in winter than over the year in average. In addition, the time-series display a few high peaks which are related to high wind speed according to the polar plots. This is compliant with long-range transport of fine particles.
Concerning Banja Luka, this factor represents 13% of the modelled factors which is extremely high for the region. In addition, the time-series display a relatively continuous source even though the polar-plots suggest that high concentrations of this factor are related to high wind speed. Regarding the ending plateau of the time-series from Feb 22, backward trajectories show that that the particular period was characterised by air-mass transport from the mainland with very low vertical exchanges. These backward trajectories together with the high occurrence in the signal of both Ca^{2+} and Mg^{2+} lead to the conclusion that the Aged sea salt factor in Banja Luka is actually blended with a Soil dust factor that haven't otherwise been identified (see Soil dust below).

Place:	Sarajevo		Banja Luka	
Winter:	2020-2021 → 2021-2022		2020-2021 → 2021-2022	
Factors:		Aged sea salt 6% [3,3 µg/m³]		Aged sea salt (shared with soil dust sources) 13% [11,4 µg/m³]

Place:	Sarajevo		Banja Luka	
Winter:	2020-2021	2021-2022	2020-2021	2021-2022
Reason according to Fingerprints: – Times series: – Polar-plots:		Significant contribution to Na^+ alone. The time-series display higher peaks at the same time the wind-flux was coming from the Adriatic Sea in stormy conditions and with low vertical mixing. Polar-plot higher concentrations at the north and south edges suggest that the sources are related to strong winds.		Significant contribution to Na^+. (Significant contribution as well to Ca^{2+} and Mg^{2+} suggest a participation of soil dust in the factor) The time-series display higher peaks at the same time the wind-flux was coming from stormy conditions in the North Atlantic with low vertical mixing. (Significant plateau at the end of the period together with northern wind flux from the mainland with very low vertical mixing suggest a participation of soil dust) Polar-plot higher concentrations at the north and south edges suggest that the source related to strong winds even if a significant background remains.

Table 30: Aged sea salt factor overall comparison

Soil dust

The Soil dust factor was isolated for both winter periods. This was possible thanks to the tracer As that should be a good crustal-source marker in the Balkan region.
The Soil dust factor in Sarajevo is characterised by As and since the source is coincidental with dry periods this suggests a road-dust suspension source. The polar plot displays a point but slightly diffuse pattern which means that the source is local and downplayed by strong winds. The total amount is comparable between the two years considering the seasonal variability.
Surprisingly, given that the Soil dust factor was rather significant in Banja Luka during the winter of 2020-21, it couldn’t be isolated in Banja Luka during the winter of 2021-22. However, the Soil dust is suspected to be blended into the Aged sea salt which was largely (see above) present in Banja Luka during the second winter.

Place:	Sarajevo		Banja Luka	
Winter:	2020-2021 ▯ 2021-2022		2020-2021 ▯ 2021-2022	
Factors:	Soil dust 8% [4,5 µg/m³]	Soil dust 2% [1,2 µg/m³]	Soil dust 19% [14,3 µg/m³]	– not discriminated –% [– µg/m³]
Reason according to Fingerprints: – Times series: – Polar-plots:		Significant contribution to As. The time-series displays higher peaks during dry periods. The inversion periods have a large influence on the major peaks. Polar-plots with a central but diffuse maximum suggest a local source that are active mostly under wind-still days. All this together might suggest a road-dust resuspension source.		

Table 31: Soil dust factor overall comparison

Chlorides-rich factor in Banja Luka

The Chlorides-rich factor isolated only in Banja Luka explains about 70% of the Cl^-. The time-series of that factor is quite unusual since the emissions responsible of that factor seems to be active during only six weeks (12/01 – 19/02) otherwise not at all. The polar plot points out at least two major directions from the sampling place. The period of activity of that factor is coincidental with dry weather with temperatures slightly below zero. This combination leads to the conclusion that this factor may be highly influenced by the dried road salt remaining on the asphalt (Almeida, 2020) and resuspended with the road dust aerosols.

In the meantime, this factor represents 14% of all the 8 modelled factors even if it was active only 1/3 of the time. In addition, it is remarkable that both peaks of the times series and the major directions of the polar plots complement the respective dips of the Aged Sea salt factor very well. Since the background of the Aged sea salt factor is believed to be the Soil dust source, it makes sense to conclude the same for the Chlorides-rich factor. A blended source would help to explain its high amount of this particular factor for the whole period.

Potassium-rich in Banja Luka

Similar to the winter of 2020-21, Banja Luka is characterised by a Potassium-rich factor that explains most of the K^+, 95% for the winter of 2021-22. As a reminder, the Potassium-rich factor for the winter of 2020-21 captured 60% of all the measured K^+, with 40% represented by the Biomass burning factor.
The share of that Potassium-rich factor in the total PM2.5 modelled is now lower, 6% instead of the previous 20%. The times series display peaks mostly during weekdays which suggests business-related activity. But the polar plots point out at least two directions in a general background which suggests a scattered number of sources. The time-series seem to be disconnected from both periods of sub-zero temperatures as well as the time series of the Biomass burning factor, therefore the Biomass burning factor shouldn't be suspected to be blended here. The top soils in the region of Banja Luka are generally acid and known to be low on K^+ (Markovic, 2015) therefore the Soil dust factor shouldn't be suspected here either. It is not possible to point out a specific source of fine particle loaded with Potassium in Banja Luka without obtaining additional information or further measurements.

Industry

The Industry factor explains between 57 to 80% of the Manganese, which usually is linked to metal industry, at least when it is not together with a soil marker. The share of that factor is similar between the two cities, about 3 to 4%.
Banja Luka have an additional factor characterised by a significant contribution to both Cr and Ni as well as Cu. The combination of Cr and Ni is known to be a signal of coal power plants but the absence of all other tracers than metals and notably the absence of SO_4^{2-} leads to reject a coal burning source here. In addition, the fact that the polar plots point out isolated directions supports the conclusion that this additional factor is also an Industry factor which is also Cr and Ni-rich.

Place:	Sarajevo		Banja Luka	
Winter:	2020-2021	2021-2022	2020-2021	2021-2022
Factors:	– not discriminated –% [– µg/m³]	Industry 3% [1,7 µg/m³]	Industry 5% [3,7 µg/m³]	Industry + Industry (Cr-Ni rich) 4% + 2% [3,9 + 2,1 µg/m³]
Reason according to Fingerprints: – Times series: – Polar-plots:		Significant contribution to Mn. The time series indicate highly variable peaks of concentration during business days, unrelated to weather variations. Polar-plots with a central but diffuse maximum suggest a local source that are active mostly under wind-still days. This may point towards a road-dust resuspension source.		For the Industry factor: significant contribution to Mn. The time series indicate significant peaks of concentration during business days, unrelated to weather variations unless the absence of rain. The polar plots point out a major source situated east of the sampling place. For the Industry (metal) factor: significant contribution to both Cr and Ni as well as Cu but not to V. The time series indicate highly variable peaks of concentration during business days, unrelated to weather variation unless absence of rain. The polar plots point out two major sources in the south and the south-west.

Table 32: Industry factor overall comparison

Combined discussion on the 2020-21 and 2021-22 campaigns

This study is exclusively focused on PM2.5, which is of particular importance because it is small enough to be transported deep into the human lungs and its most vulnerable parts, and cause lung diseases. The conclusions of this report focused on PM2.5 do not allow to anticipate effects or concentrations of other air pollutants.

The results overall show high levels of PM2.5 – on daily average 62 µg/m3. Generally, the study suggests that around 25% of PM2.5 is emitted from wood and pellet burning, which are used for heating purposes. Around 20% of PM2.5 seems to come from fossil fuel combustion including coal for heating purposes and vehicle engines. The portion of the background or long-range transport aerosols seems to represent more than 25% of the total PM2.5.

The study also shows the usefulness of results produced by the Positive Matrix Factorisation model when using the USEPA PMF 5.0 software, with the analytical results of PM2.5 daily collected on filters. The results were then further investigated using local meteorological data, including time-series and polar plots, which in some cases increased the understanding of where and when different pollution sources originate from.

The second round of measurement and modelling during winter 2021-22 confirmed the major contributors to PM2.5 in Sarajevo. Thus, it also indirectly strengthened the results for the other 5 cities in the winter of 2020-21, confirming the methodology used. In Banja Luka, however, the results showed some differences, namely that the amount of PM2.5 in Banja Luka was about 86 µg/m3 on daily average during the winter of 2021-22, higher than what was identified in winter 2020-21. The PM2.5 identified in Banja Luka was from local sources and had a complex chemical composition. This needs to be further studied.

The first sampling campaign conducted for this study occurred during the winter of 2020-2021 in 6 cities (Sarajevo, Zenica, Tuzla, Banja Luka, Bijeljina, and Brod) of Bosnia and Hercegovina and the second during the winter of 2021-2022, but in Sarajevo and Banja Luka only. That latter was undertaken using a more accurate methodology allowing measurement of more pollutants with regards to the more complex environment in these two cities. Depending on the sampling location, between 5 to 6 different groups of PM2.5 sources have been isolated in the first winter. During the second winter 8 groups of PM2.5 sources have been isolated, in both cities.

The time-series have been analysed with consideration to the surrounding geography and human activities, together with weather data provided by FHMZ and RHMZ. These analyses gave some useful insights for the identification of major groups of sources of PM2.5. The general apportionment of sources is similar to those available from other studies of the region (see REFERENCES page 125) and is shortly described below.

Secondary nitrate aerosols have been confirmed under both winter campaigns to be a significant part of the PM2.5 measured. Secondary nitrates are often between 10 to 20% of the total PM2.5. Its spatial and temporal variation patterns strongly suggest a regional contribution coming from the rest of Europe from the north.

Secondary sulphate aerosols showed a more complex situation. Secondary sulphates represented between 9 to 16% of the measured fine particles along the two consecutive winters. Modelling during both winters led to suspect that the Secondary sulphate aerosols might be mostly the result of local emissions directly from coal burning, and of a local, chemical transformation to nitrates or sulphates in wet temperature inversion conditions (Chunrong Chen, 2021).

Biomass burning has also been confirmed to be a significant source of PM2.5. In Sarajevo, the portion of this factor among the measured fine particles have been consistent during the two campaigns, with 27% and 26% respectively. The Biomass burning identification and apportionment in Banja Luka was not as coherent in the two campaigns, probably because of a specific complexity in the composition of the fine particles in this particular place. The contribution of Biomass burning during the first winter also varied between the sampled cities and was largest in the small cities where it could contribute up to 35% of the total PM2.5. Worth mentioning is also that the second measurement campaign and modelling have brought much more clarity of the different sources that affect the air quality, directly in Sarajevo, but indirectly also in the other 5 cities, since it has validated the methodology used in the study.

After the upgrade in the sampling and measurement methodology it was possible to better discriminate the fine particles emitted from Traffic. The results gave a proportion of 30% in Sarajevo and 9% in Banja Luka for the 2021-22 winter. The difference sounds big but when looking at the concentrations (15,3 vs. 7,7 µg/m³), the quantity of traffic-related fine particles in Sarajevo is only doubled compared to Banja Luka, which is in accordance with the traffic's order of magnitude (see MATCH dispersion modelling section for more details).

The Coal fossil burning was discriminated from the Traffic fossil burning in Sarajevo during the winter of 2021-22 and is estimated to be 14% of the total PM2.5. It was unfortunately not possible to isolate the Coal fossil burning alone in Banja Luka.

The Soil dust factor was discriminated well in Sarajevo also during the second winter, with a low contribution coherent with most other cities (ranging between 2-8%) during the first winter. The contribution in Tuzla, however, was estimated to 13% in the first winter. Banja Luka is an outlier with respect to Soil dust – the first winter gave a surprisingly high value of 23%, whereas the model wasn't able to discriminate this factor at all during the second winter. This needs to be further investigated.

The Industry factor plays a minor role in the total PM2.5 amount in Sarajevo. It wasn't identified during the 2020-21 campaign but was estimated to 3% the year after. In Banja Luka, that factor was first estimated to 5% and have been found again with a consistent amount of 6% but separated in two different factors. These finding are consistent with the 3% amount identified at Zenica as well.

The upgraded methodology used for the 2021-22 winter campaign allowed to identify the Aged sea salt factor which is a long-range pollutant that can be relevant particularly during the winter period. It was estimated to 6% in Sarajevo which is high but still possible during winter and similar to the estimation taken from other studies.

It is not surprising that combustion for Heating purpose is the main source of fine particles in Bosnia and Hercegovina. This study shows that wood burning is responsible for a significant part of the total PM2.5, between 19% and 36%, with a higher part in smaller towns. The burning of Coal, whereas for heating purpose or for power generation stands for 14% of these fine particles in Sarajevo. The portion of coal related particles is probably similar in the other cities, but the exact amount wasn't possible to assess in this study. The combustion of coal is probably also responsible for a significant part of the Secondary sulphate aerosols, especially during temperature inversion periods, and represent between 5% and 21% of the total PM2.5.
As expected, the combustion for transport is related to the amount of traffic. It can rise up to 30% in Sarajevo which is probably the busiest town among the six where the fine particles were sampled.

The long-range pollutants like Nitrate secondary aerosols are significant as well in the amount of PM2.5 but are also out of the scope of any local abatement strategy.
The natural sources can be meaningful with an amount of up to 10%, e.g. for Aged sea salt, mainly in wintertime, and for Soil dust, mainly during dry periods.

PART 2: ONE-YEAR DISPERSION MODELLING AND SOURCE APPORTIONMENT USING THE MATCH MODEL OVER SARAJEVO AND BANJA LUKA

Urban areas in Bosnia and Herzegovina (BiH) suffer from poor air quality and air quality standards defined for protecting human health are often exceeded. For example, the average level of PM2.5 measured in 2019 was twice the EU limit value (World Bank, 2019). According to the World Health Organization (WHO) figures from 2022, BiH is 6th among 50 European countries when it comes to the death rate attributable to air pollution. Sarajevo and Banja Luka are two cities in BiH that carry a large portion of the health burden caused by air pollution.

Sarajevo is set in a valley surrounded by complex terrain with steep mountains. Banja Luka is also surrounded by mountains but to a lesser extent. Valleys are naturally protected from wind, making valley air stagnant. Furthermore, at night when the air cools down and becomes heavy, it sinks into the valley floor from surrounding hill tops and thus promotes the development of inversions (Largeron and Staquet, 2016). These stable weather conditions are known to restrict air mass movement and greatly limit the ventilation of the air, thus having very negative effect on the air pollution load (Olofson et al., 2009) (Grundström et al., 2015). That atmospheric complexity necessitates the use of a unique dispersion model with several altitudinal layers of wind fields. In this study a 3D Eulerian model was used to be able to take into account the mass exchange between air mass flows.

Emissions come from many different sources such as industry, traffic, coal-fired power stations, and district and domestic heating. It has previously been shown that sectors such as residential combustion, power plants, industry and waste have high particle emissions in BiH (World Bank, 2019).

Emissions inventory work is a long-term task that often takes years to refine. Methodologies of data collection, managing yearly updates, maintaining relevant competencies, and keeping up to date with international standards requires long-term planning and effort. Thus, a common way to ensure that an emission inventory is good enough to depict the reality on site is to use it in a dispersion model. The air quality map and time variation are then compared against hourly air pollutant measurements from official and calibrated monitoring stations. Air quality experts should then be able to pinpoint the remaining tasks to undertake in the continuing effort toward a reliable and up to date emission inventory.

All three components – air quality monitoring, pollutant emission inventory and air quality modelling – are vital for formulating efficient reduction strategies through air quality plans and eventually emissions regulations. But the identification of sources and the estimation of their contributions are of an overarching importance for building the capacity to formulate precise and well targeted mitigation strategies for reducing air pollution levels (European Commission, 2020).

Background

The main aim of the study was to simulate concentrations of air pollutants NO_2, SO_2, PM_{10} and $PM_{2.5}$ in Sarajevo and Banja Luka using new local emission data sets from three emission sectors (district heating sources, small scale residential heating sources and traffic). Local emissions were further combined with regional emission inventories from Copernicus Atmosphere Monitoring Service (CAMS, 2019) that provided gridded distributions of European and global anthropogenic emissions, as well as global natural emissions.

The second aim was to apportion the source contributions to the general air pollution levels from the main contributors when using the high-resolution emission data (the new local emission inventories). Source apportionment links the emission source to the concentration levels by revealing the location and magnitude of air pollution levels coupled to a specific source sector. This information is important for air quality managers to formulate targeted emission reductions, which can be included in an action plan.

In a broader perspective, the dispersion modelling result can be further applied in scenario analysis to determine and evaluate the effects of certain emission reducing action plans.

Methods

Atmospheric dispersion modelling (hereafter: dispersion modelling) is a numerical simulation of how pollutants travel with the wind from their sources. Depending on the model type and its formulation, dispersion modelling can include the effects not only of the weather, but also topography, atmospheric chemical transformation and some of the effects of turbulence.
Setting the dispersion model as it was done in this study involves several steps including:

1. Preparation of emission input data. This may include different source types (point, line, area, grid and volume sources) which are usually divided into different emission sectors, such as traffic, mobile machinery, industries, domestic heating etc.
2. Meteorological data. Depending on the area of study and the type of dispersion model employed, this may be in the form of observation data at a single site, or

gridded data from a Numerical Weather-Prediction (NWP) model. Often, but not always, the dispersion model uses timeseries meteorology data to model the concentration of pollutants for a selected period of time, such as one year. In this study, hourly data from an NWP model has been used.

3. Preparing so-called physiographic data, which is topography and land-use date for the area of study.
4. Running the dispersion model. Most models calculate the concentration of pollutants on a cartesian grid for a series of consecutive timesteps.
5. Postprocessing of the results. This often includes calculating relevant statistical measures (such as yearly averages and percentiles), and extracting timeseries data at selected points in order to compare the results with monitoring data.
6. The above steps may be re-run with adjusted settings or updated input data if the model results do not agree well enough with the monitoring data.
7. For apportionment studies (for the relevant emission categories), the model setup may be used to re-run the model while scaling down a selected emission category, and then post-process this result to obtain a map of the contribution of the sector to the total concentration.

Local emissions data consolidation

One component of the IMPAQ project was to improve emissions inventories in three cities per entity – the same six cities that were examined in the PMF source apportionment study. Whereas several cities in the Federation have produced fairly harmonized emission inventories, the cities in the Republika Srpska lack emissions data for several sectors.

In Sarajevo there is sufficient traffic data available to undertake an emission data calculation from traffic counting, while for the other cities traffic data for individual roads is lacking. Only a few of the biggest industries report their emissions to the European Pollutant Release and Transfer Register (E-PRTR).
All significant industrial installations in both entities must renew their environmental permits every five years. Unfortunately, the submission for environmental permits does not include any assessment of emissions, or fuel consumption. All requests for further informal information have failed in the RS and have been conducted in the Federation under previous assessments with the formal support of the ministry of environment.

It had been decided in early 2021 to implement an alternative method of emission inventorying. The idea was instead of conducting a bottom-up inventorying to undertake a top-down emission inventory regarding the two meaningful sources of air pollutant in wintertime, the energy use for heating purpose, and the fuel burn by the traffic.

Regarding the **heating emissions** from individual households or apartments, an original methodology was set up in order to get a geographic distribution of the emissions. The detailed methodology is further described here in after but is summarized as below.
The main inputs are:

- Land use polygons from the UrbanAtlas 2012 dataset [REF?]. This dataset is surface covering and contains land use classes for all larger municipalities in most of Europe. The dataset includes population data in each polygon,
- Fuel consumption from local surveys and statistics (Survey on household energy consumption in BiH 2015),
- Bosnia and Hercegovina building typology report (Arnautović et al., 2016).

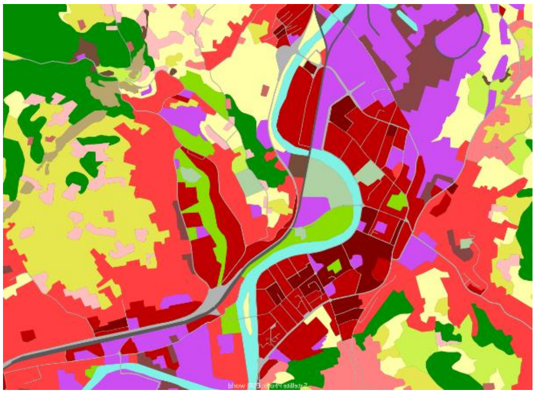



Figure 11: Urbans atlas Zenica & energy use per polygon after harmonization in Banja Luka

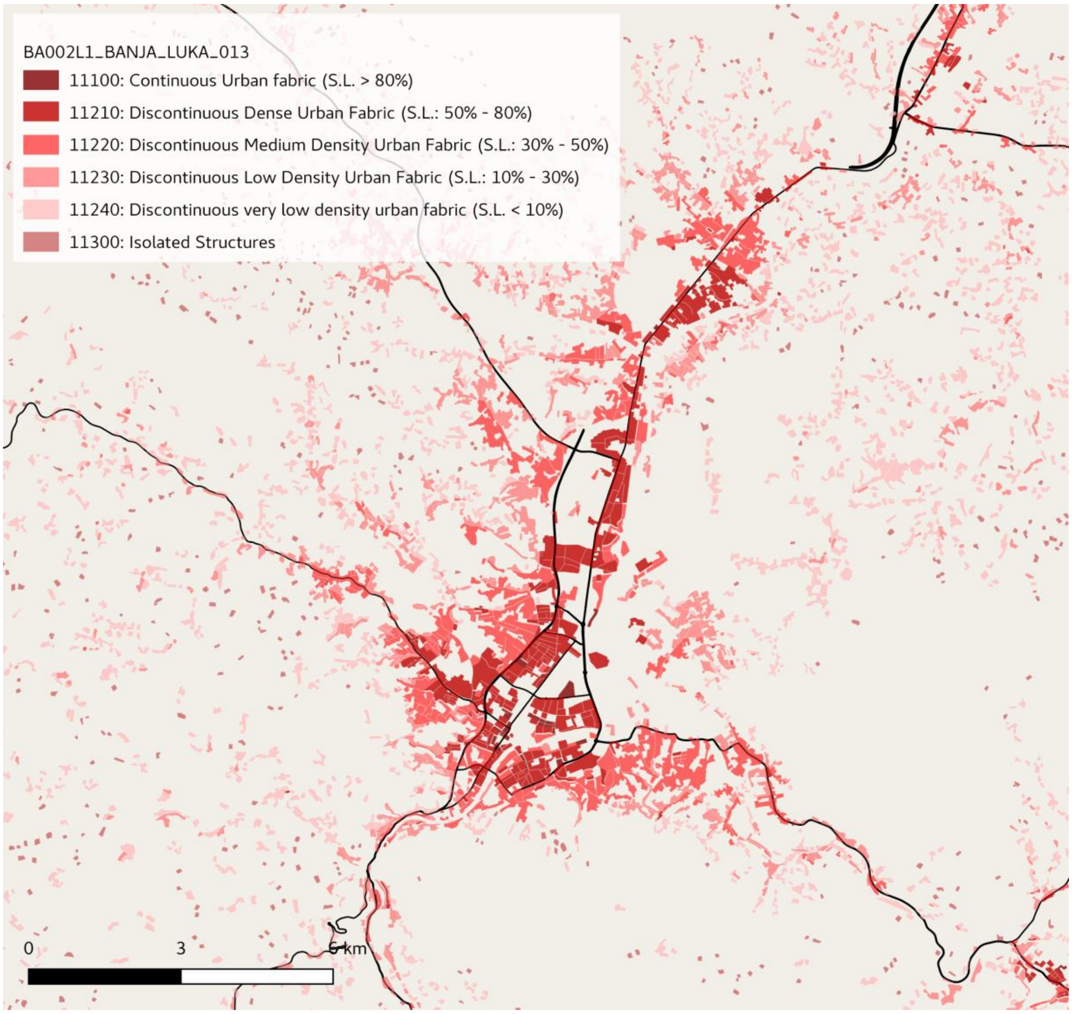


Figure 12: Banja Luka Urban Atlas 2012 land use categories that are assumed to contribute to emissions from residential heating. Major roads are shown for orientation

A blend of building types was determined for each class of urbanization. To do so the typology of residential buildings in Bosnia and Herzegovina was used (Arnautović et al., 2016). The building composition and heating energy need per type of housing were then distributed in all the calculated areas (Sarajevo, Zenica, Tuzla, and Banja Luka).

When combining these data sets with the type of fuel used either by individual households or so-called condominium it was possible to distribute the energy use in all the polygons containing housing. Further, in each polygon the energy consumption per fuel type could be estimated, which facilitated calculating the associated emissions.

The total results of energy consumption were then compared to the available statistics of energy use according to the building typology report.

All the basic data, processed data, methodology and GIS-files have been published in the HIVE data sharing portal to be freely available to all the stakeholders.

Regarding traffic and its geographic distribution, a country-wide approach was chosen. Two traffic reports from 2016 for the Federation (JP Ceste Fedreracije BiH, 2017) and for the Republika Srpska (Putevi Republike Srpske, 2020) were used. These reports rely on a network of permanent and temporarily automatic traffic counting and include at the end of the reports traffic modelling that was not possible to receive in a GIS format.

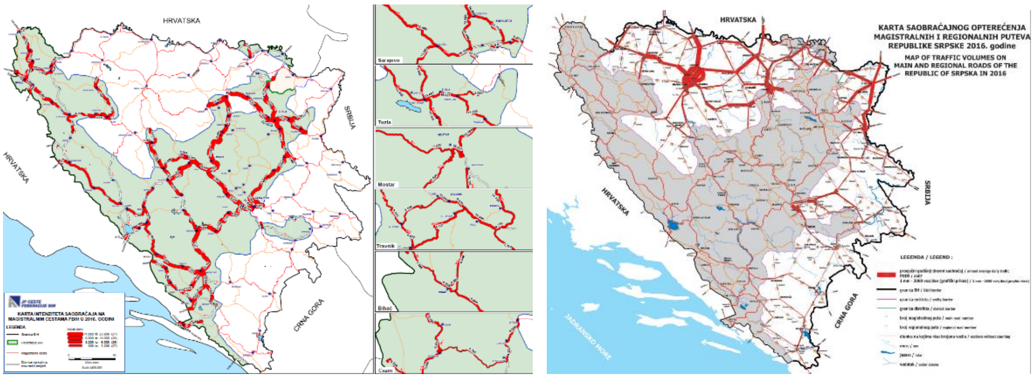


Figure 13: Traffic counting and modelling of the major roads in BiH for 2016.

Traffic counting from these two reports cover part of the major roads of both entities (Magistralna cesta). Those are only covering a fraction of the roads in BiH. That data was not available in an electronic format and SMHI had to manually implement it in a GIS database first.

It is important to note that both these reports do not include traffic flows from the segments of motorway. The requests for information have been submitted to the motorway operator in different manners but were unsuccessful.

The total traffic in the BiH has thus been estimated using the registered fleet of vehicles for the year of interest available from the Road and Transport Authorities of both entities. The yearly mileage was estimated against the Croatian statistic data for 2016 since the yearly mileage was not available for BiH. The yearly mileage depends on the class and age of vehicles.
This way of estimating the yearly mileage does not include the mileage of foreign vehicles on the Bosnian roads. That foreign traffic is estimated up to 3% for private vehicles and 15% for the heavy-duty vehicles (TRT Trasporti e Territorio, 2017). It is instead assumed that the number of foreign vehicles in BiH compensate the number of vehicles registered in BiH but driving abroad.

The yearly mileage per category have been distributed after in the existing road network using a simplified methodology. To get traffic figure for the whole road network a full-fledged traffic modelling would be preferred. Since that sort of model needs auxiliary information and is beyond the scope of the current project, a simpler approach was taken. The annual average daily traffic (AADT) flow of the roads where measurements were available is subtracted from the total estimated **traffic work** (flow multiplied by the road length) for the entity. The rest of the AADT flow is then distributed on to the rest of the road's segments according to a weight classification assumed for each road type in the Openstreet-map road network.

The same calculations were conducted for both entities. All the basic data, processed data, methodology and the programs to process the data are published in the HIVE data sharing portal.

Emission from vehicles were evaluated using the COPERT 5.4 emission model.
The basis for the model was mostly provided by RHMZ, including:

- Monthly meteorological information,
- Fuel composition from the Brod refinery,
- Statistical Energy consumption from the statistical offices of both entities, when using some conversion factors to comply with COPERT 5.4,
- Fleet of vehicles from official registration files from ministry of transport, using age of vehicles by categories from the Croatian statistical office as an approximation.
- Annual average daily traffic (AADT) from the Croatian statistical office as an approximation (Centar za vozila hrvatske, 2018).

A collaboration was done with the RHMZ in order to provide an estimation of the emissions. Some adjustments might be needed at least regarding the heavy-duty vehicle fleets and the fuel characteristics.
Calculations for the Federation were conducted after discussions with the Centar za razvoj i podršku (CRP) that have previously conducted some COPERT calculations for the Federation.
The COPERT model was run for both entities. All the basic data, processed data, and native COPERT file are published in the HIVE data sharing portal to be freely available to all the stakeholders.

Regional scale emissions

In addition of the inventoried local emissions that have been focused on the major contributors of air pollutants, there are multiple other minor but relevant sources of pollution. These other contributors take place both at the local scale and the surrounding regional scale. To take these other sources into account the compilation of emission is done using the Copernicus Atmosphere Monitoring Service (CAMS) regional emission database (CAMS, 2019).

The CAMS regional emission database is normally built on emission data nationally reported to the UN Framework Convention on Climate Change (UNFCCC), the Convention on Long-Range Transboundary Air Pollution (CLRTAP) and the EU National Emission Ceilings Directive (NEC Directive). Because of errors or incompleteness or inconsistencies it is complemented by other emission data, notably IIASA GAINS emissions.

Since most emission inventories are divided into standardized emission sectors (among others: public power, industry, road transport, aviation, waste, agriculture…) it is possible to complete a local inventory, focused on only some sectors by a selection of missing sectors and/or by the emissions of the surrounding but not inventoried areas.

The CAMS regional emission database alone is not suitable for air dispersion modelling at a local scale since the resolution for this dataset is about 8 x 5.5 km for the studied area. In order to adjust the resolution to the needs of the model, the regional emission dataset was cut out for Bosnia-Herzegovina, re-sampled to 500 meters and re-projected into the local geographical projection, MGI 1901 Balkans Zone 6 (EPSG:3908). The resulting dataset could then be used for Sarajevo and Banja Luka.

The substances in the CAMS emission dataset used are as following and fit the need of the considered modelling (Kuenen, o.a., 2021):

- NOx (nitrogen oxides)
- PM_{10}
- $PM_{2.5}$
- SO_2
- CO (carbon monoxide)
- CH4 (methane)
- NH3 (ammonia)
- NMVOC (non-methane volatile organic compounds)

Set up of the model for Sarajevo

For Sarajevo, there have been several projects to gather emission data during the last decade.

The sectors for which there is local emission data available are:

- Public Power: point sources, mainly district heating facilities, such as residential buildings and public buildings but also some industrial facilities
- Other Stationary Combustion: heating of individual houses and similar
- Transport: on-road traffic

For each of these sectors, the regional-scale emissions are replaced by the local emission data.

Point sources

A dataset containing 613 points sources for Sarajevo was recently developed by E3 (Figure 14). Most sources in the dataset are heating facilities for apartment- or public buildings, such as the facilities operated by Toplane Sarajevo.

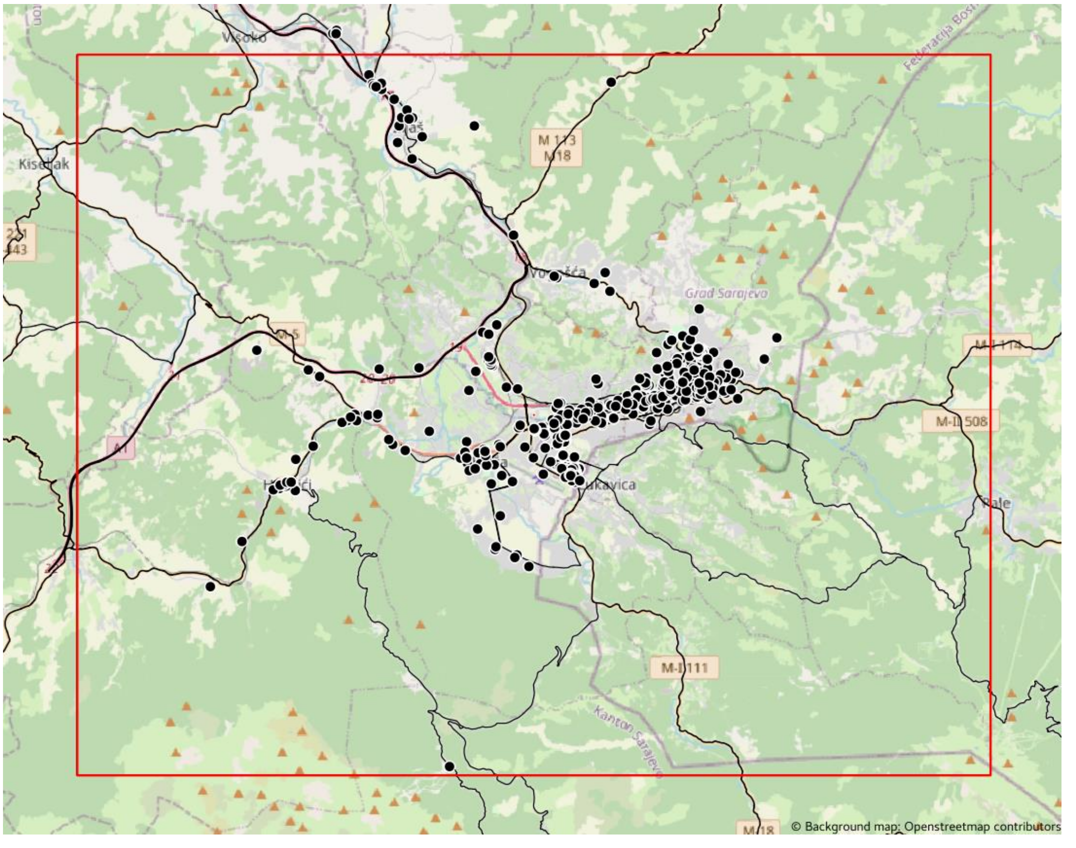


Figure 14: Point sources in Sarajevo (black dots) and modelled area (red rectangle)

For each pollutant modelled, the point-source emissions were rasterized into grids matching the dispersion model setup.

Traffic

The traffic emission dataset has been provided by E3 and contains the main roads inside Sarajevo city (Figure 15).

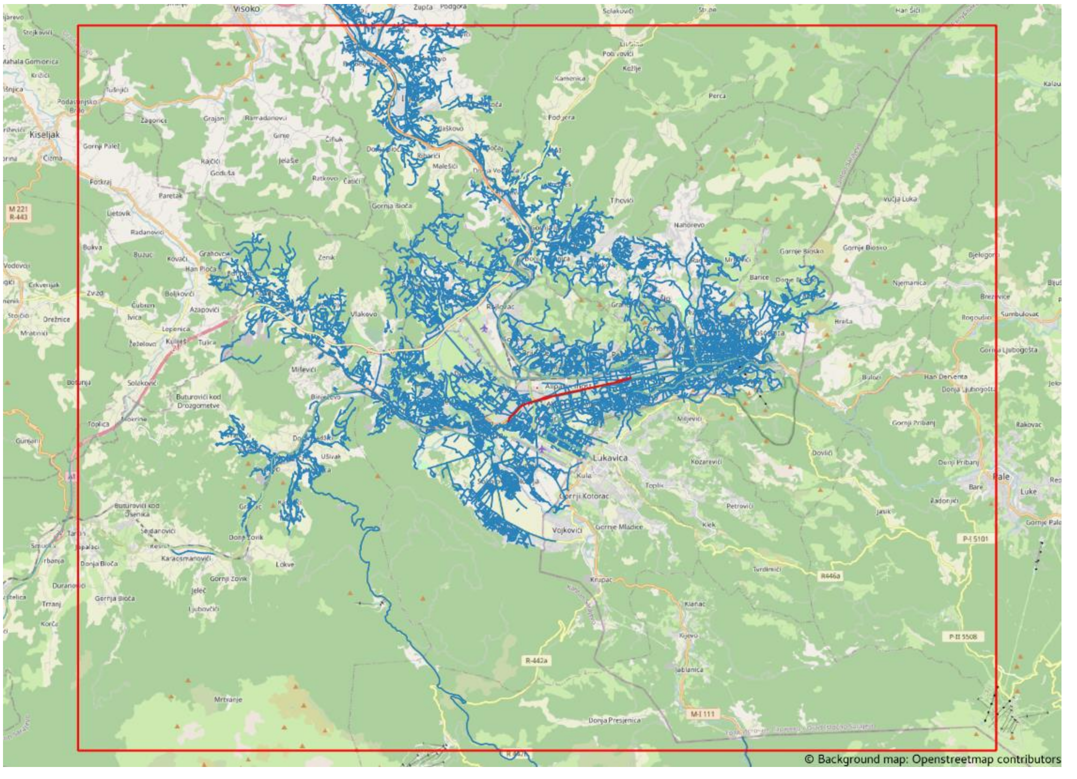


Figure 15: Road network included in the emission inventory of Sarajevo

Since this road network mainly covers the urban parts of Sarajevo and does not include roads outside the Federation of Bosnia-Herzegovina, the dataset was extended using the following method: A road network from the Openstreetmap project was extracted for the area. Using the existing emission dataset, statistics for each road type in the Openstreetmap network were extracted. The statistical values for emissions and traffic flows were then applied to the roads not covered by the local dataset. The method was limited to the larger roads only (Figure 16).

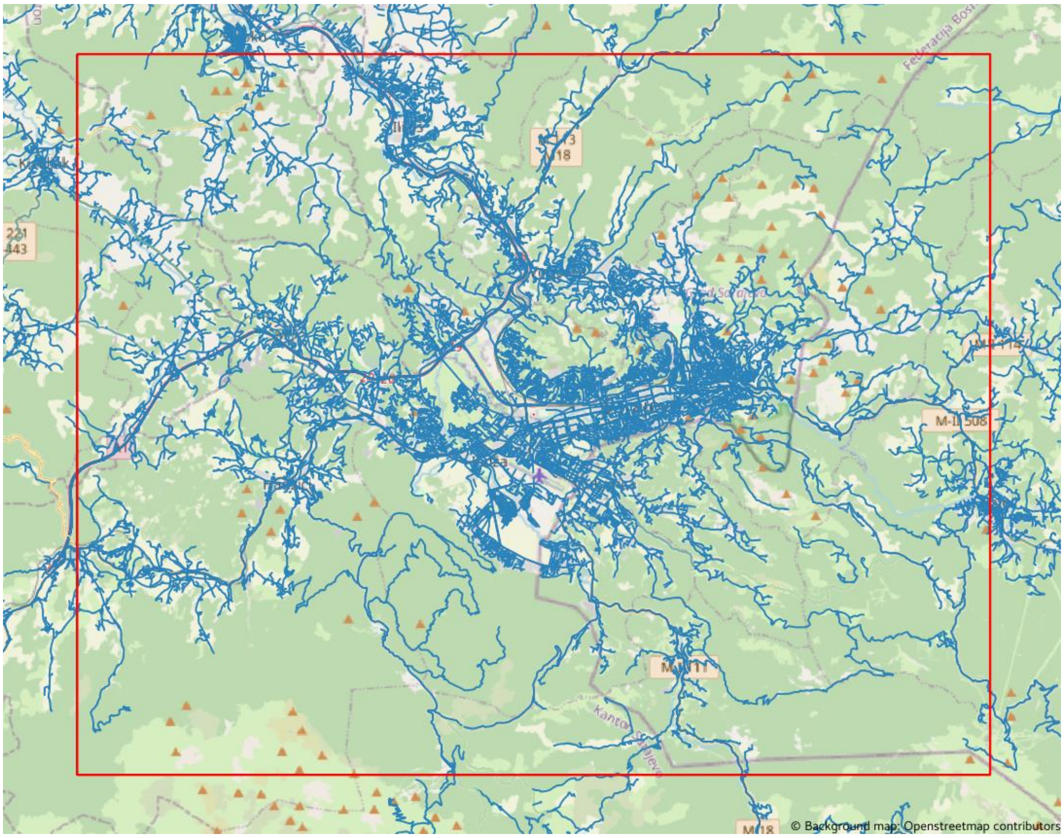


Figure 16: Road network in Sarajevo

For the first runs of the MATCH model, the traffic emissions were rescaled to equal the totals from the original (local) dataset. Since the resulting concentrations were much lower than the observed concentrations, this adjustment was later removed. For particles, wear and re-suspension were included by using aggregated emission factors (ETC/ATNI, 2021). Before importing emissions into the MATCH model, the road emission network was rasterized into 500-meter resolution, matching the grid for the dispersion model.

Residential heating

Most of the larger residential buildings, as well as public buildings, are heated by district heating and facilities localized around a block of houses (condominium). The emissions from these facilities are mostly included in the point sources dataset. The residential heating dataset, in contrast, contains primarily single- or few-family buildings.
For particles (PM_{10}, $PM_{2.5}$) and SO_2, a dataset recently developed by E3 was used, while for NOx an older dataset was selected due to the NOx emissions for the new dataset not being ready.

For the buildings belonging to the Republika Srpska part of the modelling area, heating emissions were estimated using the method using the Urban Atlas and building typology data that was developed in the frame of the project (see section residential heating below). The heating emission polygons for the different residential areas were rasterized into grids matching the dispersion model.

Regional emissions

Emission sectors are defined according to the Selected Nomenclature for Air Pollution (SNAP) source categories. The local high-resolution emissions were placed in three sector categories: Public Power (point sources of larger heating facilities of residential buildings, public houses and some industry), Other Stationary Combustion (individual residential heating sources) and Transport (on-road traffic sources). The remaining regional emissions from CAMS were placed in the remaining sectors as can be seen in Table 33. Note that for the three sectors for which there is local data, only the local data was used.

Total emissions

Total emissions of the main air pollutants for the Sarajevo model domain came from a local emission data set calculated by E3, Ceteor, SMHI, and from the CAMS regional emissions database. In the MATCH model sectors are expressed as SNAP categories.

Sector	NOx	SO_2	PM_{10}	$PM_{2.5}$
Public Power	639.7	396.7	12.2	6.6
Other Stationary Combustion	326.3	617.1	1689.8	1601.5
Industrial combustion & processes*	89.6	911.4	431.8	160.6
Fugitives*	0	0	83.7	9.6
Solvents*	0	0	0	0
Transport	3458.1	2.2	1418.8	1163.4
Other mobile sources*	0	0	0	0
Waste*	102.5	6.1	325.3	304.4
Agriculture*	0	0	38.1	8.4

Table 33: Total emissions (tonne/year) of the main air pollutants for the Sarajevo model domain from local emission data set together with the CAMS regional emissions database (*).

Set up of the model for Banja Luka

Point sources

Commissioned by the IMPAQ project in 2021, Ceteor identified 11 point-sources (mostly heating facilities) in Banja-Luka for which the installed power was known. For some of these, there was also data on used energy available (Eko Toplane Banja Luka d.o.o., 2021). For those sources that lacked data on used energy, we assumed 30% usage of the installed power during the year, based on sources for which this data is available.

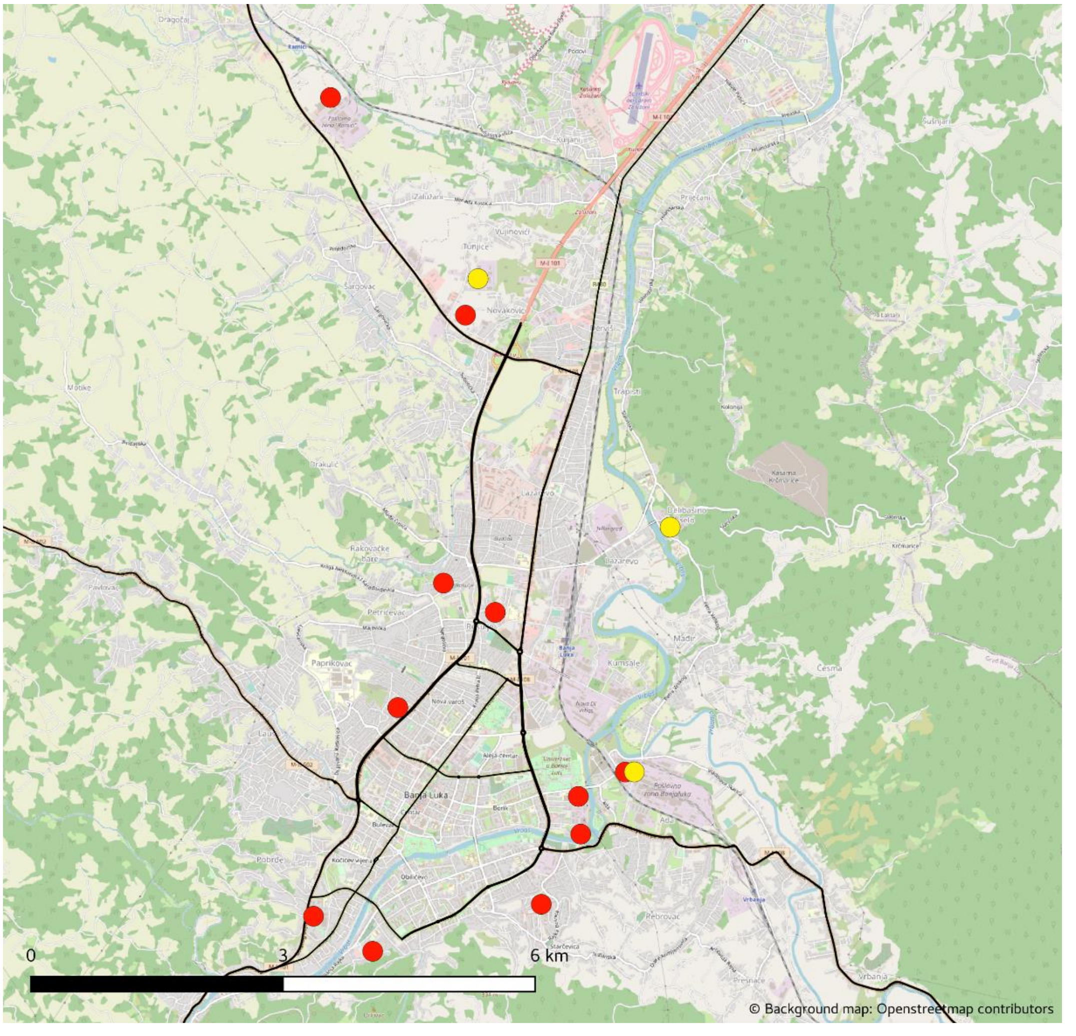


Figure 17: Point sources in Banja Luka. Red dots denote heating facilities and yellow industrial facilities

For the heating facilities point-sources, their emissions were estimated using the EEA 2019 database for emission factors (EMEP/EEA, 2019).

Regarding industrial sources, there are three sources for which the necessary information is available. There are more sources known however, they lack the necessary information to calculate emissions. This is an important area of possible improvements, not only in order to achieve high-quality pollution maps, but in order to have as detailed information about the pollution sources as possible for the city.

Finally, the point-source datasets were rasterized to 500 m resolution, matching the grid for the dispersion model.

Traffic

For the major roads outside the city of Banja Luka, there are traffic counts done by the road administration. Inside the city, however, there is a need to estimate the traffic. This was done using the method presented above under the section "Local emissions data consolidation."

The method used the total traffic from the COPERT modelling setup for Republika Srpska, which has been modelled in collaboration with the RHMZ. The total traffic work (see page 78 for definition) was then mapped onto a road network from the Openstreetmap project, where each road has a weighting factor depending on the road type.

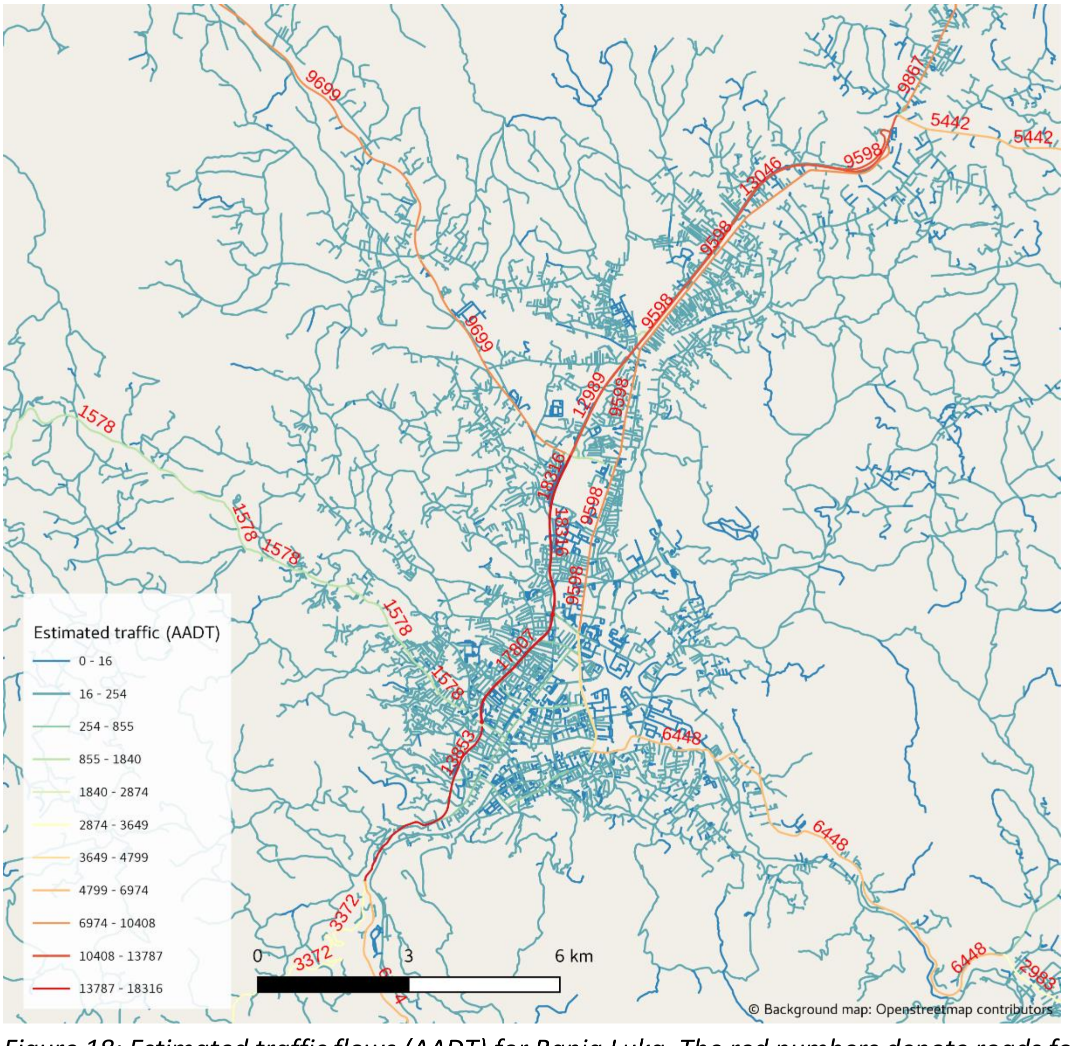


Figure 18: Estimated traffic flows (AADT) for Banja Luka. The red numbers denote roads for which there are traffic count data available

Once an estimate of the traffic work for the major roads was obtained, for each of these roads, the emissions obtained from the COPERT model was distributed with respect to the traffic work. The result is a road network in GIS format, which was rasterized into 500m cells matching the modelling grid. An example map for NOx is shown in Figure 19.

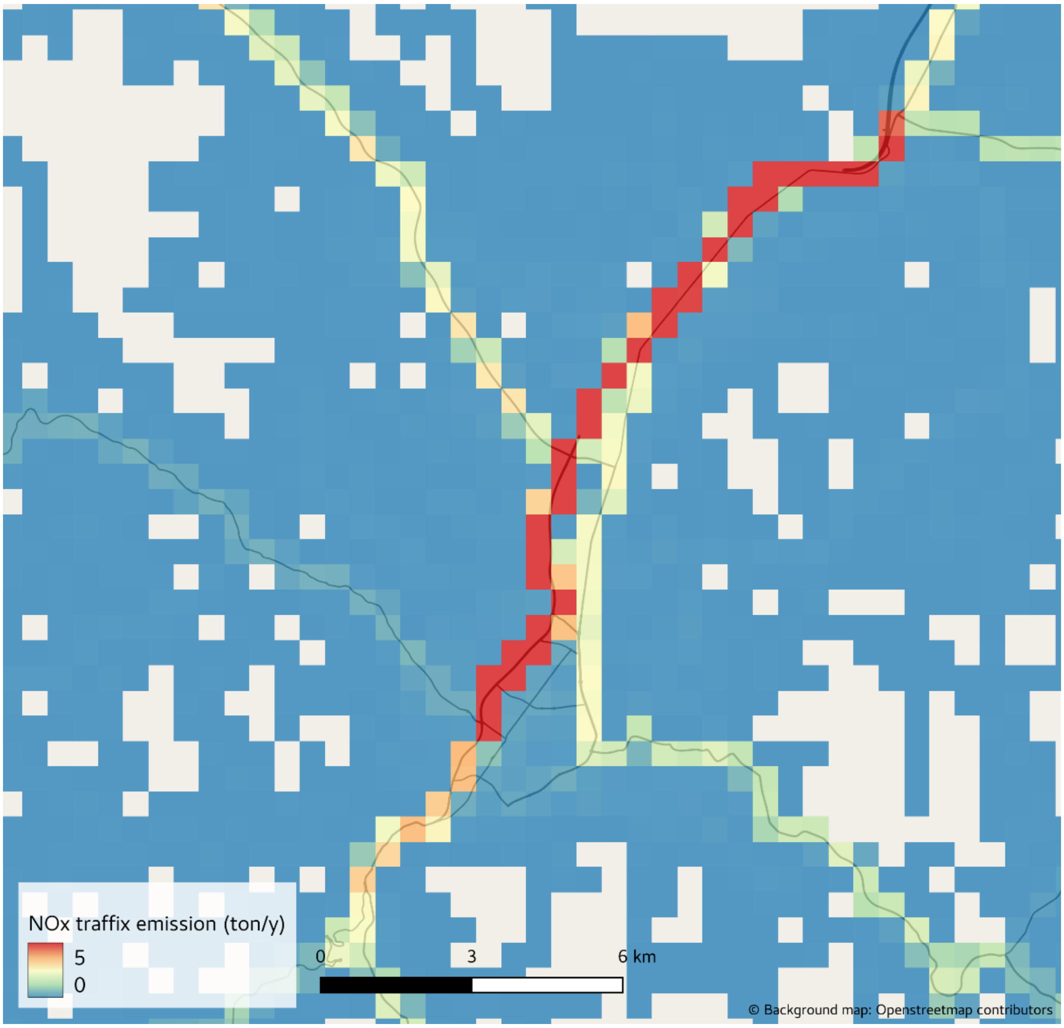


Figure 19: NOx emissions from traffic for Banja Luka, rasterized to 500 m cells

Residential heating

The emissions from heating of individual buildings was estimated using the Urban Atlas 2012 dataset (EEA Copernicus, 2018), combined with the Building typology report (Arnautović et al., 2016) and other sources of statistical data for Republika Srpska (Agency for Statistics of Bosnia and Herzegovina, 2015). Areas where estimates were used appear in light orange in Figure 20 below.

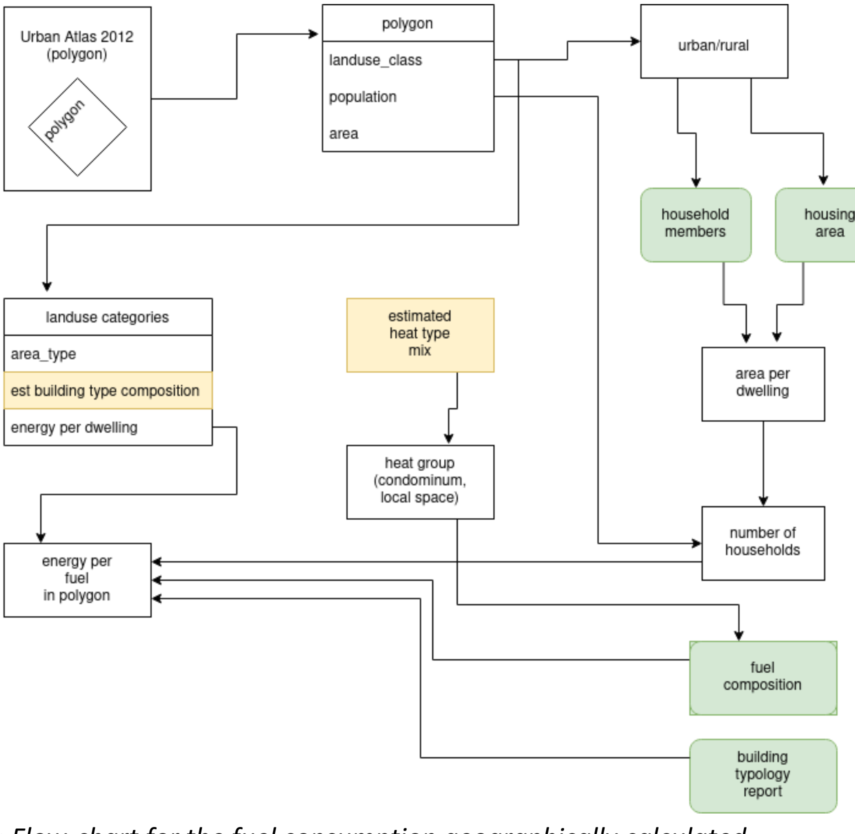


Figure 20: Flow-chart for the fuel consumption geographically calculated

The Urban Atlas GIS dataset contains polygons of different land-use classes as well as population figures for each polygon. Only some of these land-use classes are assumed to cause residential heating emissions. The resulting heating usage are shown in Figure 21.

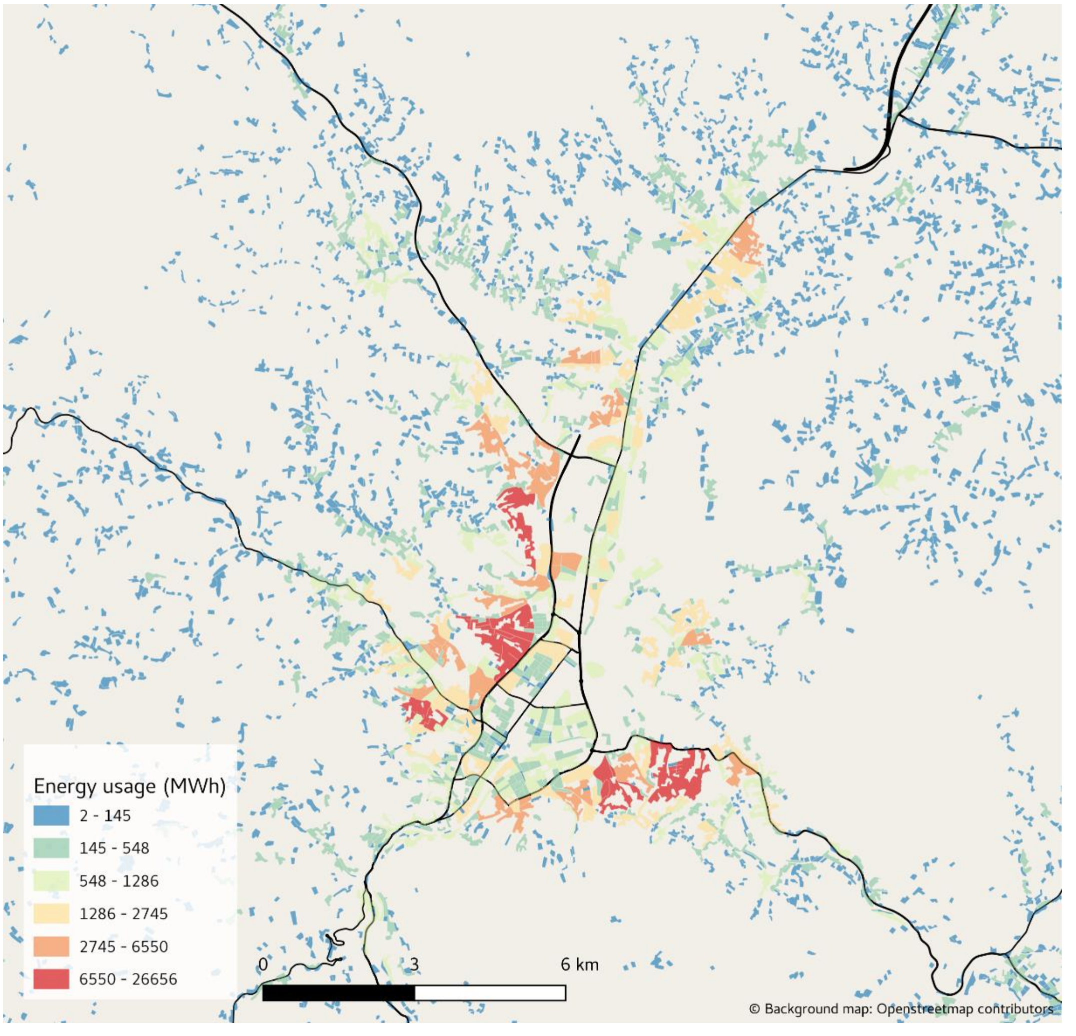


Figure 21: Residential heating usage for Banja Luka. Note that the energy usage in each polygon also depends on the size of the polygon

Once the heating energy usage in each polygon (and for each fuel type) was estimated, the corresponding emissions can be calculated. The flow-chart for this calculation is shown in Figure 22. For each fuel type, we used 2019 emission statistics from the EEA (EMEP/EEA, 2019) for the type of heating appliances used. For each appliance, fuel and substance, the corresponding emission factor may be found in the EEA database. Doing this for all polygons provided a dataset covering all relevant residential areas of Banja Luka, with emissions for all substances for which there are emission factors. The flow-chart for the emission calculation is shown in Figure 22.

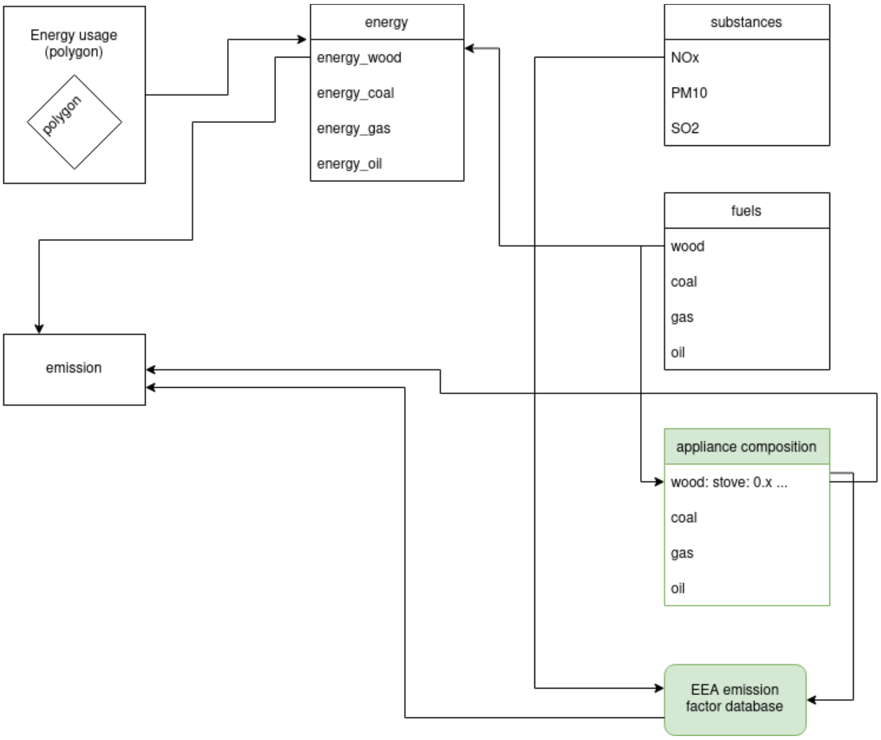


Figure 22: Flow-chart for the emission calculation

The heating emission polygons for the different residential areas were rasterized into grids matching the dispersion model. A map of the resulting NOx emissions is shown in Figure 23.

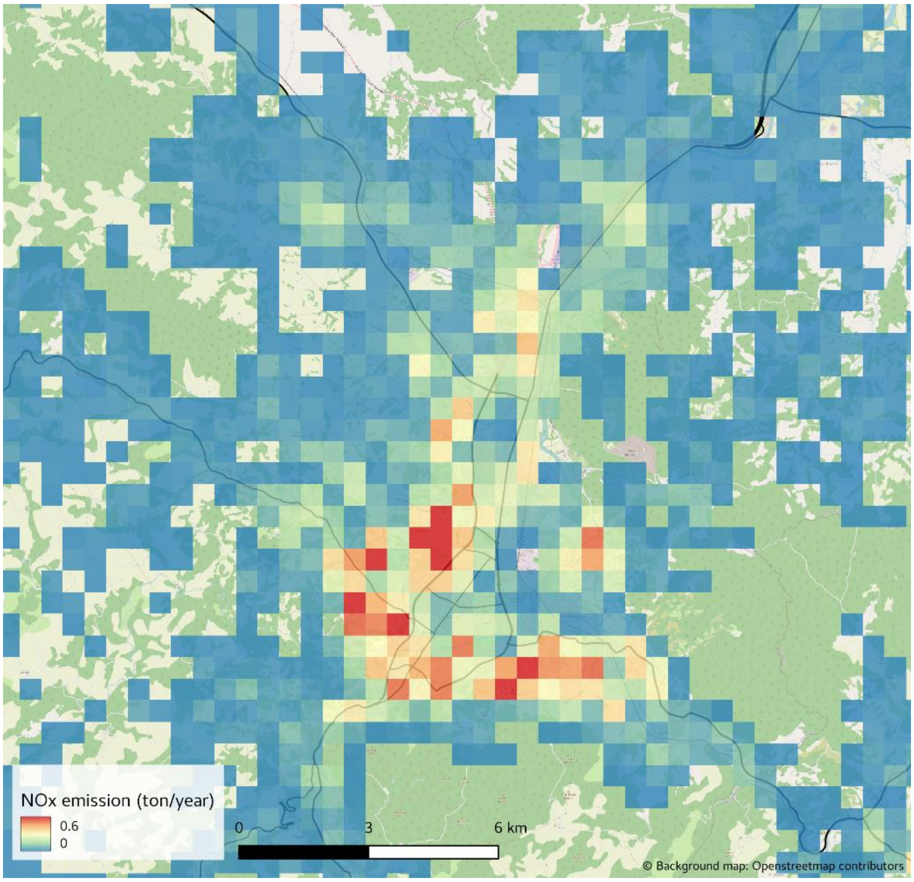


Figure 23: Residential heating emissions for NOx for Banja Luka

Regional emissions

Normally one compares the local emissions inventory with regional data from CAMS. If the emissions match, one can trust the emissions inventory as being comprehensive, and remove the CAMS layer to avoid getting double input. If the emissions don't match, one must combine the existing emissions inventory with the CAMS dataset. Here, there were so few industrial point-sources in the local emission inventory that the regional CAMS dataset was used solely. The emission totals from CAMS were kept, and the resolution was scaled down. For the other sectors, the regional data was replaced by local data, which was better. More details on emission totals for the Banja Luka model domain can be seen in Table 3: Total emissions (tonnes/year) of the main air pollutants for the Sarajevo model domain from local emission data set together with the CAMS regional emissions database (*).

Sector	NOx	SO_2	PM_{10}	$PM_{2.5}$
Public Power	87.4	61.4	181.1	89.1
Other Stationary Combustion	89.7	61.8	2371.8	1170.5
Industrial combustion & processes*	8.1	49	76.4	70.4
Fugitives*	0	0	0	0
Solvents*	0	0	0	0
Transporst	760.3	1.5	181.1	78
Other mobile sources*	0	0	0	0
Waste*	47.6	2.9	292.3	141.3
Agriculture*	0.5	0.1	125.6	23.4

Total emissions

Total emissions of the main air pollutants for the Banja Luka model domain from the local emission data set were calculated by RHMZ, SMHI, and from the CAMS regional emissions database. In the MATCH model sectors are expressed as SNAP categories.

Sector	NOx	SO_2	PM_{10}	$PM_{2.5}$
Public Power	87.4	61.4	181.1	89.1
Other Stationary Combustion	89.7	61.8	2371.8	1170.5
Industrial combustion & processes*	8.1	49	76.4	70.4
Fugitives*	0	0	0	0
Solvents*	0	0	0	0
Transport	760.3	1.5	181.1	78
Other mobile sources*	0	0	0	0
Waste*	47.6	2.9	292.3	141.3
Agriculture*	0.5	0.1	125.6	23.4

Table 34: Total emissions (tonnes/year) of the main air pollutants for the Banja Luka model domain from local emission data set together with the CAMS regional emissions database (*).

Meteorological conditions

It was early decided to conduct a dispersion modelling that could take into account the steep topography, especially in Sarajevo, and the temperature inversion effects that keep the air pollution next to the ground in some winter periods.
This entails the use of a three-dimensional high-resolution meteorological data set. The data set was created using the HCLIM38 model which is a climate version of the HARMONIE model developed by the Rossby Center at SMHI (Lindstedt, Lind, Kjellström, & Jones, 2015). HARMONIE is used by many weather institutes in Europe to make their operational weather forecasts. The HCLIM38 is a simpler and faster version of HARMONIE that fits the needs of dispersion modelling.

In order to get the proper resolution over the urban area, the geographic resolution of weather data was meshed, which means that it was progressively refined from the outskirts to the urban area. Input data for the outer edge was acquired from the global model ECMWF (18km resolution), the whole Bosnia and Hercegovina was calculated with a 12km resolution and both Sarajevo and Banja Luka urban areas was calculated with a 1 km resolution.
In each point of the grid several weather parameters (wind direction, wind speed, temperatures, humidity, precipitation, etc) were calculated on an hourly basis for the year 2018. The model provides vertical levels that are the necessary input of meteorological fields for MATCH to calculate atmospheric processes. These include advection and vertical wind fields important for assessing the influence of air mass transport on emitted air pollution species.

The MATCH-model also calculates the boundary layer height (ZI), which indicates how strong the vertical turbulence is over an area. A low boundary layer signifies low vertical air mass movement and is strongly associated with conditions promoting inversions near the

surface. In the result section for each city an analysis of the wind speed and boundary layer height was provided.
The results of the model were compared to the available weather observations at ground level and showed consistency with the daily average temperatures, and also show a significant but acceptable overestimation of the wind speed in both extremes (low and high). The meteorological weather stations used to validate the HCLIM result for Sarajevo were Bjelave, Butmir and Bjelašnica and the synoptic station in Banja Luka.

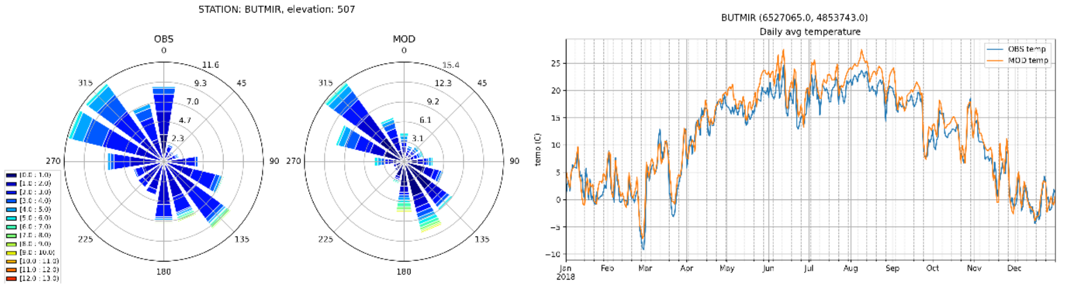


Figure 24: Comparison of Modelled and Observed temperature, wind speed and direction at the Butmir air quality and weather monitoring station for the year 2018.

Final set up of MATCH model

The regional off-line Eulerian CTM MATCH model was used with the chemical scheme based on (Simpson et al., 1993) with further extensions described by (Andersson, Helène, Robertson, Karlsson, & Engardt, 2017). The scheme considers 70 different chemical components and ca. 150 chemical reactions e.g. 28 photolysis reactions, aqueous phase oxidation of SO_2 and simple NHX chemistry. The model structure, advection, vertical winds and more are further described by Robertson and Langner, 1998.

Boundary concentrations were derived from European scale modelling with MATCH, which uses regional emission data from CAMS-REG v4.2. Background concentrations of the main air pollutants such as NO_2, SO_2, PM_{10} and $PM_{2.5}$, but also for several other pollutants such as a number of NMVOCs, NH3, CH4, and a range of other components important for e.g. chemical reaction mechanisms, were fed into the model domain every three hours.
Model simulations were run with 500 m grid resolution and hourly time steps all over the reference year of 2018. Emissions data from CAMS were re-mapped from GNFR to SNAP sectors and standard time variations defined for each sector was used.

Simulations and iterative tuning

All the different data and parameters described above come with their own uncertainty that affect the quality of the end results. For that reason, it is important to assess the results from the model against similar modelling, and against air quality measurements from official monitoring stations. These comparisons, or quality assessment, often lead to some adjustments of the model settings before a new run and a new quality assessment.

A typical quality assessment cycle would include the following steps:

Activity	Further details and purpose of activity
Prepare input data	E.g. meteorology and emissions
Setup model and run	Adjust/set and necessary settings of the model
Monitor run	Input data ingested. Errors? Warnings? Output?
Check log of runs	Errors? Warnings? Output?
Create maps	Inspect results, reasonable?
Extract data air pollution levels	For a given location, official monitoring station.
Create time series diagram	Variation of levels over time? Peaks/dips when?
Compare results with measurements	Similar trend? Peaks/dips? Underestimations/overestimations?
Analyse meteorology influence	Peaks/dips explained by meteorology?

Table 35: Example of model setting and quality assessment cycle

Several model simulations with various versions and recalculations of emission data were carried out to assess the quality of the modelled pollution concentrations. In this study, seven cycles of test simulations were carried out until no further improvements could be achieved.

Source apportionment

Source apportionment was carried out for the three sectors of most importance and where local emissions has been calculated (Public Power, Other Combustion Sources and Transport). For the remaining sectors (Industry, Solvent, Fugitives, Waste and Agriculture) containing emissions from the regional CAMS data set, were placed in one group and named “other”. This group also included the background concentrations coming in from outside of the local model domain. Source apportionment was then carried out for the remaining sectors since the apportionment of such low contribution would not be relevant because of its high uncertainty. More work on the emission data of these sectors is required before carrying out any complementary apportionment.

The methodology used to calculate the source apportionment of the mentioned sectors was to reduce one sector by 25% and run the model with the same settings otherwise. Note that reducing the amount of emission of only 25% - and not of 100% - allows the model to consider the interactions between the pollutants from different sources regarding both the atmospheric chemistry, and the local weather effects.

Air pollution levels within each sector reduced were then calculated successively, resulting in three separate simulations, each representing concentration levels with one sector reduced. To estimate the full contribution from each sector the simulation containing the total modelled concentrations were subtracted from the simulation containing the reduction of each sector and multiplied by four.

Sector contribution = (Concentration total – Concentration reduction sector) x 4

In the end, four final simulations for each city were carried out. A first simulation representing the total concentrations for 2018, followed by three additional simulations representing source contributions of the three sectors (Public Power, Other Stationary Combustion and Transport).

Results

Results for Sarajevo

Meteorology

The model shows that episodes of stable weather conditions signified by a low boundary layer height (ZI) were frequent in Sarajevo during 2018 (Figure 25). Several days with boundary layer height below 200m were observed in the period from January to March and November to December. These situations can be interpreted as representing very stable weather conditions with inversions when air exchanges with upper layers were extremely restricted or non-existent and emissions into the air accumulated near the ground. The boundary layer height varied between 100m to 600m in the winter half of the year while in summer it could reach up to over 1000m. The boundary layer was usually lower during the winter and higher during the summer which is also shown in the model.

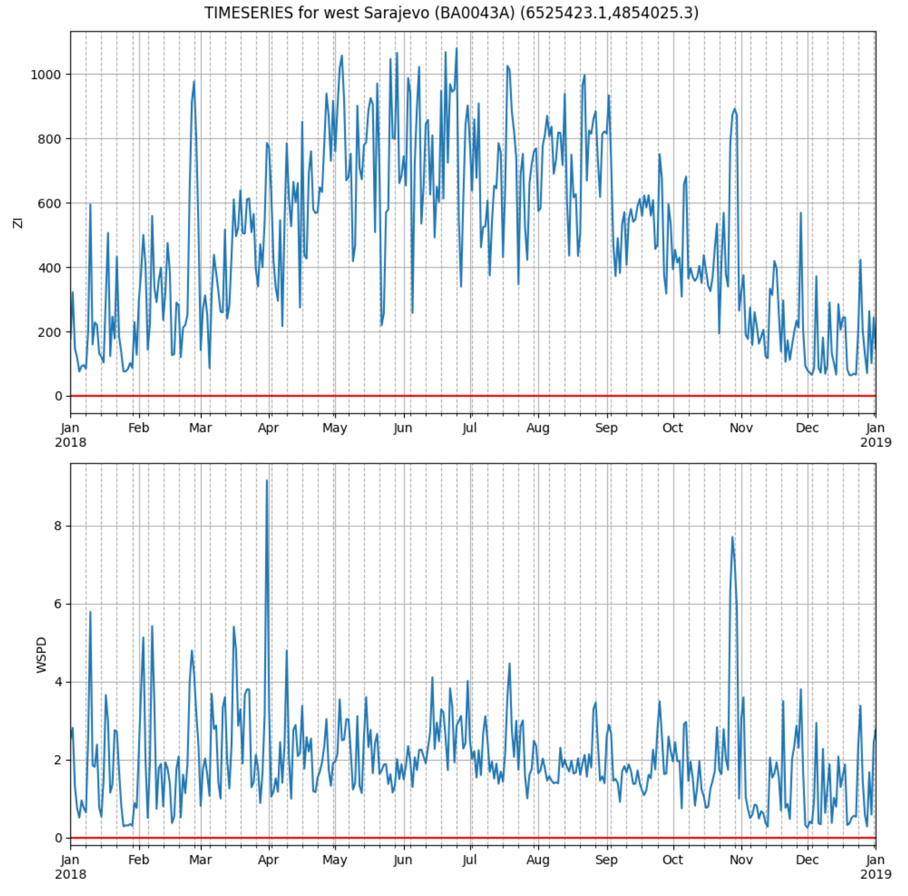


Figure 25: Daily averages of the boundary layer height (ZI) in meters and wind speed (WSPD) in m/s, calculated by the MATCH model for a grid point in Ildiza, west Sarajevo, 2018.

Interestingly, even during some days in the spring and summer, the boundary layer was quite low, especially in April and May, but also in June and July it reached below 400m. The boundary layer height was driven mainly by the temperature, wind and cloud cover. In a valley city like Sarajevo, inversions develop even easier. At night when the temperature drops, cold dense air may sink down from hillsides into the valleys, creating cold-air pools (Largeron & Staquet, 2016). Throughout the year, wind speeds were often quite low, (below 2 m/s, Figure 25b). Windier episodes were observed in January – March, with wind speeds often above 4-5m/s as a daily average. Two stormy days were also observed at the end of March and October with winds around and exceeding 8 m/s. Summertime winds were generally low to moderate, varying between 1.5 m/s to 4.5 m/s as a daily average.

NO_2 seasonal analysis

The winter half of the year showed the highest modelled levels of NO_2 in Sarajevo. Levels were high in the centre of the city and along roads surrounding the city (Annex A-1). It was also obvious that concentrations decreased with distance from local sources, confirming that NO_2 is mainly a local pollutant. Comparing the first quarter (January, February and March: JFM) with the last quarter (October, November and December: OND) the plume over the city looks more widespread with high concentrations covering a larger area over the city during the last quarter. During both quarters, the model showed several episodes with cold and stable conditions, however the first quarter had episodes of relatively high wind speeds where peaks reached 4-5 m/s, as can be seen in Figure 25b. This led to more effective ventilation of the air in between the calm episodes and thus explained the lower accumulation of NO_2 concentrations during the start of the year. While in the last quarter (OND), emissions accumulated more effectively near the ground due to less ventilation of the air. Calm episodes were interchanged with peak winds reaching only between 3-4 m/s, thus less effective ventilation was observed at the end of the year. During the summer half of the year (April May and June: AMJ) and (July, August and September: JAS), NO_2 levels were significantly lower. The air tends to be more well mixed during the summer due to higher temperatures resulting in turbulence, as can be seen in Figure 25a where the Zi value calculated by the model is high during the warm months of the year.

NO_2 source apportionment

Comparing the levels of the yearly mean (Annex A-2 a) with the estimated contribution levels for each sector it was obvious that a significant part of the modelled NO_2 comes from the transport sector (Figure 26). This can also be seen in the spatial maps in Appendix A (Annex A-2 d). Ca. 40-70% of the NO_2 concentrations throughout the year originate from transport (Figure 12). In the summer it was 70% and, in the winter, it varied between 40-50%. A relatively small fraction (~1-5%) came from large scale heating (sector 1, annex A-2) and the rest (25-30%) originated from other sectors (which includes industry and waste). Interesting to note here is that the transport sector has lower emissions during the winter half of the year. During winter months we also see that other sectors contribute to the concentrations to a larger degree, which is most likely due to the sectors for industry and waste (Figure 26) where emissions are relatively low. The model generally underestimates the observed NO_2 levels throughout the year (black line in Figure 26).

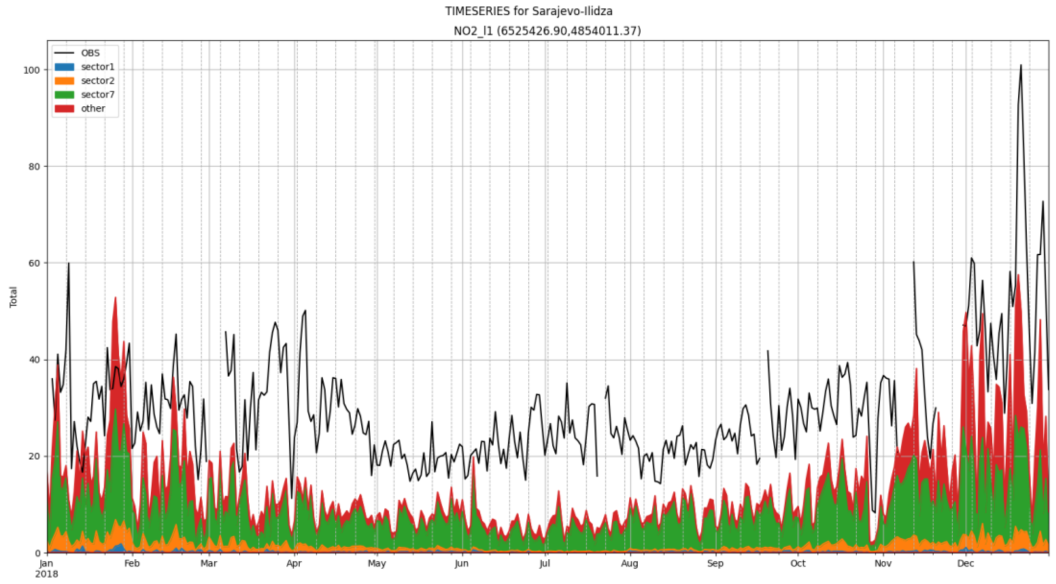


Figure 26: Daily simulated sector contributions to NO_2 levels of sectors Public Power (blue sector1), Other Stationary Combustion (orange sector 2), Transport (green sector7) and other sectors including background concentrations (red other). Daily NO_2 observations (black line) come from the measurement site in Ildiza, Sarajevo and model data represent a grid point at the same location. The total simulated NO_2 concentrations are represented by the red stack plot.

During the summer, when pollution levels are relatively low, modelled levels differ from observations with a varying factor of 2 to 3. This means that observations are generally two to three times as large as the modelled levels. During winter the underestimations are lower and reach a factor of about 1.8 in comparison with observations. The pattern from a day-to-day basis seems to follow the same trend where observations show peaks so does the model. This suggests that the model captures the atmospheric influence on pollution levels quite well while the emission totals are likely too low. Another aspect to consider is the time variations of e.g. the transport sector are not perfectly described for the traffic intensity in Sarajevo. The upward trend seen for observations in June and July is not seen in the modelled levels and could be explained by the fact that the model assumes that the emissions from traffic decrease in July – this might not be the case in Sarajevo where traffic may actually increase due to tourism. Thus, the standard time variations built into the model could be improved with access to more data regarding e.g. traffic counts on an hourly or daily basis throughout the year. Another contributing factor to low modelled NO_2-

levels is likely due to the slightly overestimated wind speeds by the HCLIM38 model. NO_2 is sensitive to wind and concentration levels are known to decrease with higher wind speed (Grundström et al., 2015). This is obvious both in the observations and modelled levels at the end of March and October where significant depressions are observed at days with stormy weather.

SO_2 seasonal analysis

From the spatial maps it can be observed that SO_2 levels were high in the winter half of the year (JFM and OND Figure A-3) but influence from a non-local source could also be observed where transport of SO_2 from the northwest is obvious. High levels were observed away from local sources especially during the first quarter (JFM Figure A-3 a). The windier conditions during the first quarter signify that transport into the model domain/Sarajevo was effective, while during the last quarter, transport occurred to a lesser degree when winds were lower. Local sources thus dominated the contribution to high levels to a larger degree in the last quarter. In other words, transport was less significant in the last quarter where winds were generally lower.

SO_2 source apportionment

Out of the local emissions data set, the domestic heating sector is the most significant contributor to SO_2 levels in Sarajevo. The sector varies between 1% in the summer to 40% in the winter with regards to its contributions to the total SO_2 levels (Figure 27). The sectors for transport (sector 7) and public power (sector 1) contribute very little to the SO_2 levels, varying between 1-10% throughout the year. A source from the public power sector was visible in the area between Opcina Vogosca and Hotonj, just north of Sarajevo, and can be seen in the spatial maps for sector contributions (Annex A-4 b). The largest contribution come from the other sector category, varying between 60-99%. Here the contribution from local industrial sources is expected to be significant since the total emissions for this sector in CAMS are relatively high (Figure 27). In the current emission setup, the contribution from these industrial point sources is smudged out over a large grid area due to the low spatial resolution of the dataset, therefore also disguising any local concentration gradients. Transport from outside the city is also important as can be seen from e.g. the yearly mean spatial map (Annex A-4 a) and the seasonal maps especially during the first and fourth quarters (JFM and OND in Figure A-3 a and d). The correlation between modelled and observed values is quite poor for SO_2 (Figure 27).

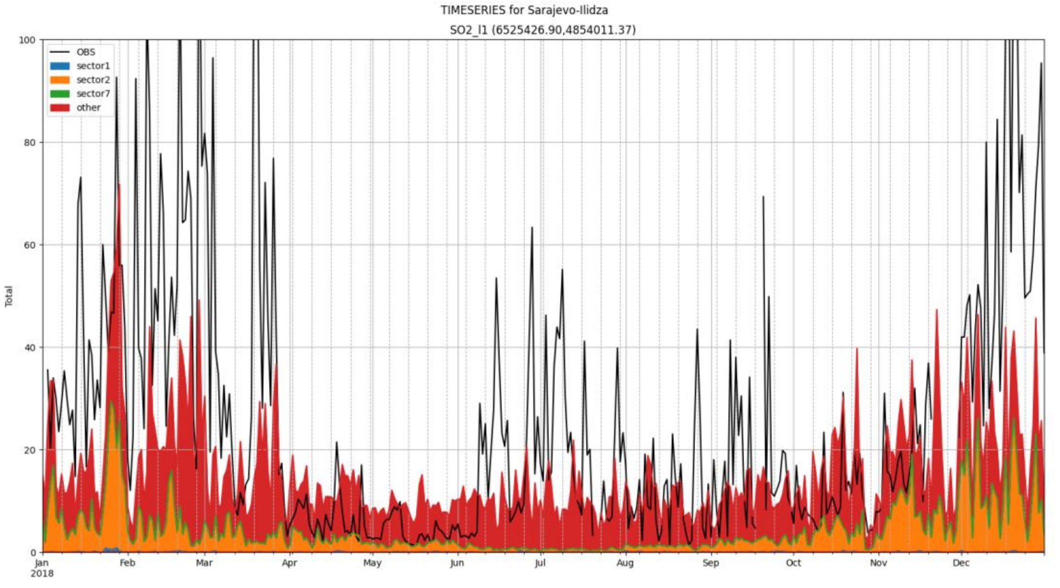


Figure 27: Daily simulated sector contributions to SO_2 levels of sectors PublicPower (blue sector1), OtherStatComb (orange sector2), Transport (green sector7) and all other sectors including background concentrations (red other). Daily SO_2 observations (black line) come from the measurement site in Ildiza, Sarajevo and model data represent a grid point at the same location. The total simulated SO_2 concentrations are represented by the red stack plot.

Modelled levels were generally underestimated by a factor of three or more during observed peaks both in winter and summer. Furthermore, modelled levels were largely overestimated between April and June. These fluctuations between under- and overestimations by the model may signify that the emission data needs significant improvement to correctly describe the actual source emissions in Sarajevo.
From June to September there were large observed peaks in SO_2, recurring on a weekly basis, which the model did not capture. These peaks are likely driven by a local source, which most probably is missing in the emission data. The peaks may also be a result of transport from outside the region as indicated in the second and third quarters in the seasonal maps (Figure A-4 b and c). North westerly winds were quite common in June and July thus transport form this direction could carry plumes of emissions from both local and distant sources. The emissions from industry and electric power in the city of Kakanj (Tvornica cementa and Termoelektrana) could both contribute significantly to long distance transport of air pollutants into the Sarajevo region. These plants are located 30-40 km north-west of Sarajevo and the emission plumes from these plants are anticipated to be diluted before reaching Sarajevo. Extending the model domain to include these regions could reveal more information about the emission contributions from these plants and the transport from north-west. Furthermore, a back-trajectory model such as HYSPLIT could be used to track the origin and air mass movement over time. This can provide more

information regarding where an air mass has been transported and the potential sources that may have influenced the plume.

Particles, seasonal analysis

Both PM_{10} and $PM_{2.5}$ show high levels during JFM and OND (Annex A-5 and Annex A-7), especially OND similar to NO_2 pattern. Levels decrease with distance from the local sources, suggesting that the concentration load of particles is predominantly driven by local emission sources. Indication of transport from outside the model domain is apparent during OND from the northwest. However, this could also be an effect of the city plume being transported away from the local sources in Sarajevo with south-easterly winds. The most frequent wind directions in Sarajevo are north-westerlies and south-easterlies which push the city plume in these directions while also transporting particles into and out of the city. Levels during AMJ were relatively low and uniform over the whole model domain with a tendency to form higher levels around the centre of Sarajevo. Emissions were generally lower during summer but well mixed conditions were common and driven by strong thermal turbulence during the warmer months, creating large vertical eddies in the atmosphere resulting in particles being spread both vertically and horizontally over the model area. Levels during late summer and early autumn (JAS) were relatively low but concentrated to areas where local emission sources exist.

Particles, source apportionment

Domestic heating (sector 2 in Figure 28) was a significant contributor to the PM_{10} levels, up to 50-60% in the winter, while quite low in the summer ~15%. The transport sector contributed ca 15% to PM_{10} levels in winter and was also very low in the summer. The other sector category was significant during the summer half of the year.

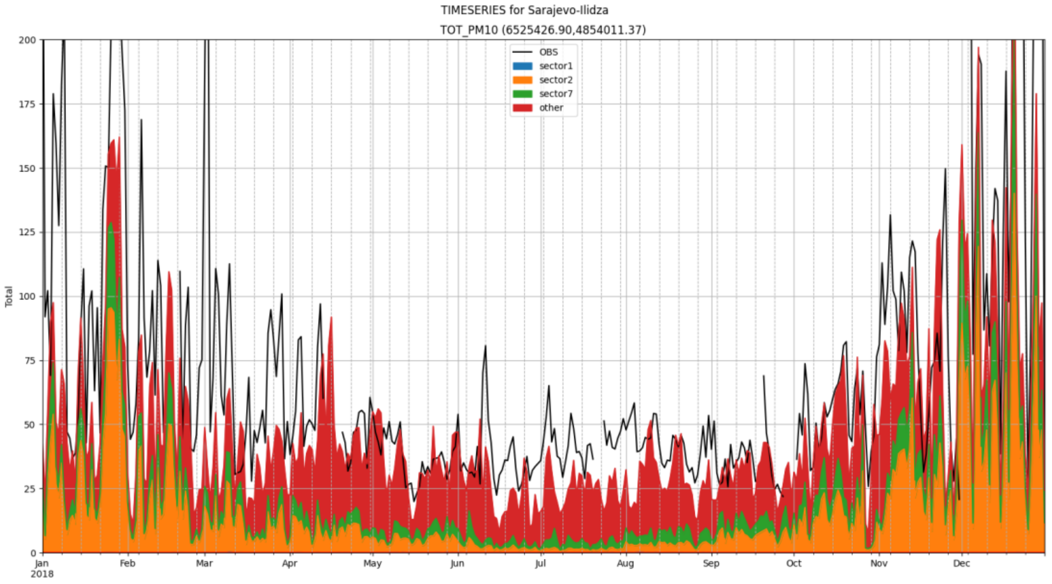


Figure 28: Daily simulated sector contributions to PM_{10} levels of sectors PublicPower (blue sector1), OtherStatComb (orange sector2), Transport (green sector7) and all other sectors including background concentrations (red other). Daily PM_{10} observations (black line) come from the measurement site in Ildiza, Sarajevo and model data represent a grid point at the same location. The total simulated PM_{10} concentrations are represented by the red stack plot.

Waste and industrial sources are likely very important here with their relatively high emissions but there are also emissions from fugitives and agriculture, though they are low in comparison to other sectors. These emissions are from the CAMS regional emission data set with a coarser resolution than the new local emission dataset. It would be useful to review these emission sources and calculate them on a higher grid resolution in order to carry out a qualitative source apportionment on these sectors. Furthermore, PM_{10} levels were also influenced by transport from outside the city (Annex A-6 a). Comparing the total modelled levels (red stackplot in Figure 28) with observations (black line in Figure 28) it is obvious that modelled PM_{10} levels correlate quite well with observations with some exceptions for a couple of peaks observed in e.g. January, February and March. The overall difference between modelled PM_{10} levels are ca half of the observed levels. During peak PM_{10} levels the difference is much larger in e.g. March where observed levels were about five times larger than the modelled levels. For the fine particle fraction a similar pattern is observed to that of PM_{10}.

A large part of the PM_{10} fraction consist of $PM_{2.5}$ and thus the analysis hereby made for PM_{10} also applies for $PM_{2.5}$. No analysis can be made between modelled and observed values for $PM_{2.5}$ however since there is a lack of observation data for this component.

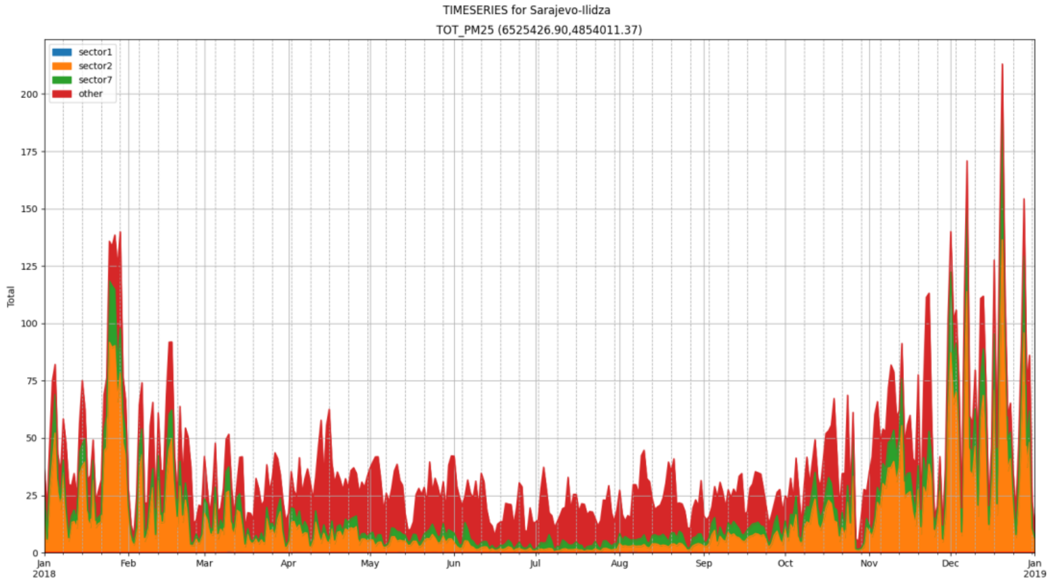


Figure 29: Daily simulated sector contributions to $PM_{2.5}$ levels of sectors PublicPower (blue sector1), OtherStatComb (orange sector2), Transport (green sector7) and all other sectors including background concentrations (red other). Daily $PM_{2.5}$ observations (black line) come from the measurement site in Ildiza, Sarajevo and model data represent a grid point at the same location. The total simulated $PM_{2.5}$ concentrations are represented by the red stack plot.

Results for Banja Luka

Meteorology

The boundary layer height varied throughout the year in Banja Luka as expected, (Figure 30a) with low heights in the winter and higher in the summer. Very low boundary layer (<200m) was frequently observed in January-February and November-December, but also one day in May. Wind speeds varied between 0.5 to 5 m/s as a daily average throughout the year (Figure 30 b).

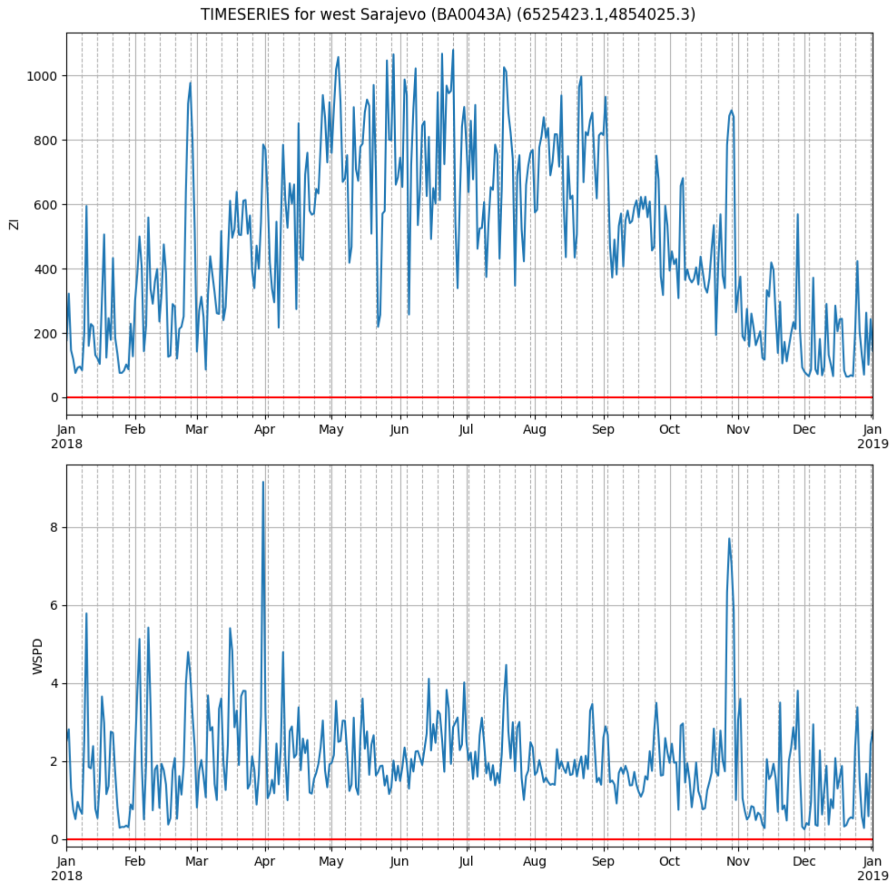


Figure 30: Daily averages of the boundary layer height (ZI) in meters and windspeed (WSPD) in m/s, calculated by the MATCH model for a grid point in the central part of Banja Luka, 2018.

In the summer period relatively low winds were observed, varying between 1 to 3 m/s and episodes with very low winds (<1 m/s) were frequent in January-February and November-December, but also some days in April-October. At the end of March, April and October winder conditions were observed, with wind speeds reaching above 4 and 5 m/s. March was in general a very windy month.

NO_2 seasonal analysis

As can be seen from the spatial maps in Appendix B, modelled concentrations for NO_2 in Banja Luka are generally quite low, with quarterly means reaching up to 8 ug/m3 in winter (JFM and OND Annex B-1 a and d) and 5 µg/m^3 in summer (JAS, Annex B-1 c). This is expected since the emissions are very low (Table 2). Winter seasons show the highest levels and more so at the end of the year (Annex B-1 a and d). In the summer NO_2 is lower (Annex B-1 b and c). High levels are observed near the centre of Banja Luka and along traffic routes.

NO_2 source apportionment

The concentrations for NO_2 in Banja Luka are low on average. Overall, the traffic sector represents the largest contributions to the total levels of the year. Peaks stand for ca. 60% (sector 7 in Figure 31).

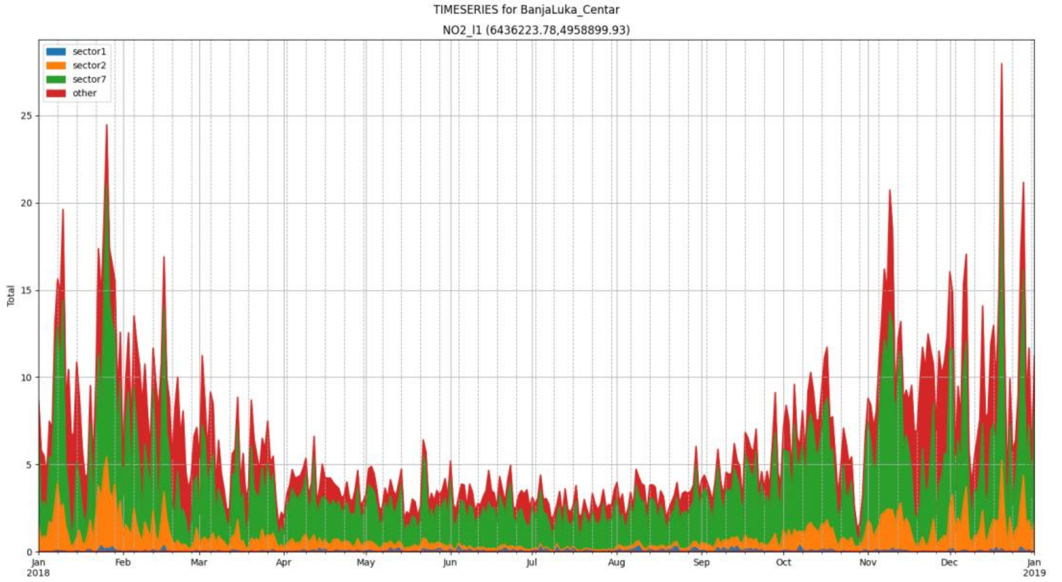


Figure 31: Daily simulated sector contributions to NO_2 levels of sectors PublicPower (blue sector1), OtherStatComb (orange sector2), Transport (green sector7) and all other sectors including background concentrations (red other) for a grid point in the centre of Banja Luka. The total simulated NO_2 concentrations are represented by the red stack plot.

Domestic heating adds to the concentrations mainly in the winter, especially in January-February and October- December (sector 2 in Figure 31) when it varies between 1-16% of the total modelled concentration levels. Other sectors vary between 20-80%, with high contributions during winter peaks. In this category several sectors are included together with the background levels. Background levels are expected to have a very small or negligible effect on the levels in Banja Luka since NO_2 is mainly driven by local sources. Since the modelled concentrations are very low to start with, the result of the modelled concentrations in Banja Luka are to a large degree uncertain. Further work with the emission totals is needed to improve the modelling result.

SO_2 seasonal analysis

Looking at SO_2 levels, a general observation is that the modelled concentrations in Banja Luka are very small throughout the year. Quarterly means only reach up to about 8 ug/m3 and a source area can be noticed in the north-eastern part of Banja Luka (Annex B-3 a-d) where the city's highest modelled SO_2 levels can be observed. High levels are observed in the winter half of the year, especially linked to a point source in Banja Luka (Annex B-3 a and d). There is also some influence from the background in westerly, southerly and easterly directions during the first months of the year (Annex B-3 a). The visible source area in Banja Luka is likely a steel and iron plant (Jelšingrad Livar Livnica čelika) and originates from the regional CAMS emission data for the industrial sector. To further identify and confirm that it is this actual source, CAMS emissions need to be further assessed to identify the specific source(s) included in the coarser grid data and re-calculated onto a higher grid resolution. A higher grid resolution will then allow for a more precise source location and thus also identification.

SO_2 source apportionment

For SO_2 it is obvious that the other sector category represents the largest proportion of the total modelled daily levels and contributes up to 90% of the daily SO_2 levels (red stack plot in Figure 32). The industrial sector together with the background levels are the most significant emission sources in this sector category as other sectors have very small or no emissions (Table *3: Total emissions* (tonnes/year) of the main air pollutants for the Sarajevo model domain from local emission data set together with the CAMS regional emissions database (*).

Sector	NOx	SO_2	PM_{10}	$PM_{2.5}$
Public Power	87.4	61.4	181.1	89.1
Other Stationary Combustion	89.7	61.8	2371.8	1170.5
Industrial combustion & processes*	8.1	49	76.4	70.4
Fugitives*	0	0	0	0
Solvents*	0	0	0	0
Transporst	760.3	1.5	181.1	78
Other mobile sources*	0	0	0	0
Waste*	47.6	2.9	292.3	141.3
Agriculture*	0.5	0.1	125.6	23.4

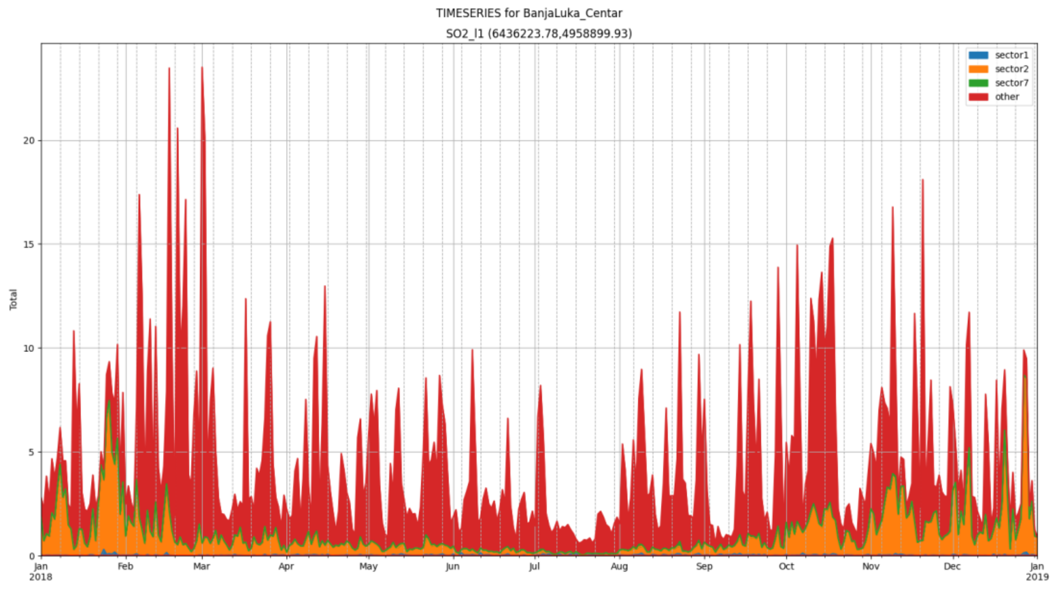


Figure 32: Daily simulated sector contributions to SO_2 levels of sectors PublicPower (blue sector1), OtherStatComb (orange sector2), Transport (green sector7) and all other sectors including background concentrations (red other) for a grid point in the centre of Banja Luka. The total simulated SO_2 concentrations are represented by the red stack plot.

Some peaks, however, are dominated by the domestic heating sector, e.g. in January and December and can reach up to 65-90% of the total concentration level. Transport and public power represent a very small proportions of the pollution levels. Looking at the yearly mean in the spatial maps (Annex B-4) these findings are also noticeable where domestic heating varies from 0-13% (Annex B-4 c), while Public Power and Transport look rather empty over the model domain (Annex B-4 b and d). The fact that PublicPower does not contribute as much to the pollution load at ground level is explained by the fact that a large proportion (99.75%) of the emissions from this sector are injected into the atmosphere at a higher level (184 m height or higher). Emissions at higher levels will to a larger degree be subject to vertical transport in the atmosphere and a smaller fraction of the emissions will reach the ground. Thus, concentration levels at ground level will be calculated lower. Emissions from domestic heating sources, (placed in the Other Stationary Combustion sector) however, are all emitted at ground level by the model.

Particles, seasonal analysis

Both coarse and fine particles (PM_{10} and $PM_{2.5}$) show high levels during the winter in Banja Luka (Annex B-5, B-7 a and d). The lowest levels are observed in late summer early autumn (annex B-5 c). Concentrations decrease with distance from the city centre and from roads around indicating that concentrations are dominated by local sources. However, in the end of the year (OND, Annex B-5 d) there seemed to be relatively high PM_{10} levels over the whole model domain, potentially indicating transport from outside regions.

Particles, source apportionment

In the winter, when particle levels are high, the domestic heating sector frequently represent the largest proportion (up to 90%) of the modelled peak levels in the central parts of Banja Luka (Figure 34). During spring and summer months the other sector category dominated the PM_{10} pollution levels, varying between 50-90% of the total levels, which are generally lower in magnitude.

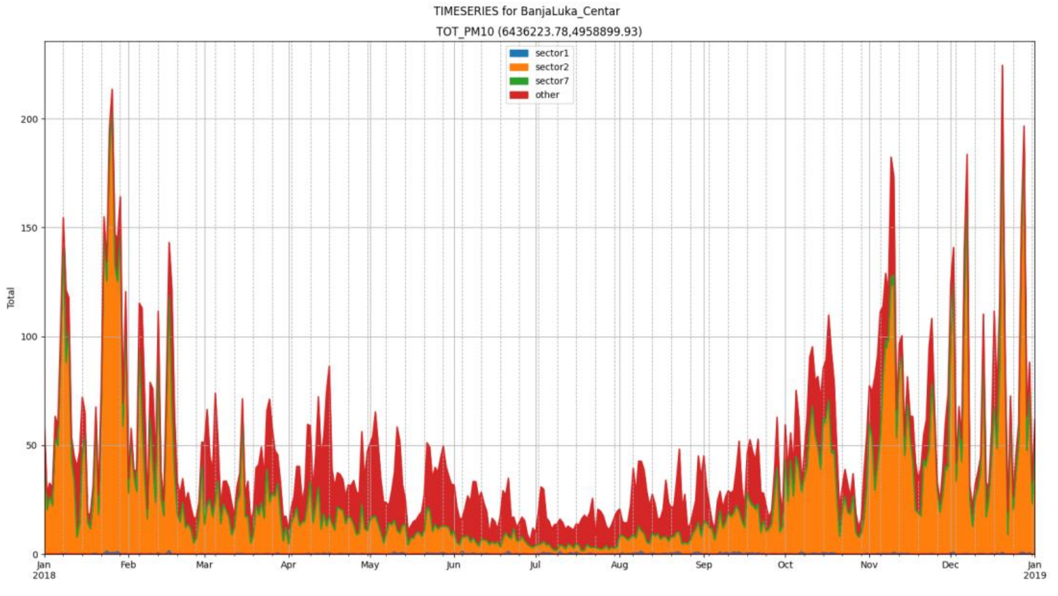


Figure 33: Daily simulated sector contributions to PM_{10} levels of sectors PublicPower (blue sector1), OtherStatComb (orange sector2), Transport (green sector7) and all other sectors including background concentrations (red other) for a grid point in the centre of Banja Luka. The total simulated PM_{10} concentrations are represented by the red stack plot.

Public power and the transport sector contribute very little to the PM_{10} levels daily. Looking at the yearly mean, however, the Transport sector contributes to between 0-4% of the yearly mean levels along roads (Annex B-6 a and d).

For $PM_{2.5}$ (Annex B-8) the pattern is very similar. During winter and autumn months (January, February, November and December) the highest levels were observed (Figure 34). Peak levels varied between 80-120 ug/m3 and were dominated by the domestic heating sector 55-90% (Figure 34). During the rest of the months (March-November) the peak levels were generally half as low, varying between 10-70 ug/m3 and dominated by the other sector category varying in proportion between 50-90%.

On a yearly basis domestic heating stands for ca. 50% of the contributions to the total PM_{10} levels in the areas where levels are high (Annex B-7 a). The transport sector stands for a much smaller portion, ca. 4% and Public Power is much smaller even than that. Other sectors, such as industry and waste make up the rest of the contributions on a yearly basis, also with some influence from the background.

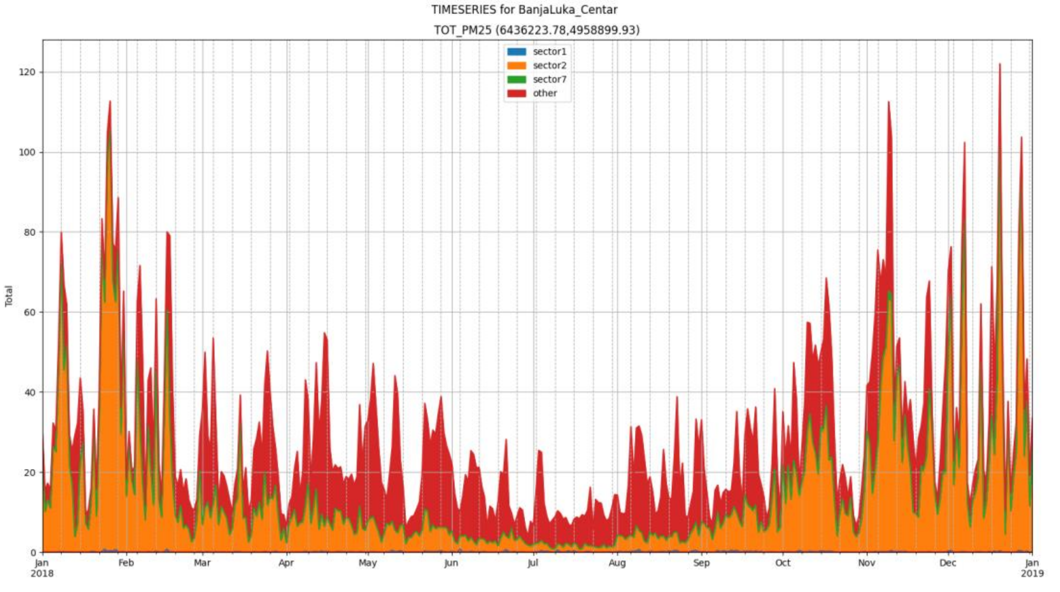


Figure 34: Daily simulated sector contributions to $PM_{2.5}$ levels of sectors PublicPower (blue sector1), OtherStatComb (orange sector2), Transport (green sector7) and all other sectors including background concentrations (red other) for a grid point in the centre of Banja Luka. The total simulated $PM_{2.5}$ concentrations are represented by the red stack plot.

For fine particles, the pattern on a yearly basis is very similar to that of PM_{10} (Annex B-8 a). Domestic heating represented 50% and transport 3% in areas with high $PM_{2.5}$ levels (Annex B-8 c and d), while Public Power was extremely small and insignificant in the current model setup. The other sector category represents the remaining sources where industry and waste are likely the most important contributors.

Uncertainty and future improvements

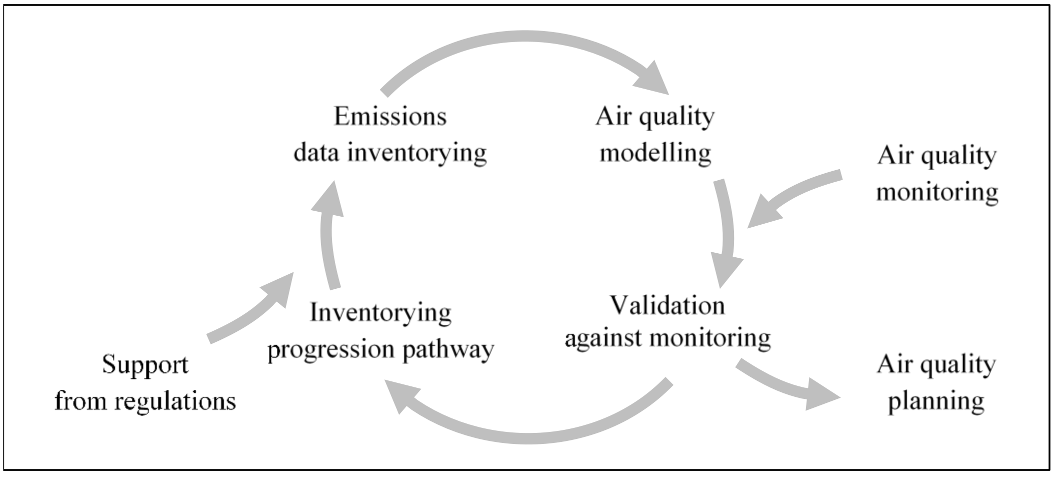
The uncertainties in the model results are, as we see it, mainly driven by three factors: emission totals, overestimated wind speeds and time variations of sectors. The largest improvement would be obtained by improving the emission inventory.

The point sources from industry and district heating need to be better described in terms of the emission composition and amount as well as their variation of activity (eg: district heating is linked to outdoor temperatures, industry working on a 24/7 basis or business hours, complex emissions of some industries and notably the metal industry). That sort of information is considered sensitive by owners and operators. Therefore, organizations in charge of the emission inventorying must have a clear mandate from the government to do so, and the owners clear instructions to share that data for a dedicated purpose.

Depending on the location, the emission inventorying effort already done is essential and very important. The current emission inventory will however gain from continuous updating of inventorying methods with more information on locations, emission factors and time variation of the related activities of the sources.
More information about fuel types used in residential heating is often required as well as a better information about appliances and their fuel consumption.
Thanks to the vehicles registration system the vehicle fleet is known but data of the annual mileage would be an improvement. Additionally, the removal of catalyser and particle filters from vehicles is often cited as a common problem in BiH which makes it difficult to accurately estimate the traffic.

The centralization of emission inventorying could be of a great help in order to enhance and accelerate the circle assessment of a good air quality modelling capacity:

Emission data inventorying à air quality modelling à validation against air quality monitoring à identification of progression pathway for inventorying.



Running the emissions data as they are through a dispersion model shines some light on how well the simulated pollution levels relate to measured pollution levels. In the current emission inventory, information was provided for three sectors. These are believed to be sometime underestimated, and some important points sources are missing. More information from other sectors is also necessary for a more detailed model simulation result.

Wind speeds on a day to day basis were on average 0.5 m/s higher than observed wind speeds. This can have an effect on the modelled concentration levels. If a better wind speed data can be obtained the effect of lower wind speeds on the modelling result can be tested by running a new simulation with updated wind speed input data.

The standard time variations of sectors built into the MATCH model are based on information about emission sources located in north-western Europe. This might be improved in BiH for e.g. traffic may have a slightly different monthly variations. The daily time variation of traffic may also be adjusted to the BiH habits. Furthermore, time-variation could be further developed to include consideration to the ambient temperature. When temperatures are very low, emissions from residential heating may be even higher than described in the current time-variations for this sector. In order to make such changes more information about the intensity of emission sources during cold temperatures in BiH is needed. Further experimenting with and testing of different time-variations and simulation tests are also needed.

Concluding summary on the MATCH study

In this study we have simulated the air pollution levels of NO_2, SO_2, PM_{10} and $PM_{2.5}$ in Sarajevo and Banja Luka using high resolution emission data for three sectors; Public Power (large scale heating facilities and some industrial facilities), Other stationary combustion (individual domestic heating sources) and Transport (on-road traffic) and coarse resolution emission data from CAMS for other sectors (e.g. Industry, Fugitives, Waste and Agriculture). We have also estimated the source contributions for the three sectors with high resolution emission data.

Overall, the study has shown that the air pollution load in both Sarajevo and Banja Luka, is to a large degree dominated by local emission sources. A seasonal variation of pollution levels was obvious for all modelled air pollutants. Concentrations were generally low in the summer half of the year and much higher in the winter due to frequent calm and stable weather conditions combined with high emissions related to heating. This was further supported by the model's capacity to capture several days in the winter with low boundary layer height, signifying a high potential for inversions. There was also significant transport of air pollutants (SO_2 and particles) from outside the cities, especially during the winter.

The simulated source apportionment suggest that the transport sector dominated the NO_2 levels, while individual residential heating dominated particle levels. This was obvious in both Sarajevo and Banja Luka. Thus, emission reduction strategies targeted at these sectors would most likely improve the air quality situation in both cities. For SO_2, domestic heating contributed to a large degree, but a large proportion of the modelled concentrations also originated from the other sector categories, mainly industry. The emission data of the industry sector came mainly from CAMS and the totals of this dataset are considerable but cover a coarser resolution grid than the local emissions. Thus, obtaining more information about the specific industrial sources and recalculating their emissions with a higher spatial resolution is necessary to pinpoint the locations of the sources included in this data set and to further reveal any concentration gradients over a finer grid within the model domain. With a coarse grid resolution concentration, gradients are difficult to determine. This also makes it difficult to determine which areas are significantly impacted by which emission sources.

Another reason to further review SO_2 emissions is the fact that they were surprisingly low in Banja Luka. For particles, the waste and agriculture sectors had significant emission totals for particles originating from the CAMS emission data, highlighting once again the importance of reviewing coarse emission data and obtaining more detailed information about the emission totals of the specific sources within these sectors.

Underestimations are mainly explained by low emissions in general (probably large underestimates), and partly by slight overestimation of wind speeds, and time variations (SO_2 and NO_2 in the summer were not captured well, requiring more info on the time variation of emissions). Quite a large discrepancy was observed between measured and modelled concentrations of SO_2 in the summer in Sarajevo, where the model underestimated peak levels by a factor of between 2 and 3 between June-October. This can either be explained by a missing local source in the emission inventory data or a large influence from sources outside of the model domain. For the model to capture this, further work with the emission data both locally and in surrounding regions would be necessary. Furthermore, for particles, the model underestimates the observed levels by 50% or less, except for some peaks. Overestimated wind speeds in the HCLIM input data is likely contributing to this underestimation thus better meteorological input data may also result in a better correlation between modelled and observed levels. A dispersion model run at 500m horizontal resolution will not produce exactly the same concentrations as a monitoring station placed close to emission sources, but will rather produce slightly lower concentrations, even when using very accurate emissions data and meteorological data.

Once emission inventories of all source sectors are of good quality and can describe the current situation in more detail, the results from the dispersion modelling can be further applied in scenario analysis. Emission reduction scenarios can be computed and simulated in the model and thus give city planners a powerful tool to test whether a certain, or a combination of several emission reductions strategies, are likely to result in an intended air quality improvement. Furthermore, the dispersion model can also be used as a substitute in locations where measurements are non-existent. It's quite a common practice to use a dispersion model in the initial stages of mapping and assessing the air quality situation over a broad geographical area.

Sustaining and expanding the air pollution monitoring network to cover urban, sub-urban and rural areas are important. A larger monitoring network would be beneficial for gathering data from different types of locations in the cities. Some locations may have quite specific local features and emission sources which affect the air pollution load in an area in a unique way. Such locations do not represent the general air pollution situation, having several monitoring stations will thus show the variation between locations and help with illustrating the overall air pollution situation in the cities. A larger monitoring network can then also be used to validate model results and thus also help improve the whole model setup.

In order to achieve a complete picture of the link between emission sources and the air-quality situation in any of the two cities studied, a combination of continued emission inventorying, dispersion modelling and comparison with monitoring data is needed. The work that has taken place so far in BiH by local authorities and consultancy companies has provided important steps to achieve this goal.

It is clear that the air quality is the worst in both cities when the major air pollutant emitters – individual and district heating and traffic – adds up during wintertime. The situation is made even worse during the periods of temperatures inversion that delay the dispersion of the pollutants and lead instead to their concentrations. It would be useful to focus the mitigation strategies on the abatement of the emissions of NOx and particles from both residential heating and traffic. This could mean creating subsidies for cleaner fuel for heating, effective burning appliances, isolation enhancement, and district heating extension. Further actions that could be considered, include improving the vehicles technical inspections and finding feasible ways to both reduce people's dependency on cars for mobility and to promote the modernisation of the fleets of vehicles. This would provide enhanced benefits if conducted in synergy with the promotion of clean public transport, biking and pedestrian paths.

It would also be very useful to improve the monitoring of temperature inversions in order to be able to disseminate awareness toward sensitive groups of peoples during these periods. This would be the simplest way to develop a local prediction of upcoming alarm levels for pollutants in the ambient air. A continued effort towards maintaining the meteorological stations in and around the cities is also important to understand how the weather situations affect the air quality in general, as well as providing important input and validation data for meteorological models used for dispersion modelling.

Better knowledge of the emissions that are specific to the BiH would benefit urban and air quality planning in the near future. Maintaining this knowledge and understanding of air pollution drivers would allow authorities to accurately inform communities of their ongoing and continued commitments for improving the environment. It could also help authorities to prioritize future initiatives or actions to be undertaken in air quality action plans. In addition, it could contribute to the monitoring of the efficiency of former policies. Finally, sustaining such an effort in the air pollution investigations could contribute to maintaining a community of informed people and experts, which is critical for making good decisions and accepting relevant recommendations.

CONCLUDING REMARKS ON BOTH STUDIES IN THIS REPORT (PMF RECEPTOR MODELLING AND MATCH)

This source apportionment study has addressed source apportionment from several different perspectives. Receptor modelling of PM2.5 measurements in the six cities provided a more basic analysis of where PM pollution originates in the selected cities. The second round of PM2.5 measurements and receptor modelling provided more detailed information regarding sources of pollution in Banja Luka and Sarajevo. The final round of analysis, the dispersion modelling, focused not only on PM pollution but also other sources of pollution such as NO_2 and SO_2. That analysis was based on emissions inventories, and the data was compared with data from air quality measuring stations to verify its accuracy.

This study is unique in that it analyzes source apportionment based on several modelling techniques and is based on multiple different types of data sets. While there are some overarching conclusions that can be made based on the results of this study, for example that traffic and fossil fuel combustion are the primary sources of PM pollution in Banja Luka and Sarajevo, due to the complexity of the study, different approaches yielded slightly different results. The differences in the results should be viewed as areas where there needs to be further investigation. For example, there were several instances where output from the MATCH model did not completely align with air quality measurements. This indicates that more detailed emissions data should be gathered. Similarly, the PM2.5 measurements found instances of unexplained potassium-rich PM2.5 in Banja Luka and cadmium-rich PM2.5 in Sarajevo that are difficult to explain and are not represented in the emissions data for these cities. This is also an indication that emissions inventories should be improved.

In addition to highlighting places where more detailed data collection is needed, this study has provided diverse information for policy makers. For example, the source apportionment analysis can be useful for policy makers looking to create emissions reduction programs. Furthermore, dispersion modelling offers insight into how air pollution moves within Banja Luka and Sarajevo. The output of the dispersion modelling can be used, for example, to create forecasts, develop action plans, and inform city planning.

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APPENDICES

Appendix 1

The following pie-charts, similar to the one in Figure 35, show two separated information for each sampling place.

- The first pie-chart shows the apportionment of the sources, or group of sources, that explain the measured weight of PM 2.5 during the sampling period. The colours of the named sources are the same in a page, but differ from one page to another.
- The other pie-charts show the distribution of each measured species weight within the different apportioned sources, or group of sources (see the colours of the first pie-chart for names). 0% is displayed when the contribution from a factor is under 1%.

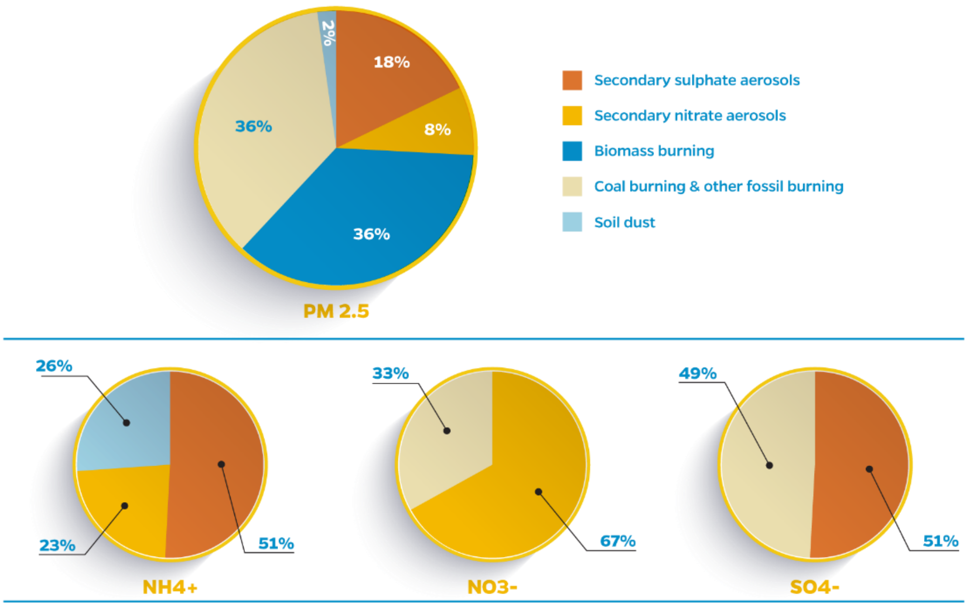
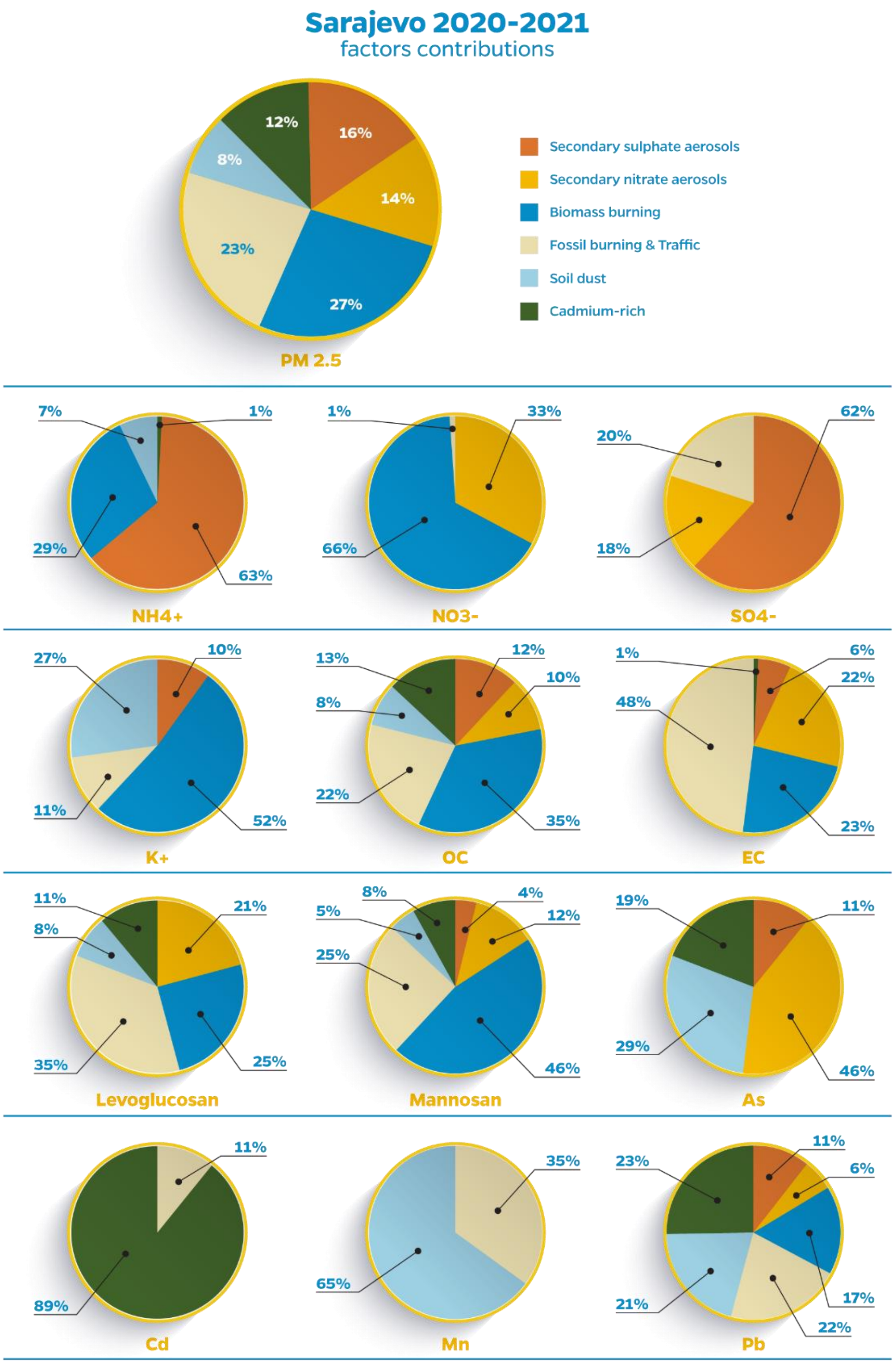


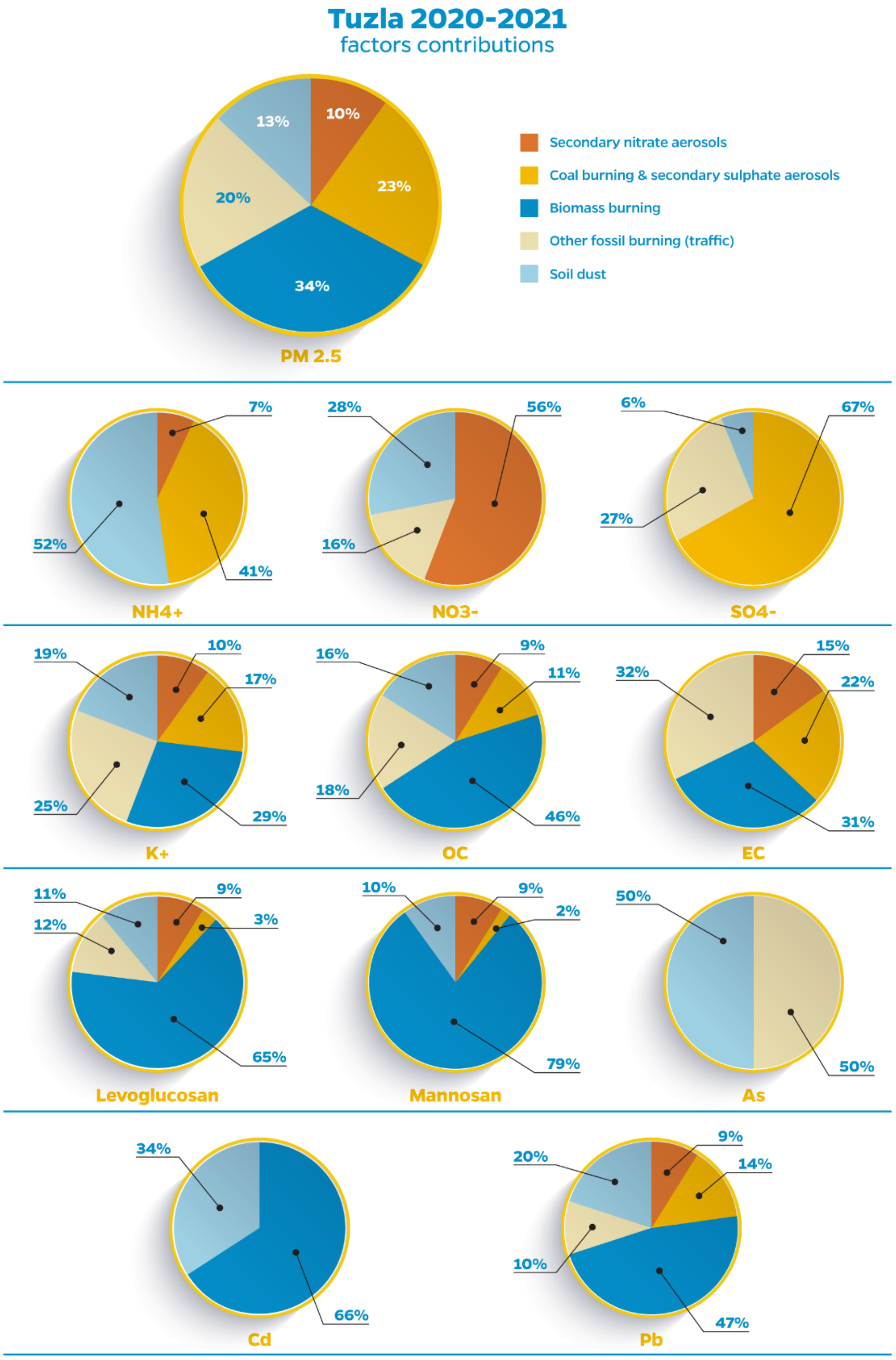
Figure 35: Pie chart in appendixes

This information is already shown by the black square-dots in the graphs displaying the composition of factors for each sampling places. But it is gathered by species here. Some species trend to be tightly related to only a few sources whereas some species are less specific.

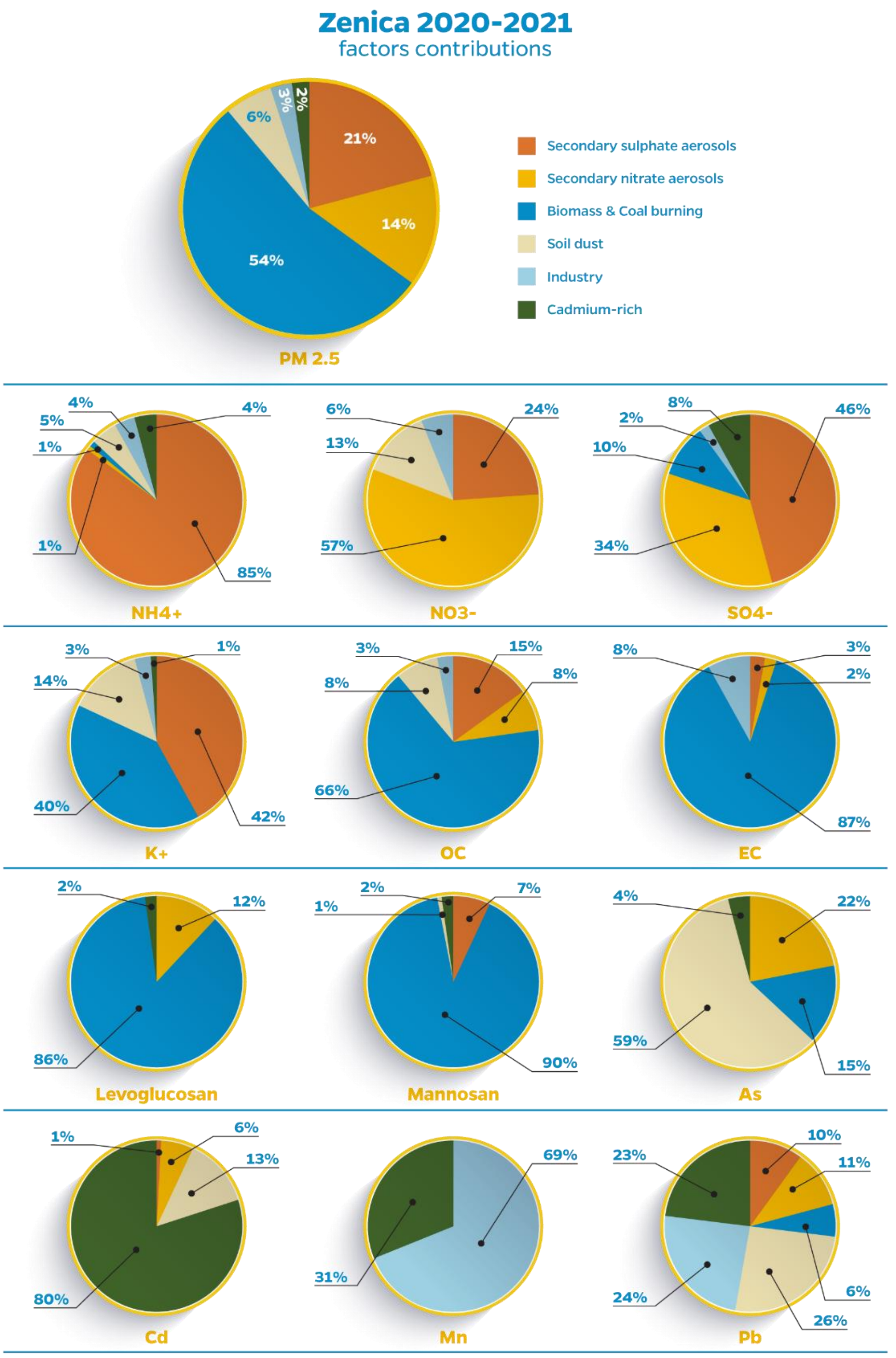
Sarajevo 2020-2021 – factors contributions



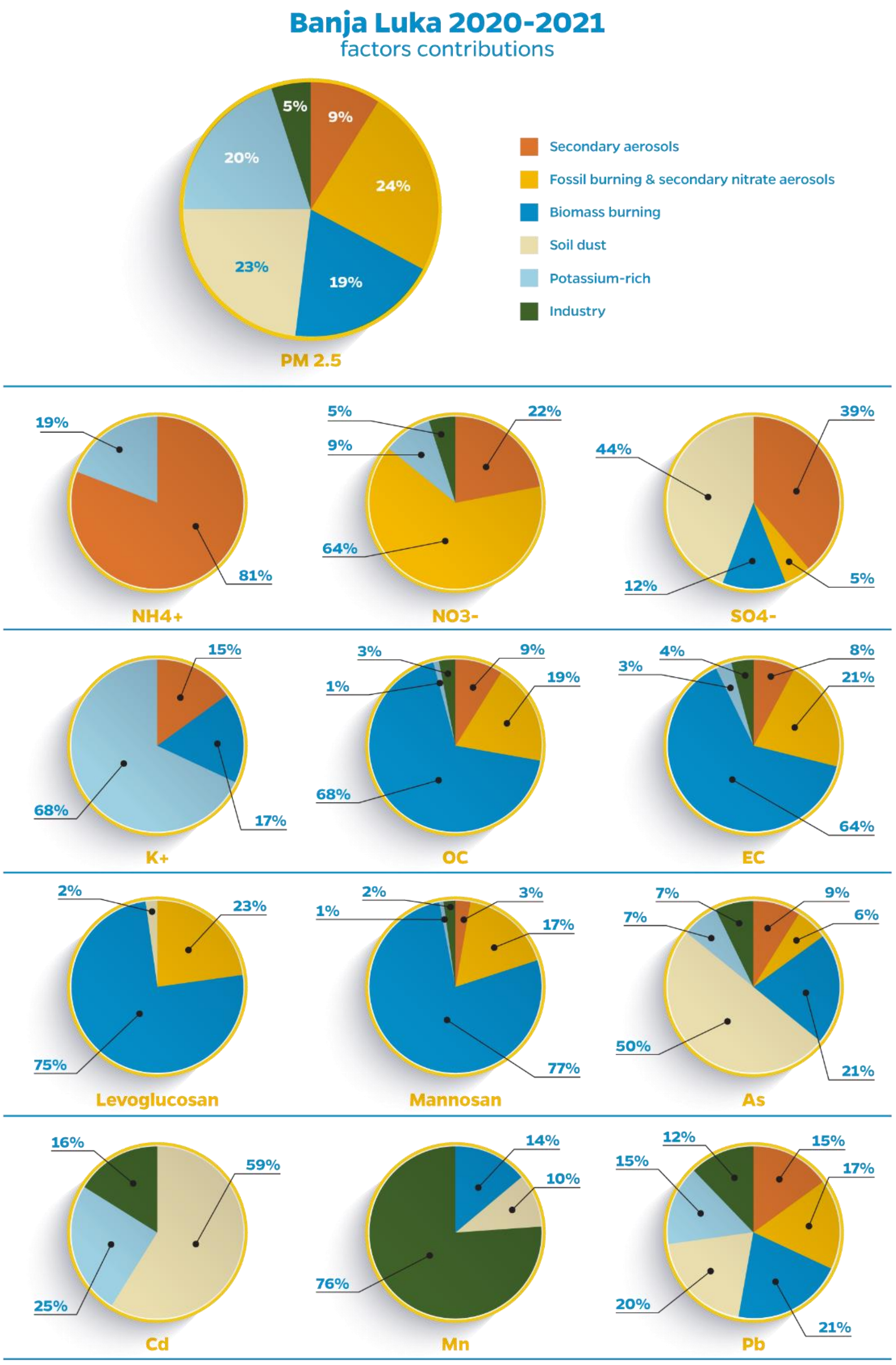
Tuzla 2020-2021 – factors contributions



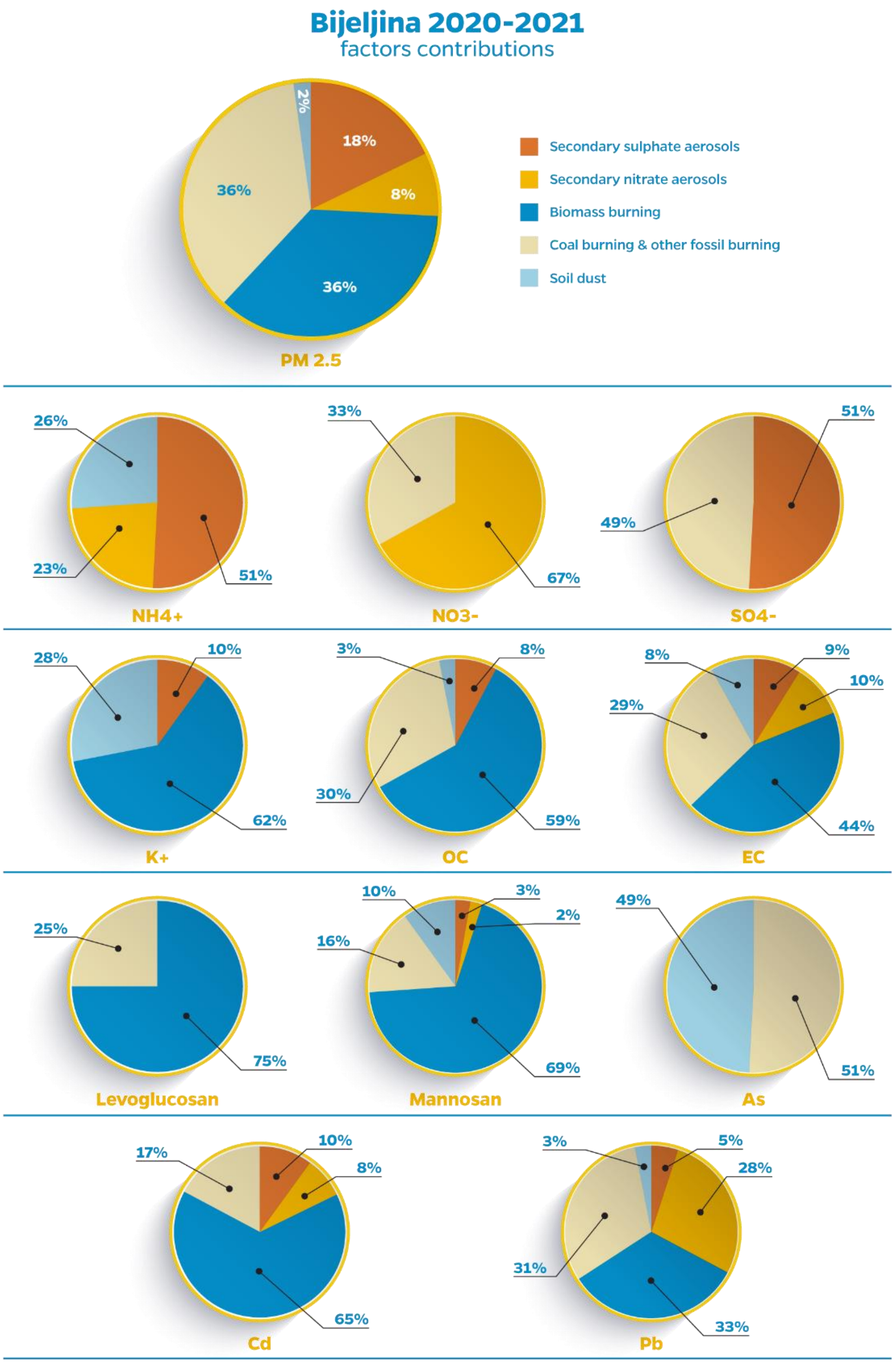
Zenica 2020-2021 – factors contributions



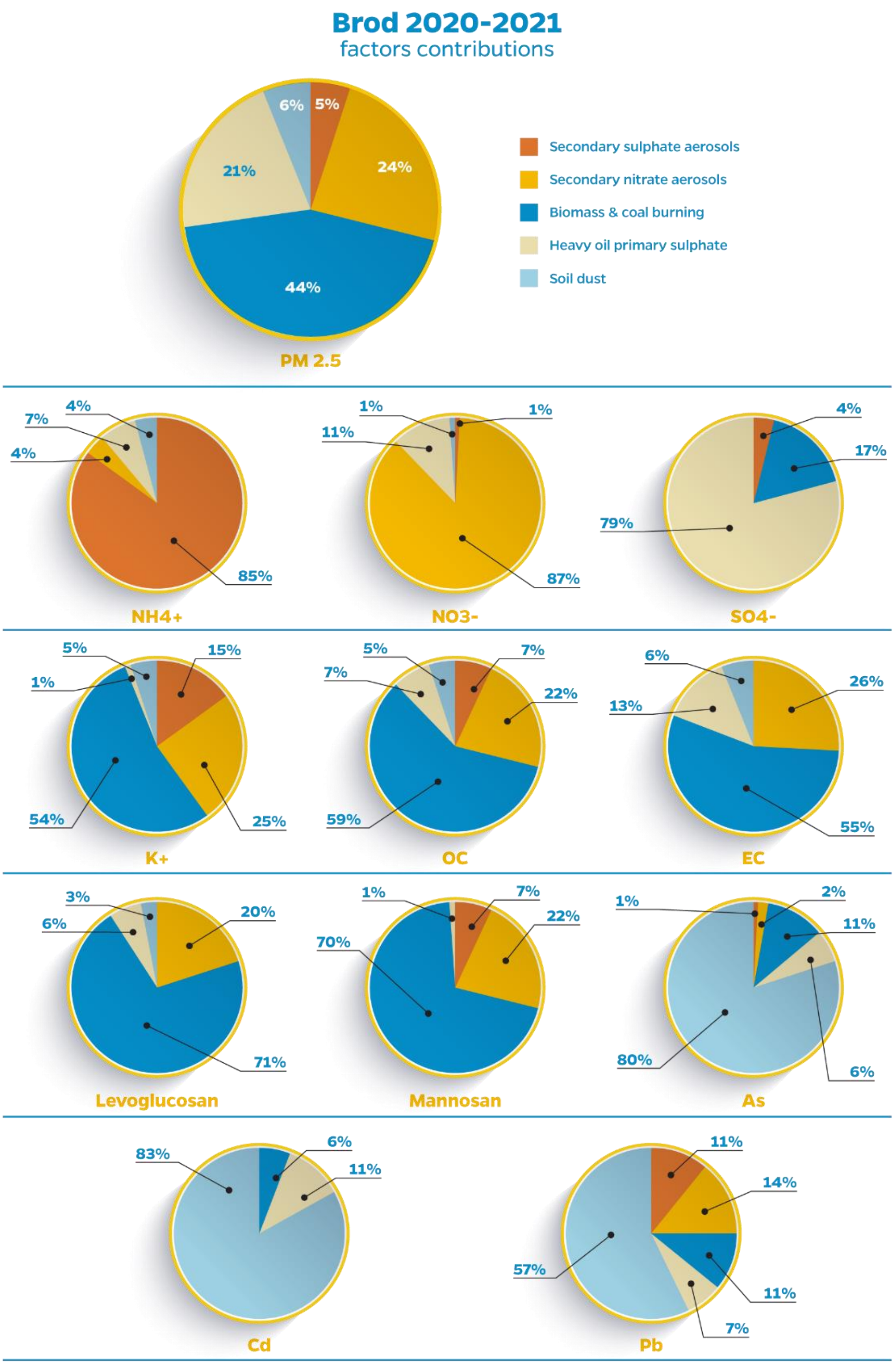
Banja Luka 2020-2021 – factors contributions



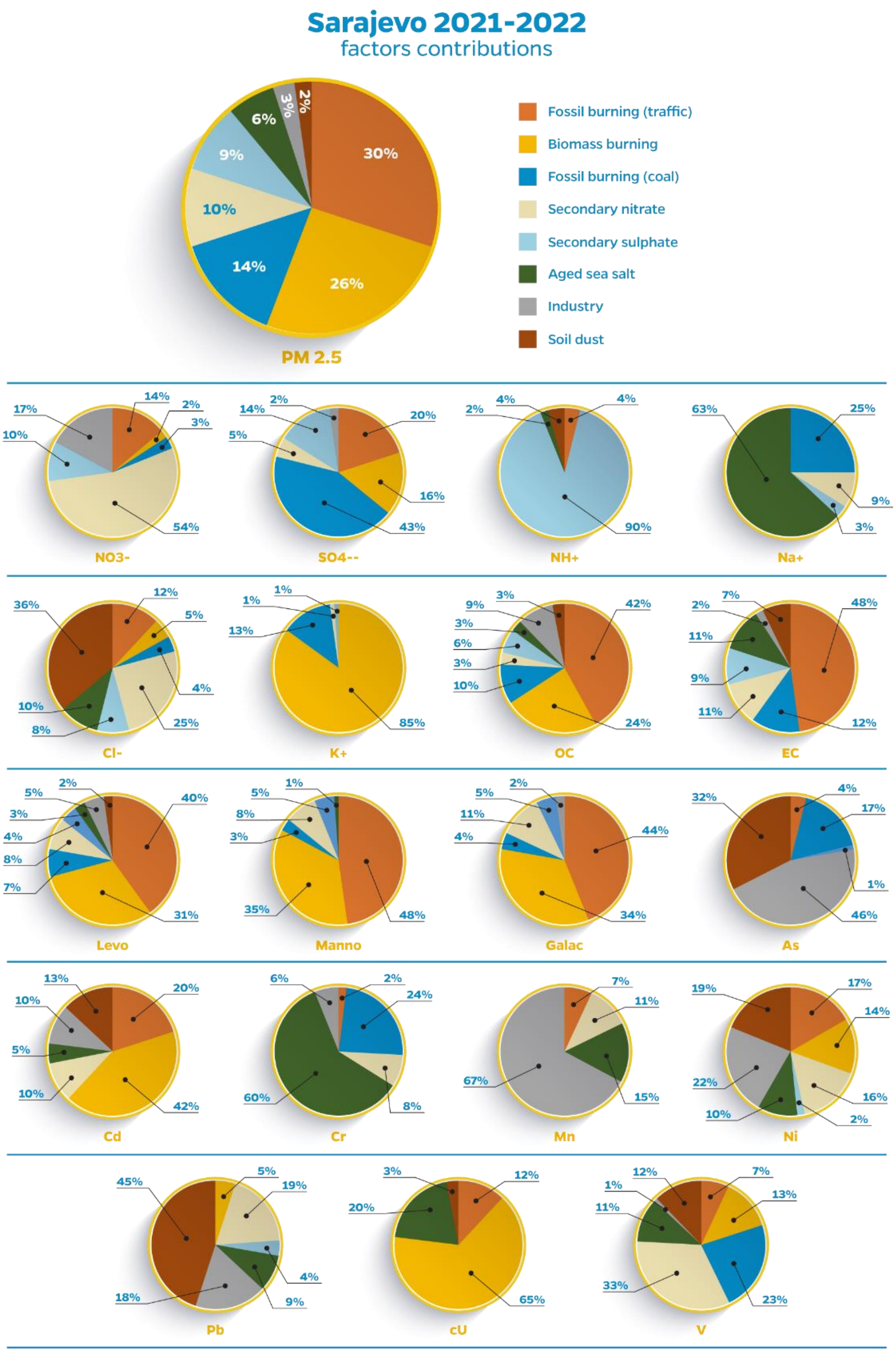
Bijeljina 2020-2021 – factors contributions



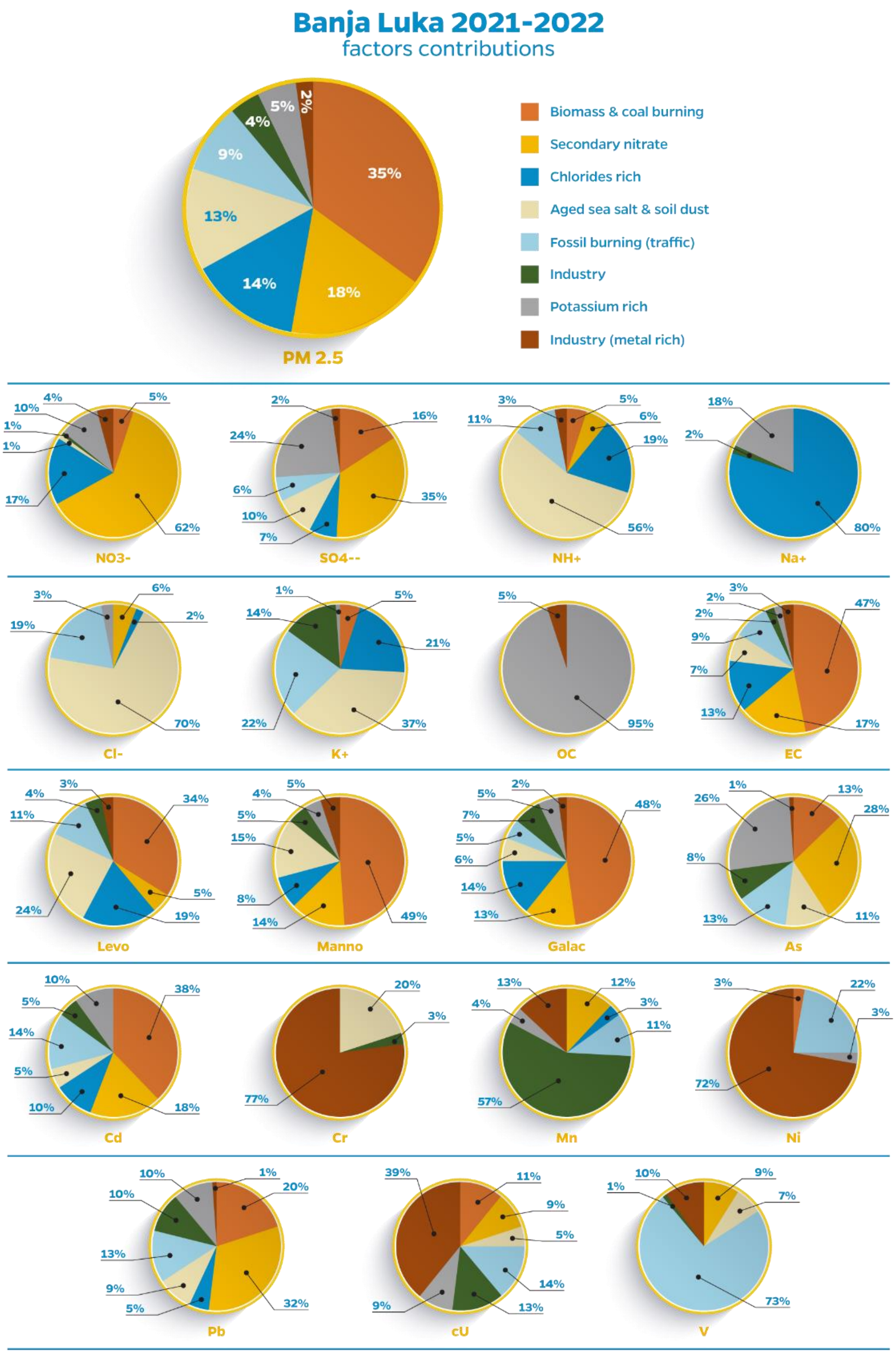
Brod 2020-2021 – factors contributions



Sarajevo 2021-2022 – factors contributions



Banja Luka 2021-2022 – factors contributions



Appendix 2 – Sarajevo, dispersion and source apportionment maps

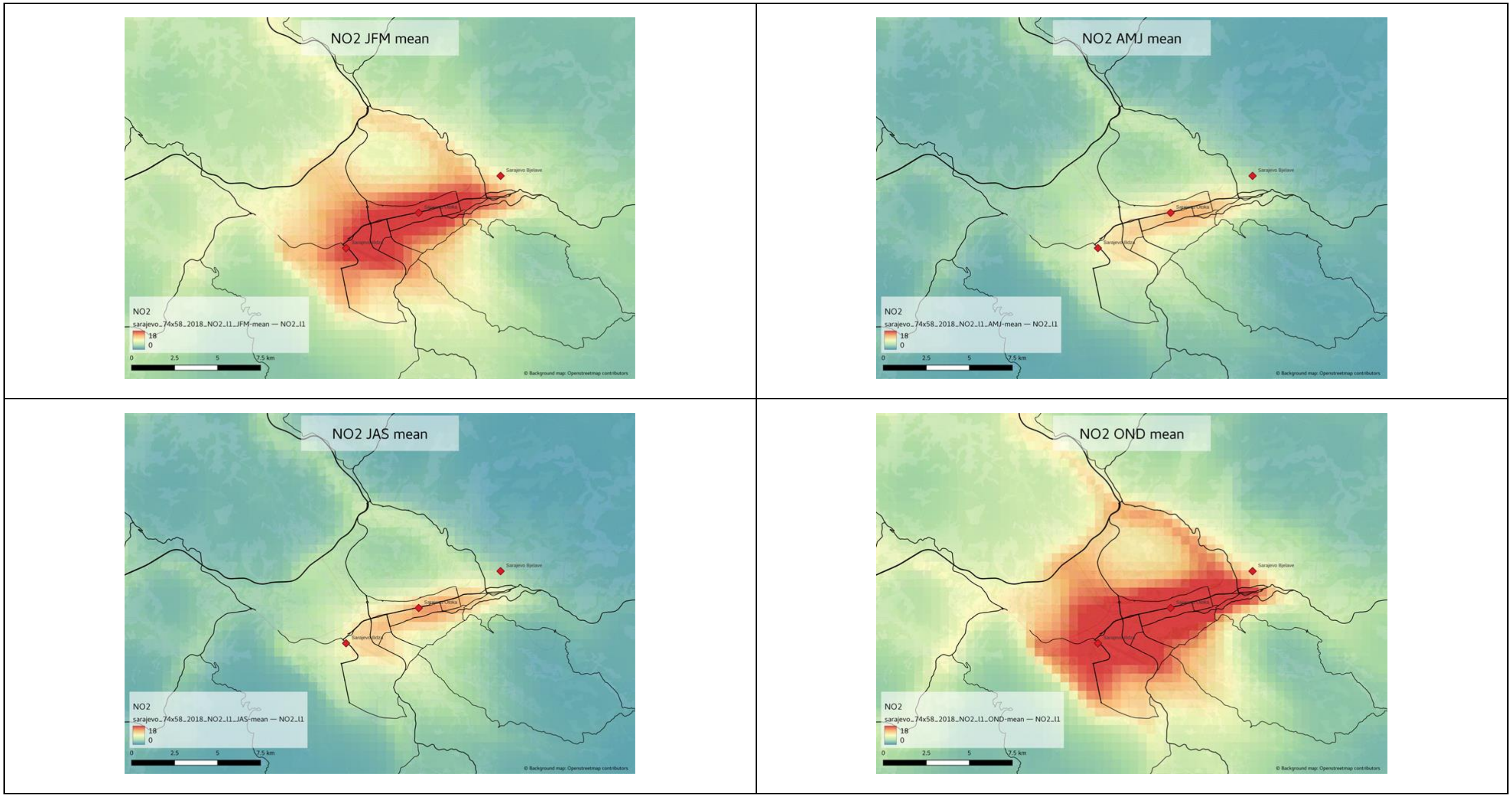


Figure A-1. Spatial maps of modelled seasonal NO2 averages during; a) January-March (JFM), b) April-June (AMJ), c) July-September (JAS) and d) October-December (OND) in Sarajevo, 2018. Units are in ug/m3.

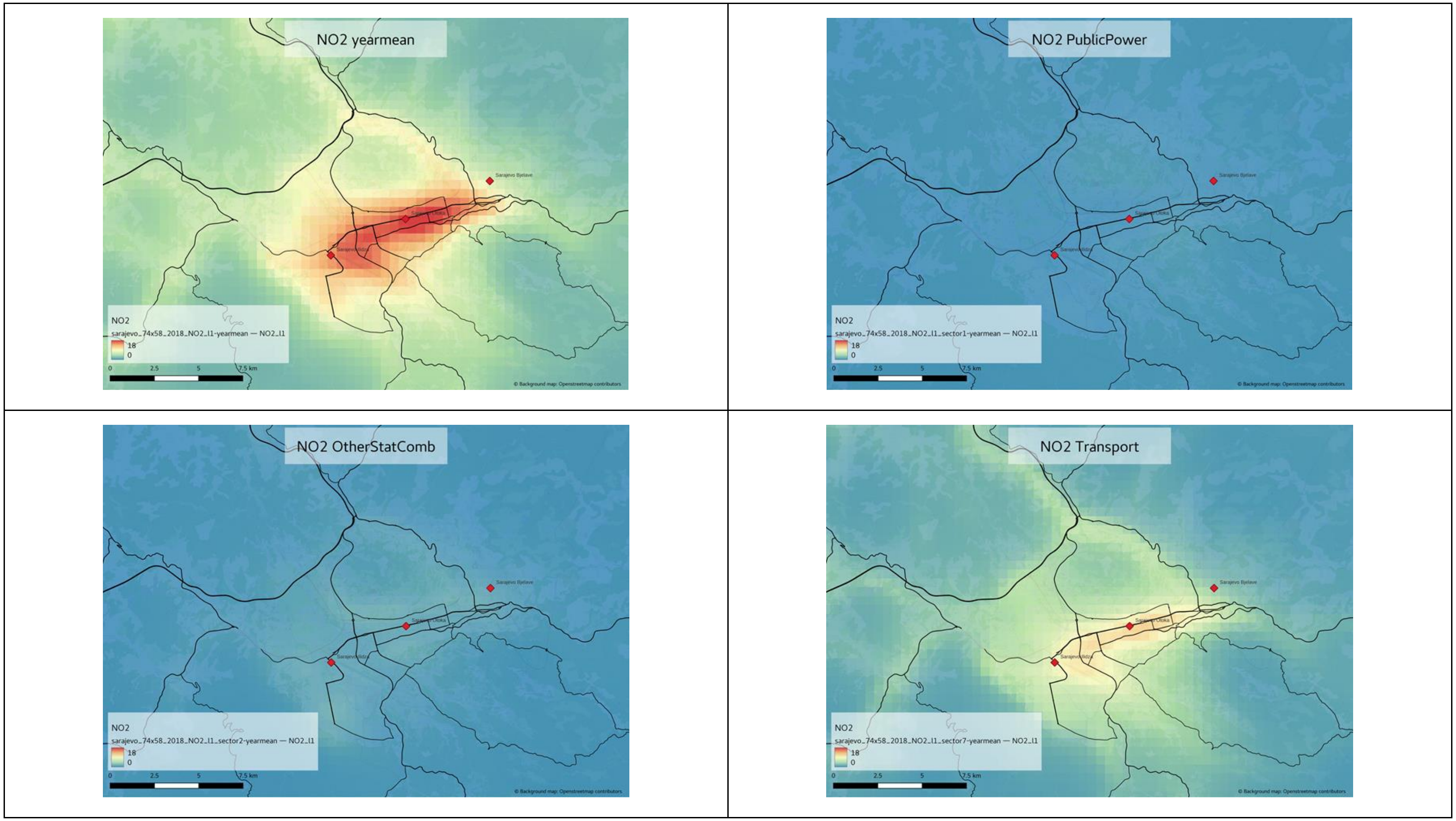


Figure A-2. Spatial maps of modelled NO2 concentrations in Sarajevo, 2018 for a) the total yearly mean based on all available sector emissions within the model domain, including background concentrations from the boundary. Source apportionment of NO2 concentrations for three different sectors calculated with the new high resolution dataset of local emission from b) PublicPower, c) OtherStatComb and d) Transport

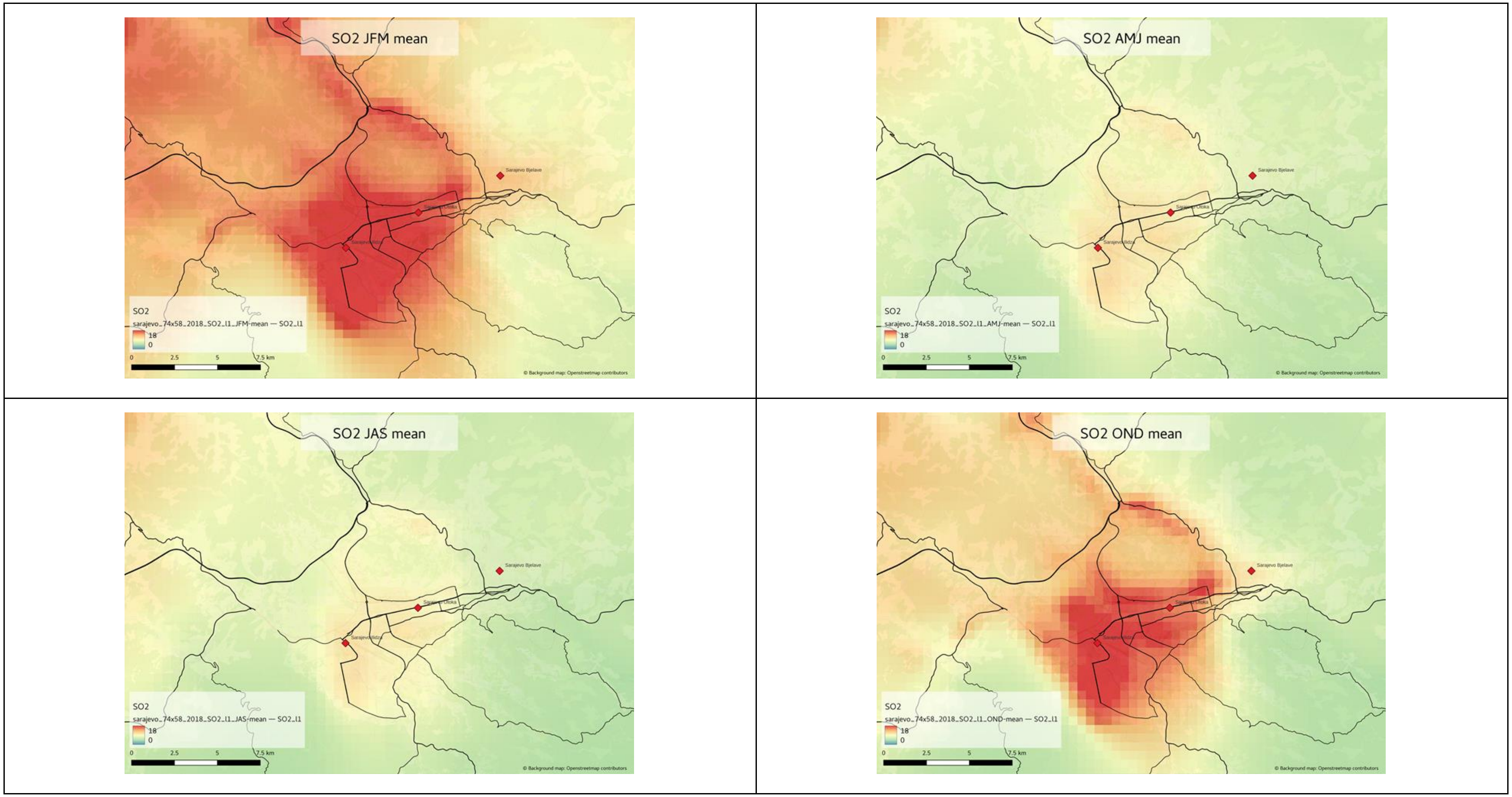


Figure A-3. Spatial maps of modelled seasonal SO2 averages during; a) January-March (JFM), b) April-June (AMJ), c) July-September (JAS) and d) October-December (OND) in Sarajevo, 2018. Units are in ug/m^3

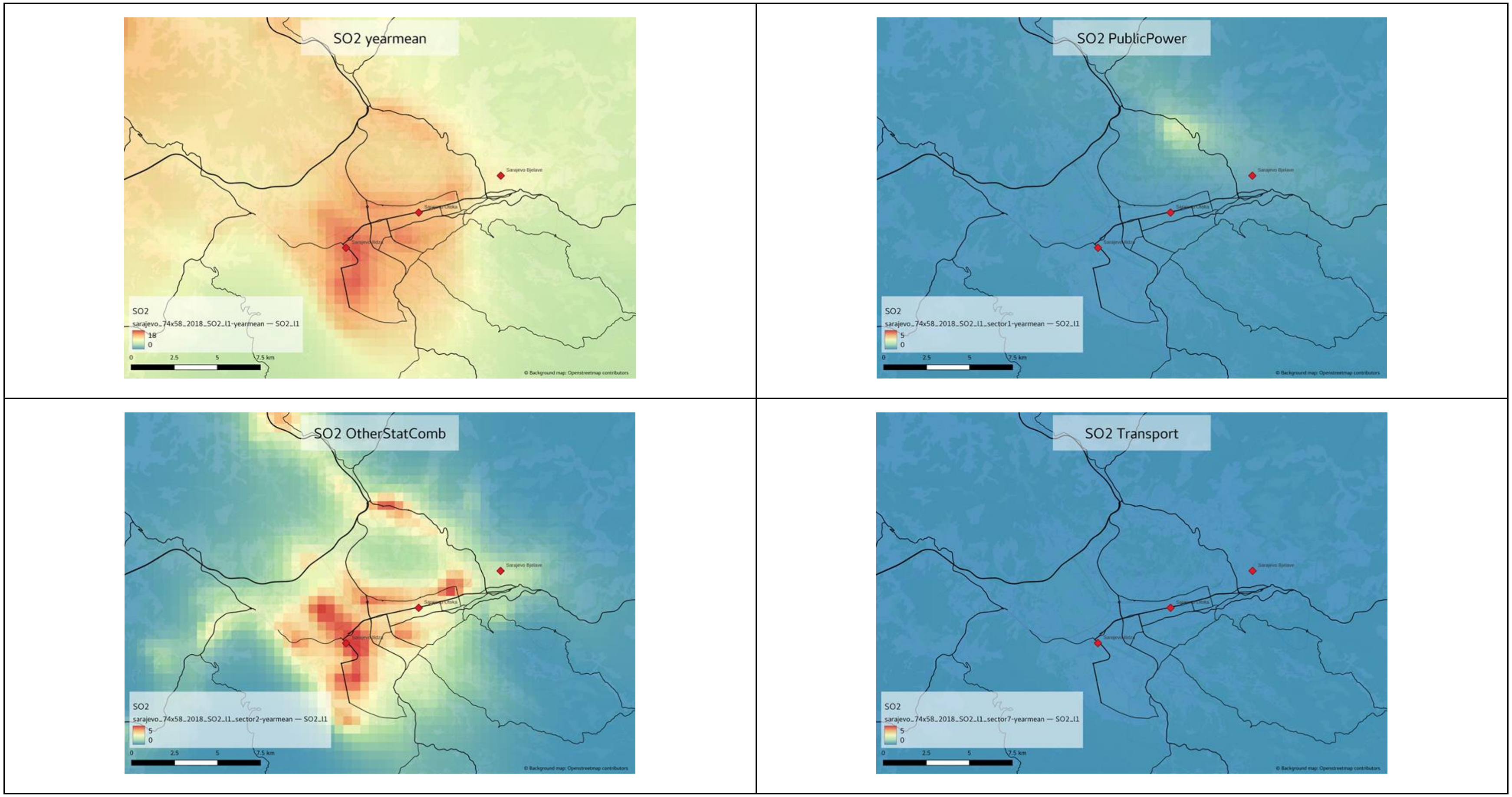


Figure A-4. Spatial maps of modelled SO2 concentrations in Sarajevo, 2018, for a) the total yearly mean based on all available sector emissions within the model domain, including background concentrations from the boundary. Source apportionment of SO2 concentrations for three different sectors calculated with the new high resolution dataset of local emission from b) PublicPower, c) OtherStatComb and d) Transport

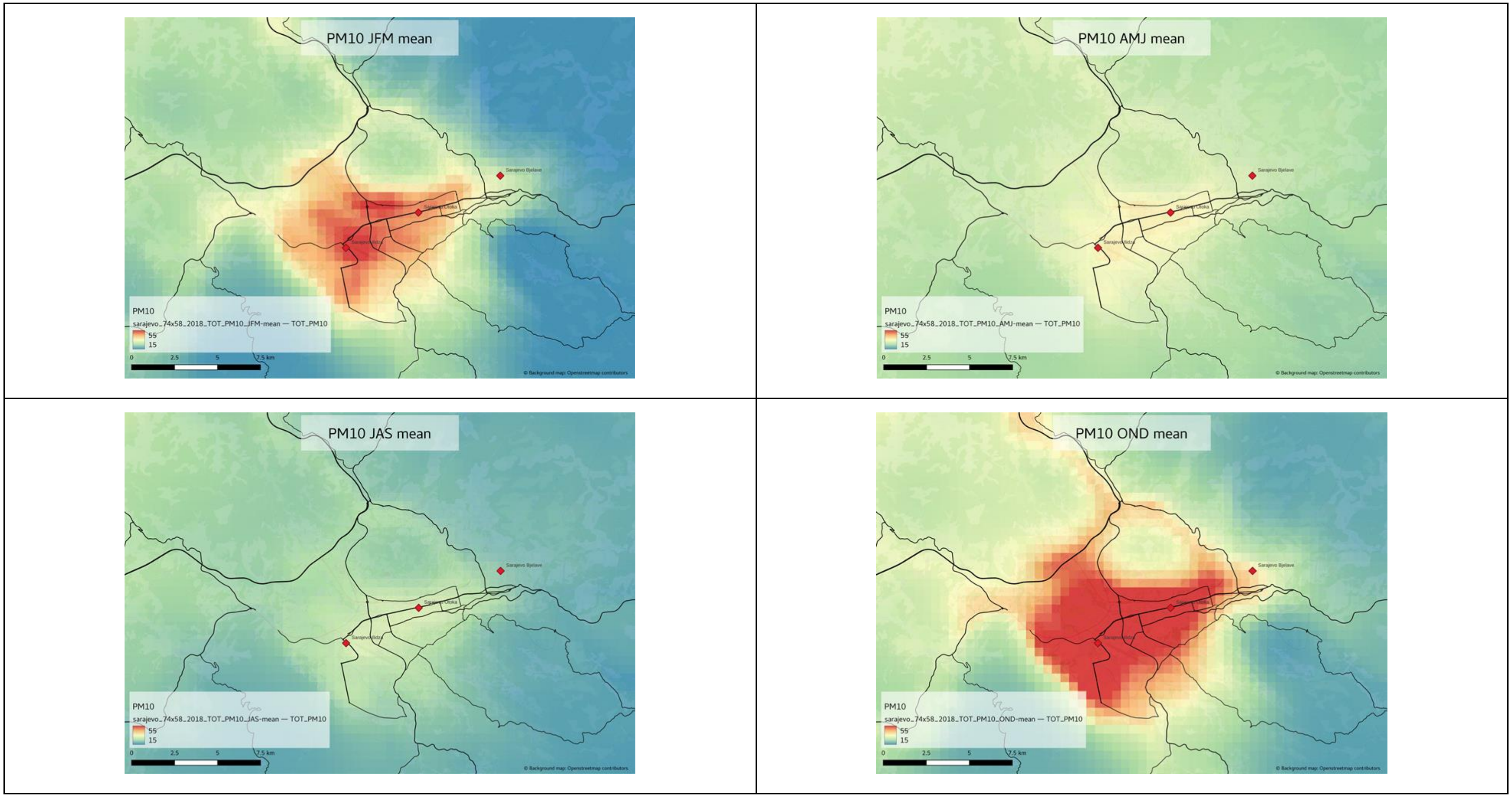


Figure A-5. Spatial maps of modelled seasonal PM10 averages during; a) January-March (JFM), b) April-June (AMJ), c) July-September (JAS) and d) October-December (OND) in Sarajevo, 2018. Units are in ug/m^3

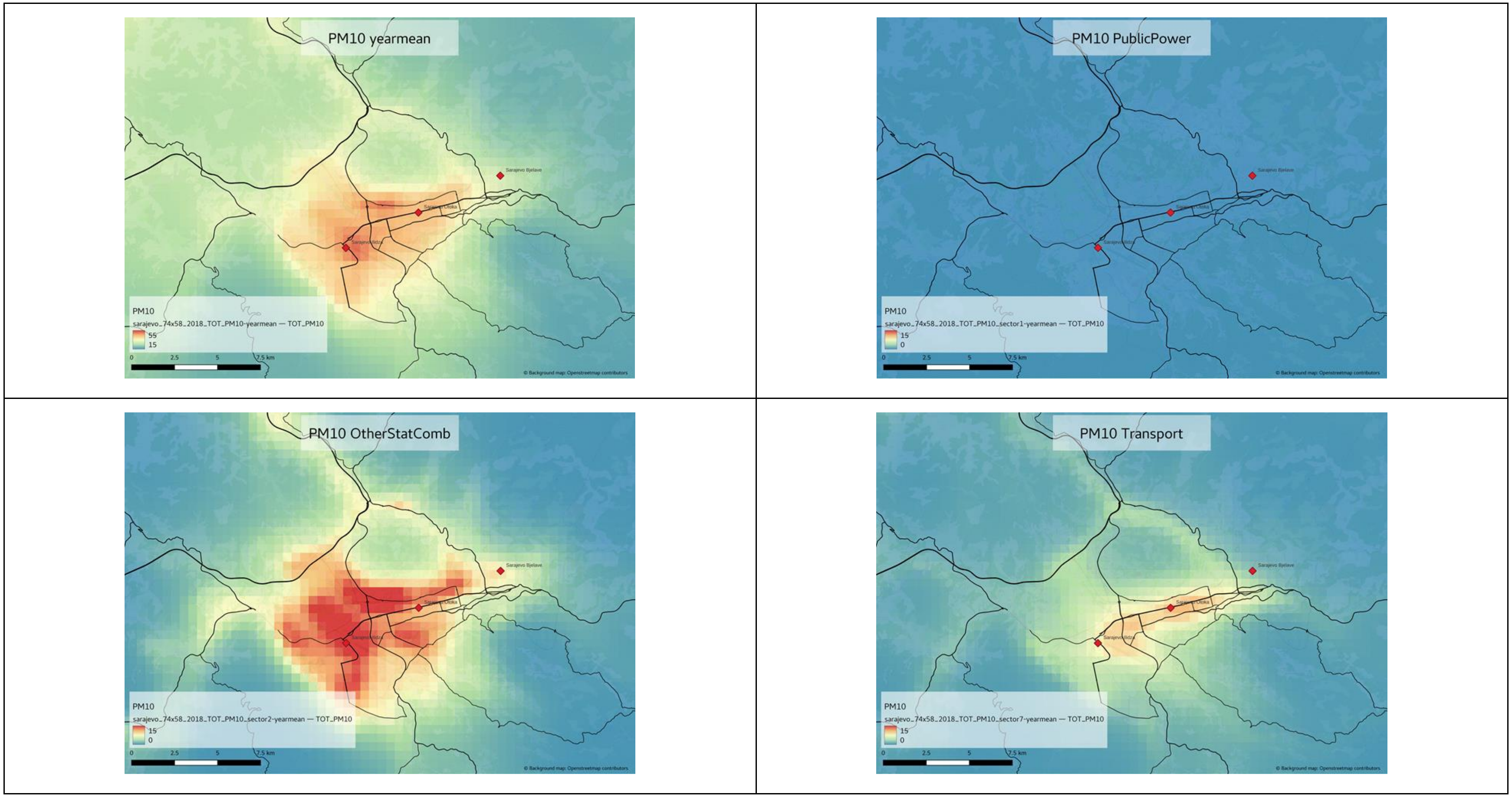


Figure A-6. Spatial maps of modelled PM10 concentrations in Sarajevo, 2018, for a) the total yearly mean based on all available sector emissions within the model domain, including background concentrations from the boundary. Source apportionment of PM10 concentrations for three different sectors calculated with the new high resolution dataset of local emission from b) PublicPower, c) OtherStatComb and d) Transport

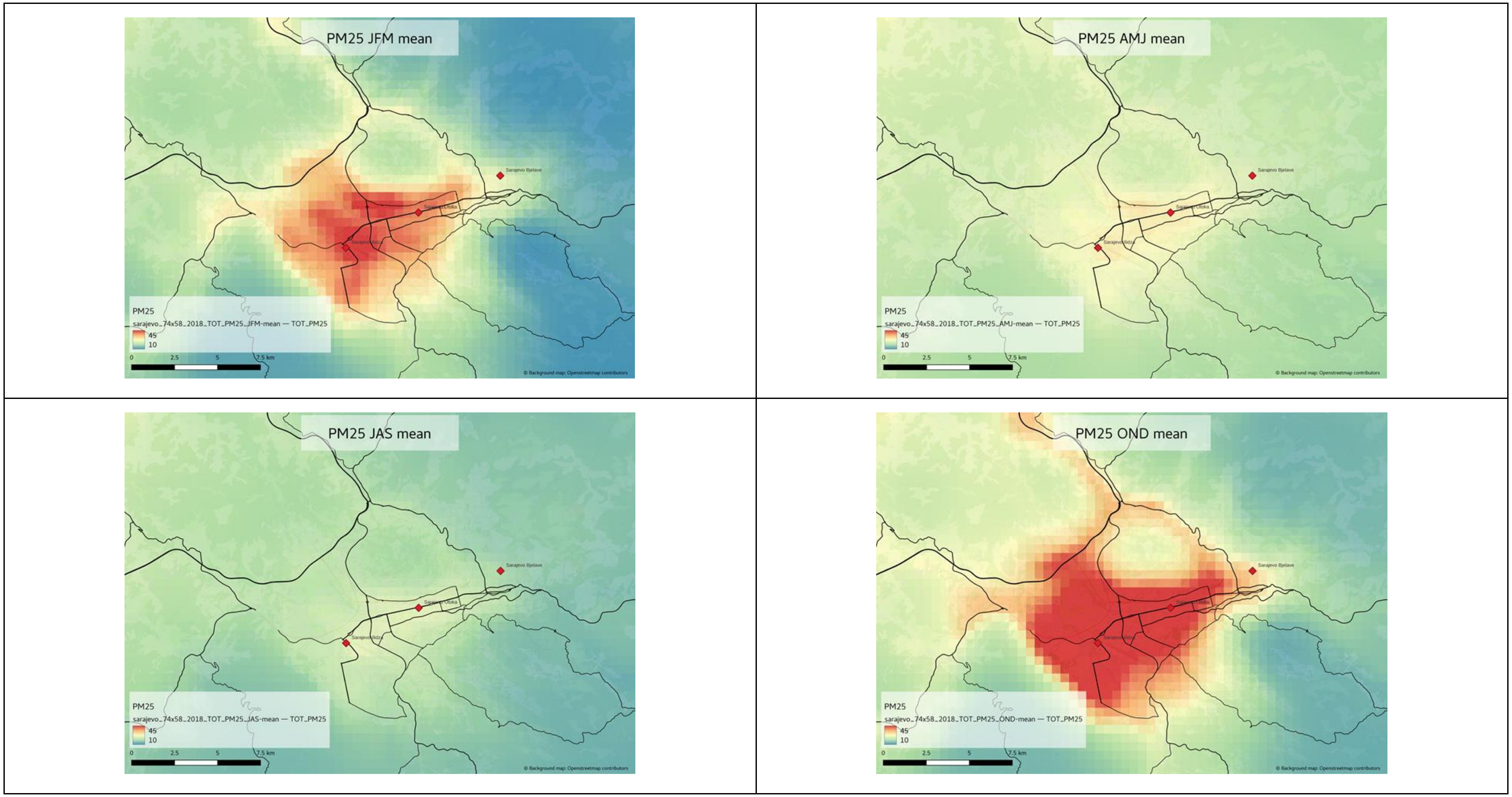


Figure A-7. Spatial maps of modelled seasonal PM2.5 averages during; a) January-March (JFM), b) April-June (AMJ), c) July-September (JAS) and d) October-December (OND) in Sarajevo, 2018. Units are in ug/m3

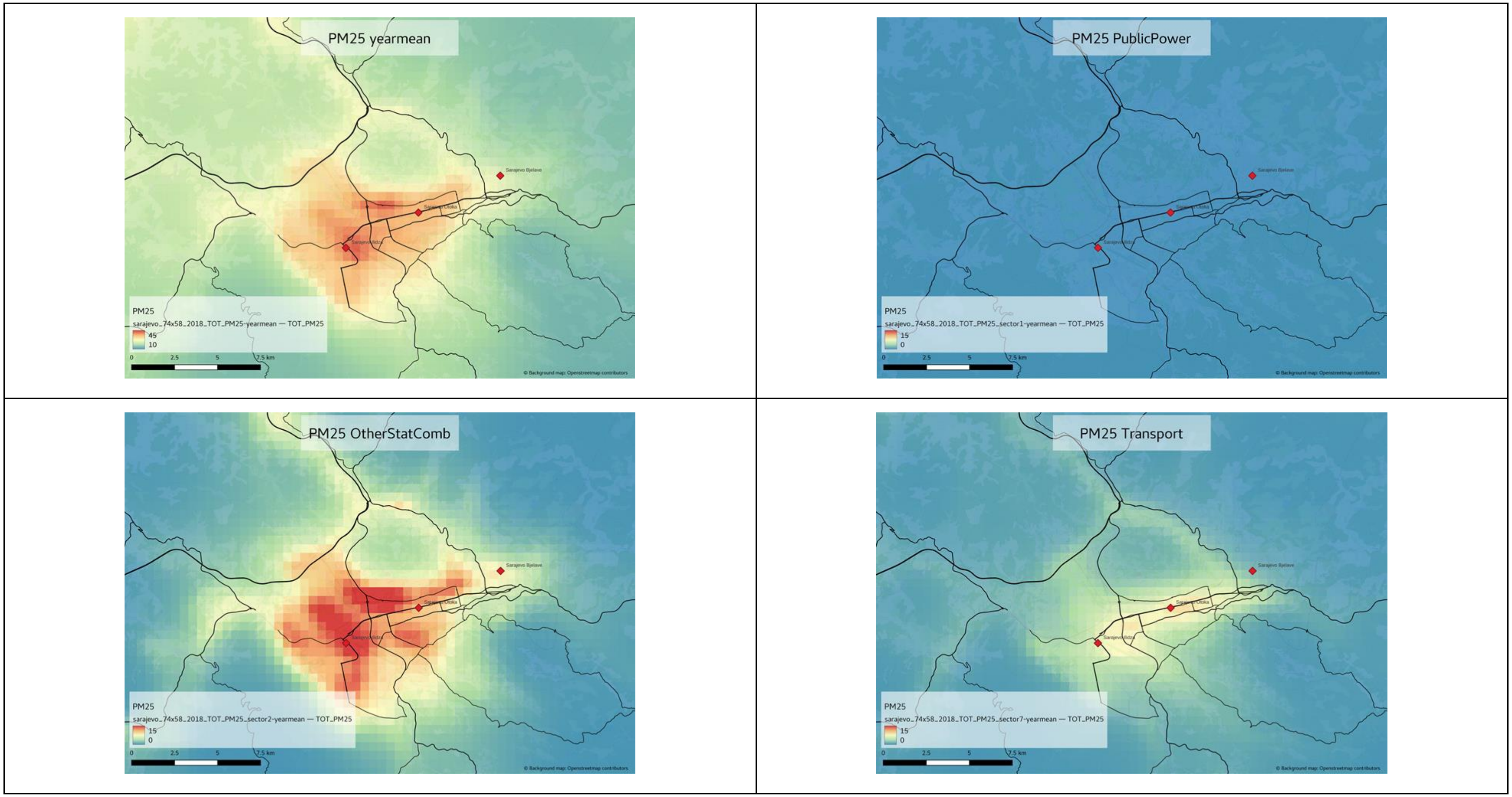


Figure A-8. Spatial maps of modelled PM2.5 concentrations in Sarajevo, 2018, for a) the total yearly mean based on all available sector emissions within the model domain, including background concentrations from the boundary. Source apportionment of PM2.5 concentrations for three different sectors calculated with the new high resolution dataset of local emission from b) PublicPower, c) OtherStatComb and d) Transport

Appendix 3 – Banja Luka, dispersion and source apportionment maps

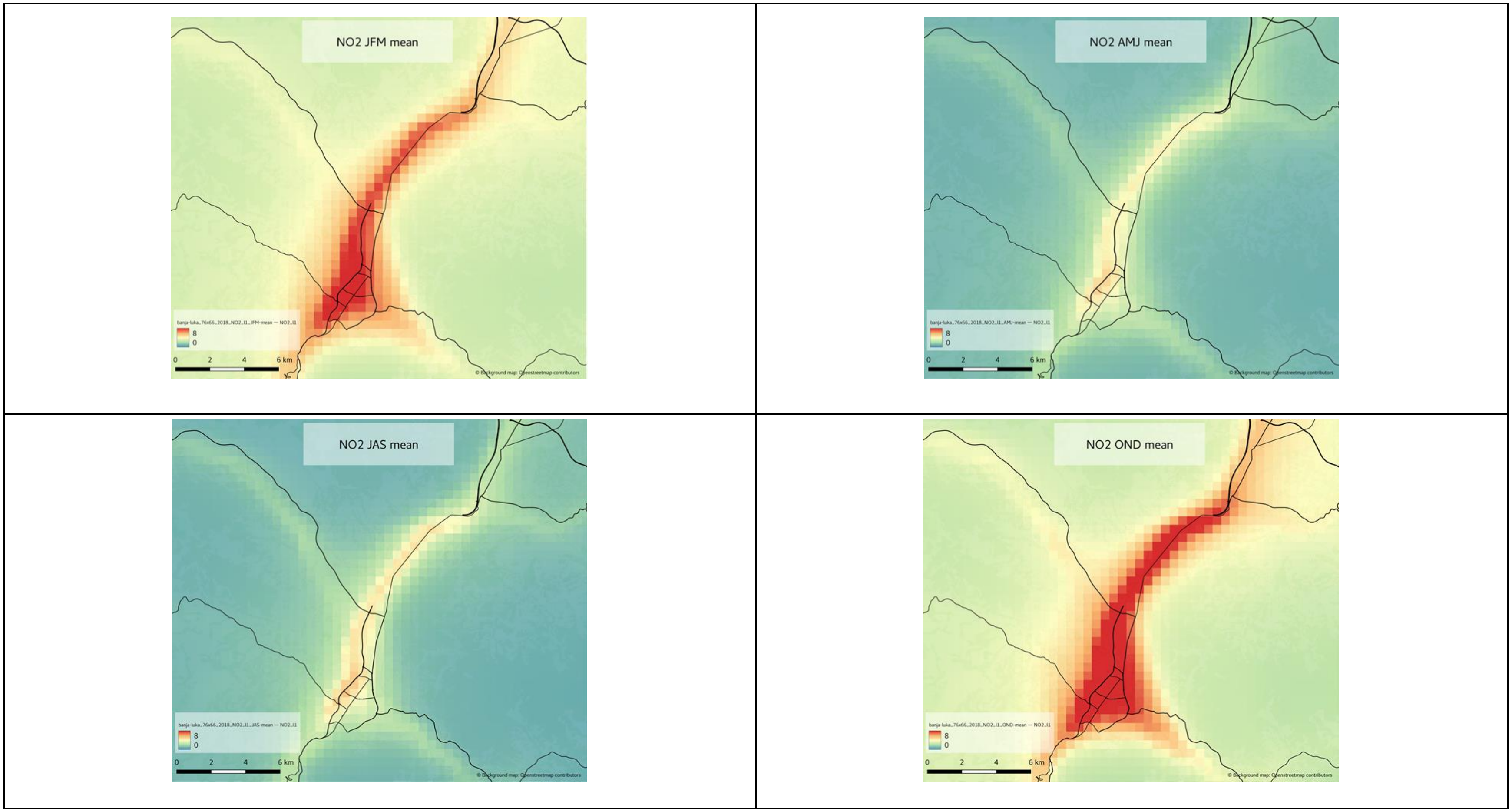


Figure B-1. Spatial maps of modelled seasonal NO2 averages during; a) January-March (JFM), b) April-June (AMJ), c) July-September (JAS) and d) October-December (OND) in Banja Luka, 2018. Units are in ug/m^3

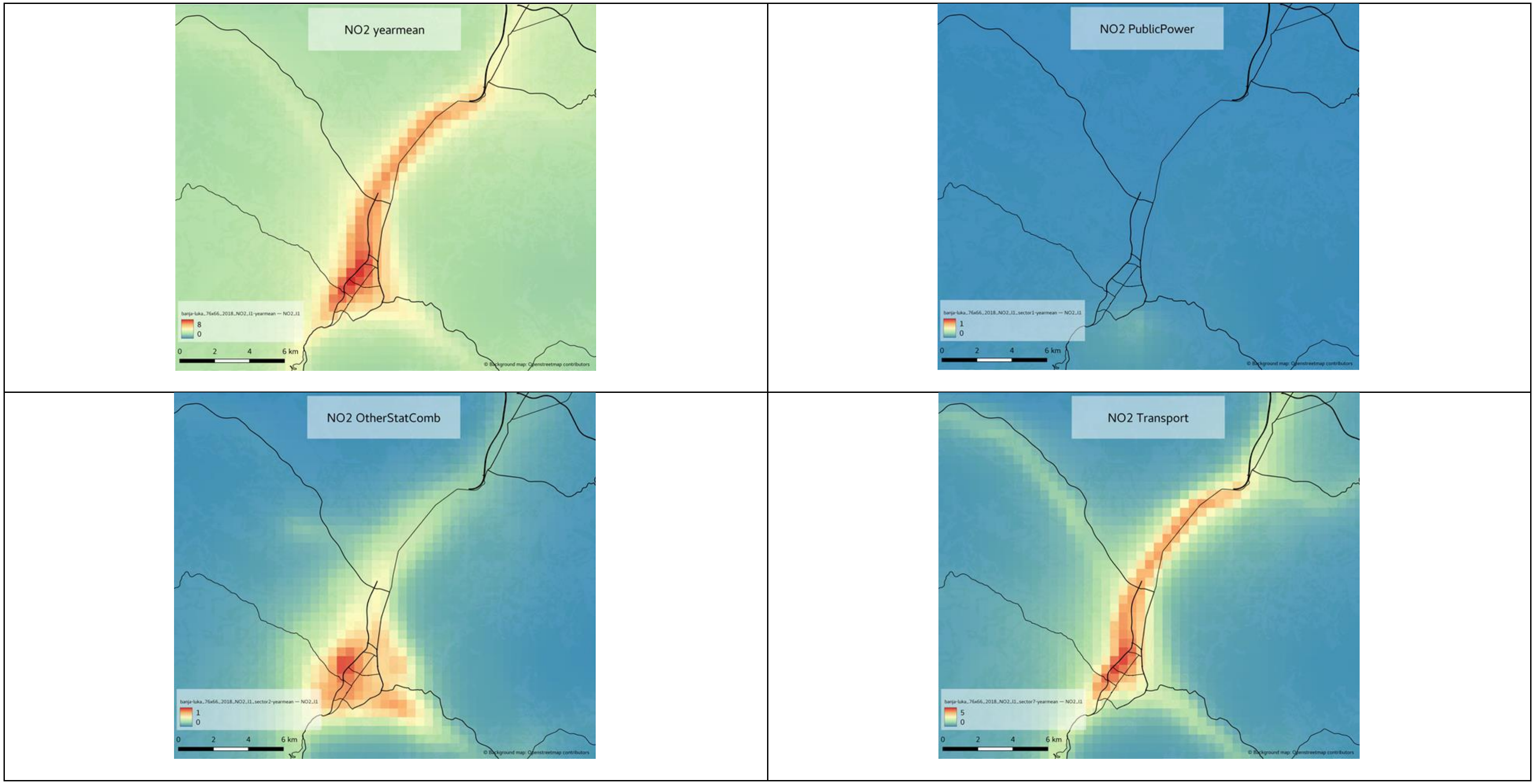


Figure B-2. Spatial maps of modelled NO2 concentrations in Banja Luka, 2018 for a) the total yearly mean based on all available sector emissions within the model domain, including background concentrations from the boundary. Source apportionment of NO2 concentrations for three different sectors calculated with the new high resolution dataset of local emission from b) PublicPower, c) OtherStatComb and d) Transport

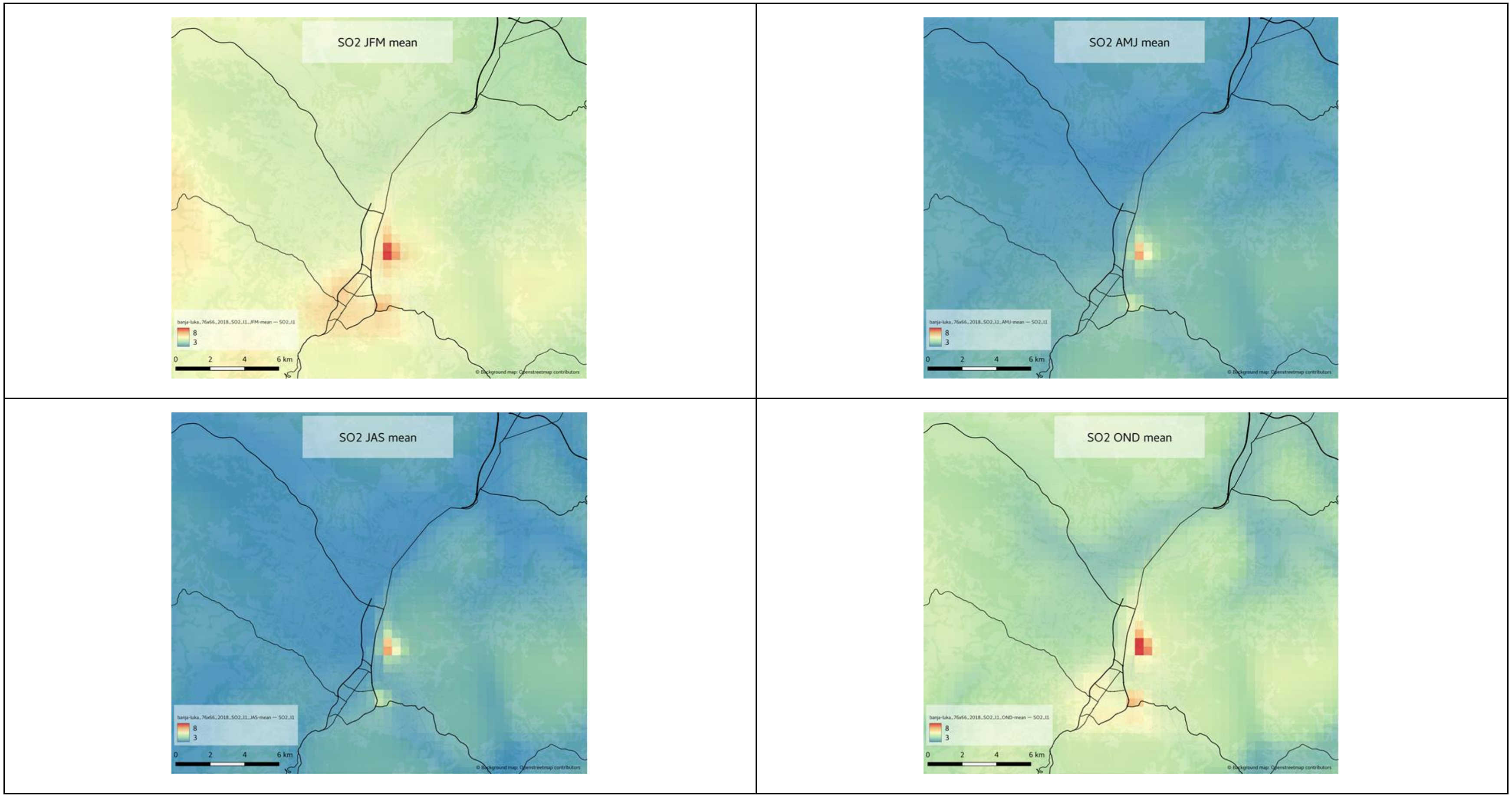


Figure B-3. Spatial maps of modelled seasonal SO2 averages during; a) January-March (JFM), b) April-June (AMJ), c) July-September (JAS) and d) October-December (OND) in Banja Luka, 2018. Units are in ug/m^3

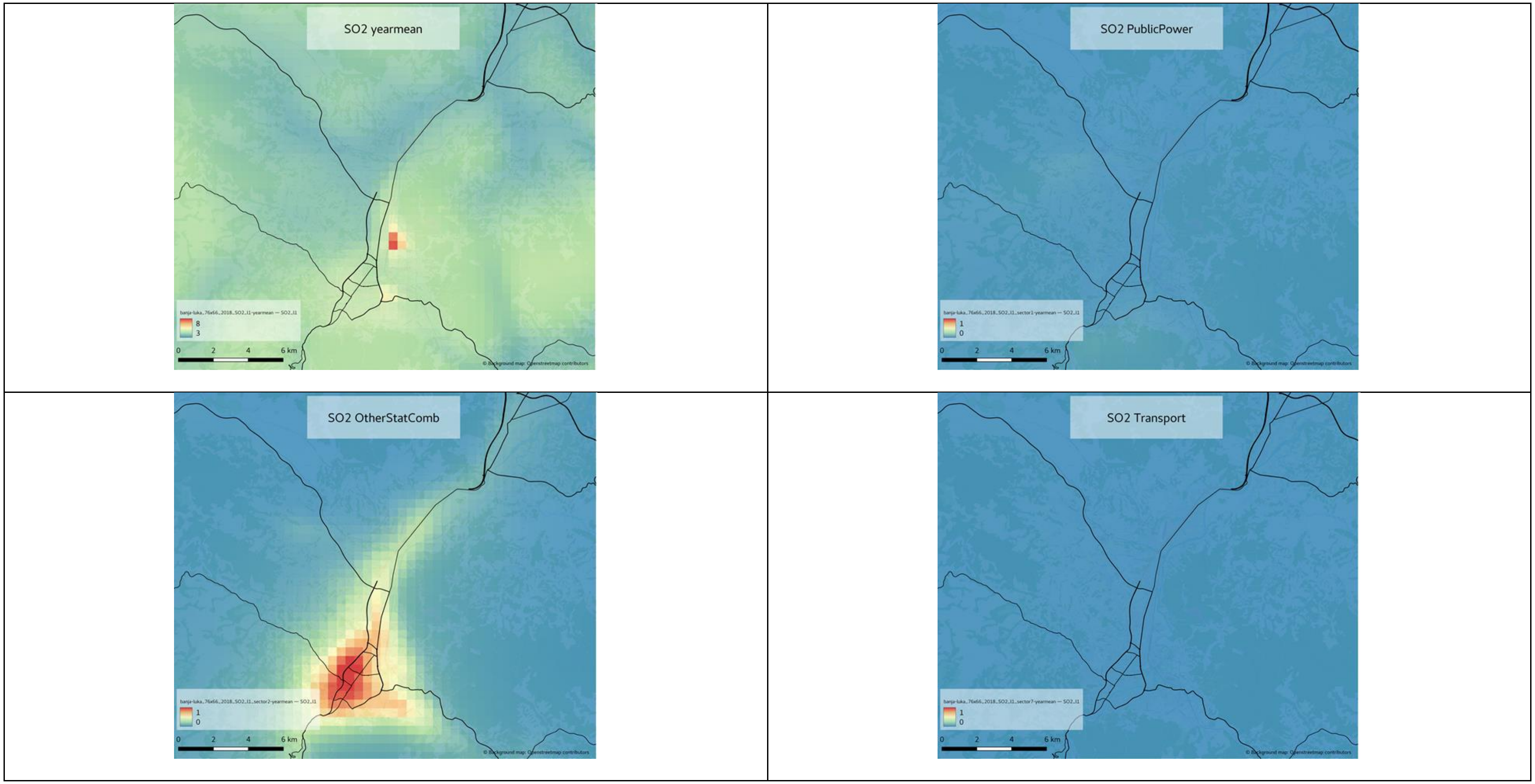


Figure B-4. Spatial maps of modelled SO2 concentrations in Banja Luka, 2018 for a) the total yearly mean based on all available sector emissions within the model domain, including background concentrations from the boundary. Source apportionment of SO2 concentrations for three different sectors calculated with the new high resolution dataset of local emission from b) PublicPower, c) OtherStatComb and d) Transport

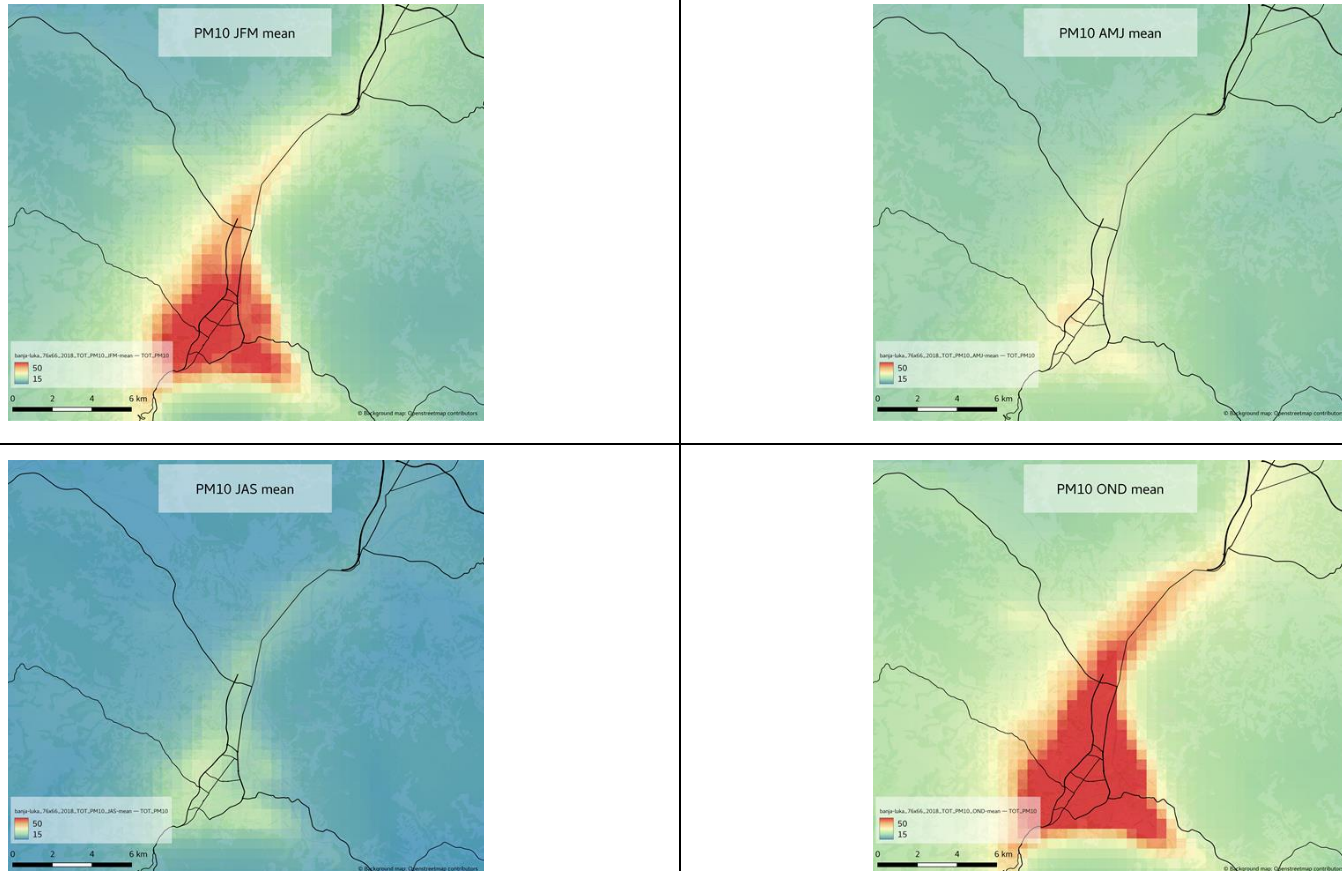


Figure B-5. Spatial maps of modelled seasonal PM10 averages during; a) January-March (JFM), b) April-June (AMJ), c) July-September (JAS) and d) October-December (OND) in Banja Luka, 2018. Units are in ug/m^3

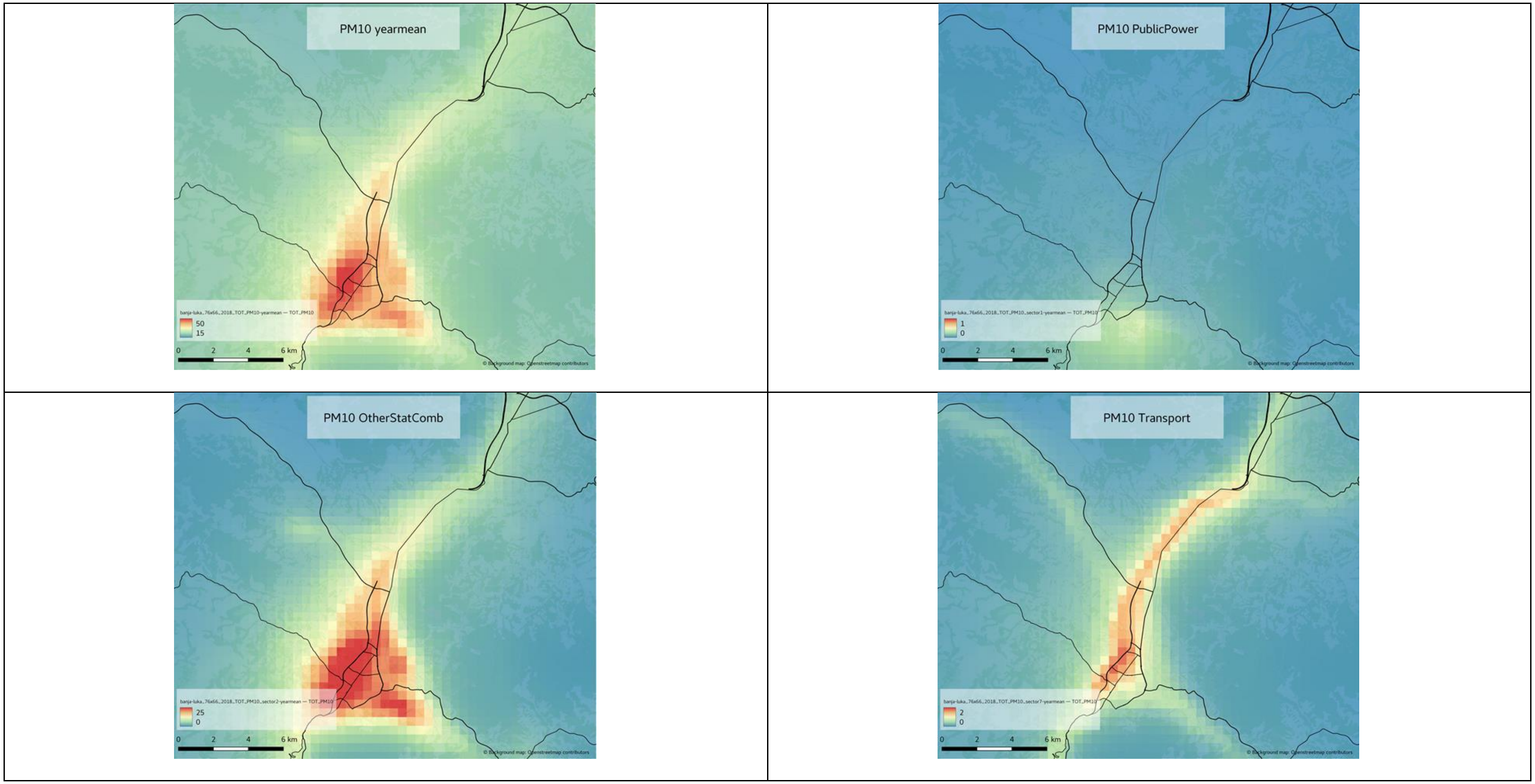


Figure B-6. Spatial maps of modelled PM10 concentrations in Banja Luka, 2018 for a) the total yearly mean based on all available sector emissions within the model domain, including background concentrations from the boundary. Source apportionment of PM10 concentrations for three different sectors calculated with the new high resolution dataset of local emission from b) PublicPower, c) OtherStatComb and d) Transport

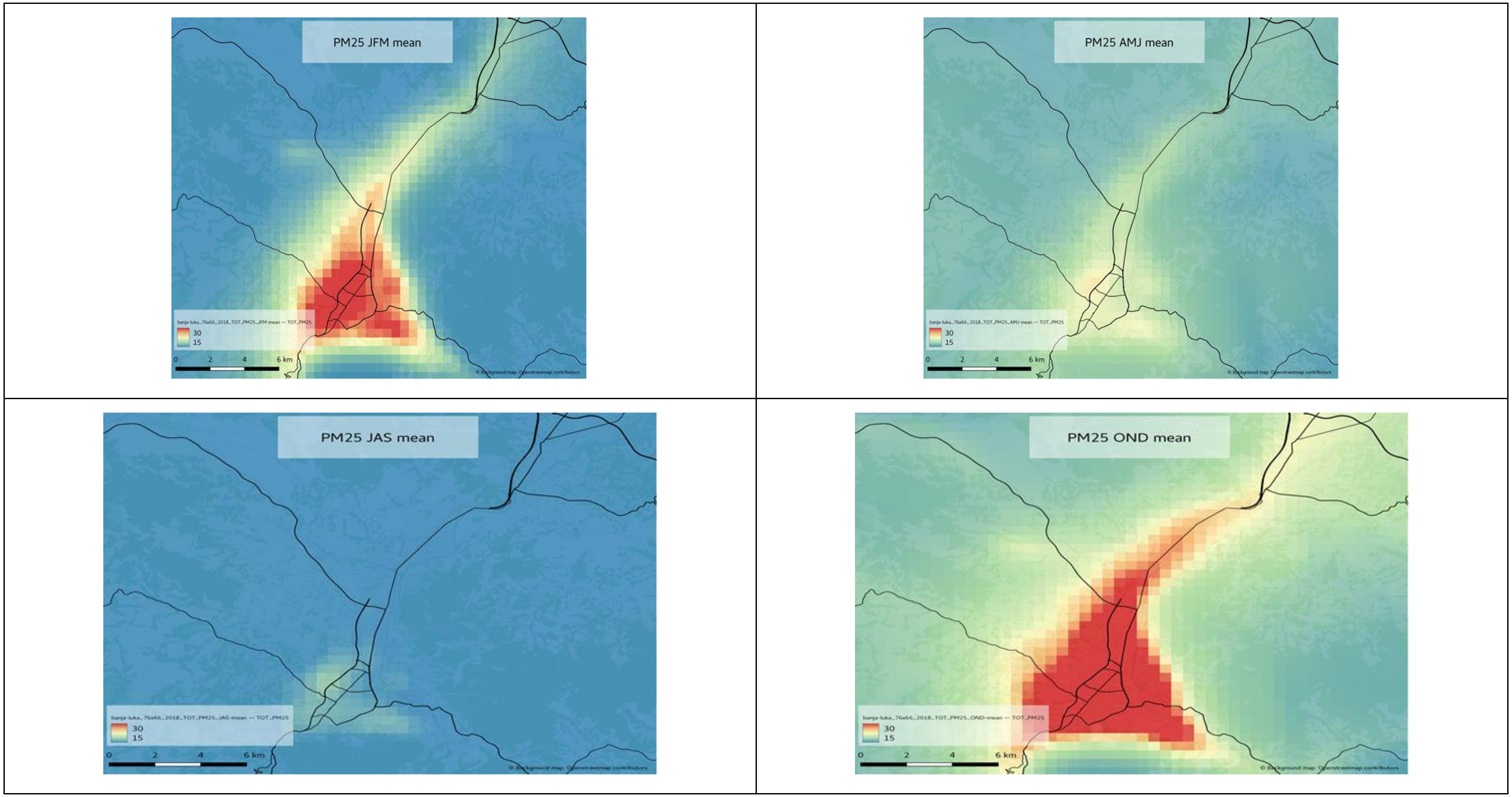


Figure B-7. Spatial maps of modelled seasonal PM2.5 averages during; a) January-March (JFM), b) April-June (AMJ), c) July-September (JAS) and d) October-December (OND) in Banja Luka, 2018. Units are in ug/m^3

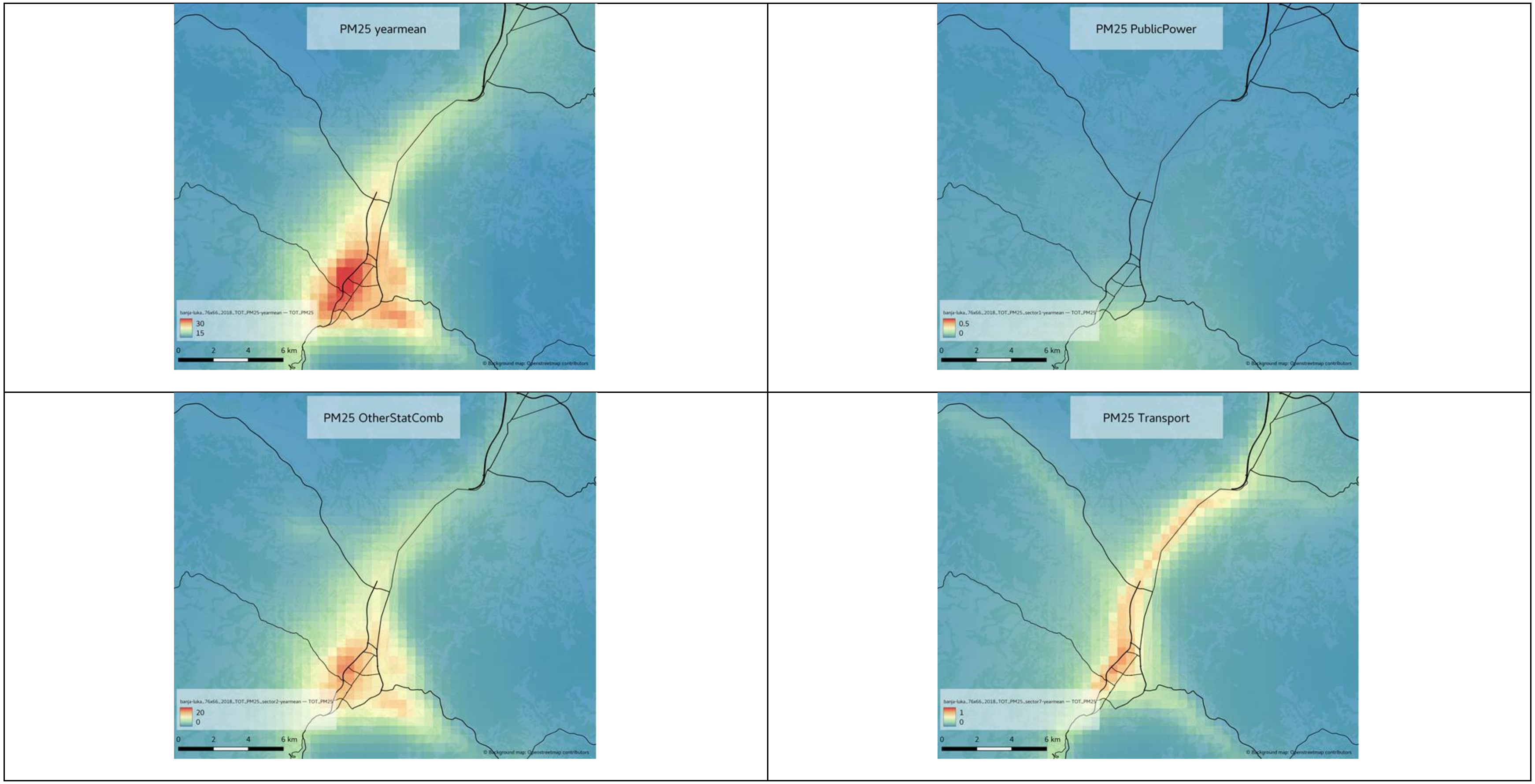


Figure B-8. Spatial maps of modelled PM2.5 concentrations in Banja Luka, 2018 for a) the total yearly mean based on all available sector emissions within the model domain, including background concentrations from the boundary. Source apportionment of PM2.5 concentrations for three different sectors calculated with the new high resolution dataset of local emission from b) PublicPower, c) OtherStatComb and d) Transport