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SPATIO-TEMPORAL VARIABILITY OF ANNUAL $PM_{2.5}$ CONCENTRATIONS AND POPULATION EXPOSURE ASSESSMENT IN SERBIA FOR THE PERIOD 2001–2016

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Abstract: The long-term exposure to $PM_{2.5}$ (particulate matter with aerodynamic diameter ≤ 2.5 μm) is the leading global health risk factor. The spatio-temporal variability of annual values of the total $PM_{2.5}$ concentrations in Serbia is analyzed using the high-resolution (0.01° × 0.01°) gridded data set V4.EU.02 for the period 2001–2016. Population counts and density data were used to calculate the population exposure while the urban land cover data were used to estimate the relations between the size of urban area and the concentration of $PM_{2.5}$. The mean annual values vary in the range 13.93 μg/m^3–28.91 μg/m^3. The regional differences exist, but the highest values were obtained for urban environments (> 22.5 μg/m^3). Negative trend of annual $PM_{2.5}$ is present in most of the parts of the Serbian territory, especially in the eastern parts reaching –0.37 μg/m^3 per year (p ≤ .05). More than 99% of the territory of Serbia has the mean annual values under the national safe limit established by regulation (25 μm/m^3), but comparing to the World Health Organization (WHO) guideline (10 μm/m^3), all the territory is above the safe level. In line with the trend of urbanization, there is a clear upward trend in the number of population exposed to the higher concentrations of $PM_{2.5}$. The share of the population exposed to values higher than 25 μg/m^3 increased from 6.65% in 2005 to 11.40% in 2015, while comparing to WHO standard, the total population in Serbia is exposed to the values higher than the safe one.

Keywords: air pollution; $PM_{2.5}$; trend; population exposure; Serbia

Introduction

Atmospheric aerosol particles or particulate matter (PM) are classified as air pollutants that are directly emitted (primary PM) or formed in the atmosphere from precursor gases through reactions (secondary PM). They can either have natural (marine aerosol, mineral dust, biological aerosol, volcanic

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ash, etc.) or anthropogenic origin (fuel combustion for power generation, domestic heating and transport, industry and waste incineration, etc.) (European Environment Agency [EEA], 2018; Fuzzi et al., 2015; Tasić, Rajšić, Novaković, & Mijić, 2006). The PM consists of complex mixtures of organic and inorganic compounds, solid and liquid particles, with a wide spectrum of physical and chemical properties and different aerodynamic diameter (*ultrafine* with diameter ≤ 0.1 µm—$PM_{\leq 0.1}$, *fine* with diameter > 0.1 ≤ 2.5 µm—$PM_{>0.1 \leq 2.5}$, and *coarse* with diameter > 2.5 ≤ 10 µm—$PM_{>2.5 \leq 10}$) that determine their climate and health-related effects (Reizer & Juda-Rezler, 2016; Vicente & Alves, 2018).

The air pollution has been recognized as a risk factor for human health by the World Health Organization (WHO) since 1958 (WHO Regional Office for Europe, 2017). Ambient (outdoor) air pollution was estimated to cause 4.2 million premature deaths worldwide in 2016, mainly due to $PM_{2.5}$ exposure (WHO, 2018). Numerous studies found a strong exposure-response to the airborne PM in the form of short-term effects (hospital admissions) and long-term or cumulative health effects (morbidity, lung cancer, cardiovascular and cardiopulmonary diseases, etc.) (Beelen et al., 2008; Brauer et al., 2003; Brunekreef & Holgate, 2002; Pope & Dockery, 2006; Vodonos, Awad, & Schwartz, 2018). The $PM_{2.5}$ are small in size and rich in toxic substances (known as lung particles) which determine their greater impact on human health than the coarse PM (Shisong et al., 2018). Deaths worldwide attributable to ambient $PM_{2.5}$ increased from 3.5 million in 1990 to 4.2 million in 2015, making long-term exposure to ambient PM as a fifth-ranked global health risk factor in 2015 (Cohen et al., 2017).

According to the EEA (2018), the $PM_{2.5}$ concentrations reached in 2015 were responsible for about 422,000 premature deaths emerging from long-term exposure in Europe (over 41 countries). The highest numbers of premature deaths and YLL (Years of life lost) were estimated for the countries with the largest populations (Germany, Italy, Poland, France, and the United Kingdom). When considering YLL per 100,000 inhabitants, the largest impact was recorded in central and eastern European countries (Bulgaria, Serbia, North Macedonia, and Hungary) where the highest $PM_{2.5}$ concentrations were also registered. The urban population is mostly affected by air pollution due to numerous activities (traffic, industry, household heating, etc.) that increase the level of PM in the atmosphere (Karagulian et al., 2015). Yang et al. (2018) stated that the increased $PM_{2.5}$ concentration in cities became the major social issue together with rapid urbanization in many regions of the world. Taking into account the effects of PM on the humans, especially when it comes to $PM_{2.5}$, Shaddick et al. (2018) emphasize the need for assessing the population exposure at the national and lower territorial units' level.

Previous studies about air pollution by PM in Serbia are mainly focused on PM_{10}, while those related to $PM_{2.5}$ are rare and often include PM_{10}, $PM_{0.1}$ or other air pollutants (NO_2, SO_2, etc.). Some authors discussed the chemical composition and physical properties of airborne particles, related to different sources of air pollution (e.g., Cvetković, Logar, Rosić, & Ćirić, 2012; Joksić et al., 2009; Šerbula, Antonijević, Milošević, Milić, & Ilić, 2010), while the conducted epidemiological studies examined the association of exposure to airborne PM with health effects (Kovačević, 2019; Stevanović, Jovašević-Stojanović, & Jović-Stošić, 2016; WHO Regional Office for Europe, 2019). Todorović, Radenković, Rajšić, and Ignjatović (2019) point out that the health risk assessment studies for Serbia are scarce due to the lack of long-term air quality monitoring data and medical records, such as hospital admissions and deaths due to specific causes, on daily basis. In order to overcome the disadvantage associated with insufficient and poor spatial coverage of measurement sites (Vuković et al., 2015), several studies use the method of biomonitoring to assess air quality (Aničić, Spasić, Tomašević, Rajšić, & Tasić, 2011; Stamenković et al., 2016). Certain studies discuss low-cost sensing technologies for air quality monitoring and exposure assessment (Jovašević-Stojanović et al., 2015; Tasić, 2017). The studies are mainly related to the so-called "hot spots", larger cities, especially for (the capital) Belgrade (Rajšić, Tasić, Novaković, & Tomašević, 2004; Tasic, Rajsic, Novakovic, & Mijic, 2007) and industrial centers, e.g. the City of Bor (Djordjevic et al., 2013; Tasić, Milošević, Kovačević, Jovašević-Stojanović, Dimitrijević, 2012). On the regional level, Malinović-Milićević, Mihailović, Nikolić-Đorić, and Jevtić (2015) analyzed the impact of air pollution sources (household heating and traffic) on air quality in Vojvodina region for the period 2001–2008. So far, there is a lack of studies on the spatio-temporal variability of $PM_{2.5}$ for the whole territory of Serbia as well as on population exposure.

Assessing the effects of $PM_{2.5}$ exposure is often limited by the absence of ground-level measurements with convenient spatial and temporal coverage. In recent years, remote sensing data have enabled the estimation of the global distribution of ambient $PM_{2.5}$ concentrations. Satellite data are processed together with Chemical Transport Model (CTM) simulations and ground-level measurements to produce the high-resolution datasets enabling an insight into long-term (decade or more) spatio-temporal changes and offer information about the magnitude of human exposure for epidemiological studies (van Donkelaar et al., 2010; van Donkelaar, Martin, Brauer, & Boys, 2015; van Donkelaar et al., 2016; van Donkelaar, Martin, Li, & Burnett, 2019). These data are used in studies on global (Li, Han, Jin, Zhang, & Wang, 2019; Wang et al. 2019; Yang et al., 2018), regional and national levels (Bai, Ma, Chang, & Gao, 2019; Larkin, van Donkelaar, Geddes, Martin, & Hystad, 2016; Lu, Mao, Yang, Zhao, & Xu, 2018). In this research, spatial variability and temporal trend are analyzed using the high-resolution (0.01° × 0.01°) dataset of the total annual $PM_{2.5}$ concentrations in Serbia for the period 2001–2016. In addition, the variability of $PM_{2.5}$ exposure is analyzed depending on the size of the urban area. The same spatial resolution datasets about population count and density are used to estimate the population exposure.

Study area

Serbia is positioned in southeast Europe on the Balkan Peninsula and covers an area of 88,499 km^2 (Figure 1) (Statistical Office of the Republic of Serbia, 2019). The northern part of the country is represented by low land, which is the southeast part of the Pannonian plain. The altitude increases to the south where the hills and mountains interspersed by river valleys and basins are dominant forms of relief. The highest altitude is 2,656 m a.s.l. (Đeravica Peak, Prokletije Mountain) situated on the southwest of the country.

According to the last Census data from 2011 (Statistical Office of the Republic of Serbia, 2014), the number of inhabitants is 7,186,862

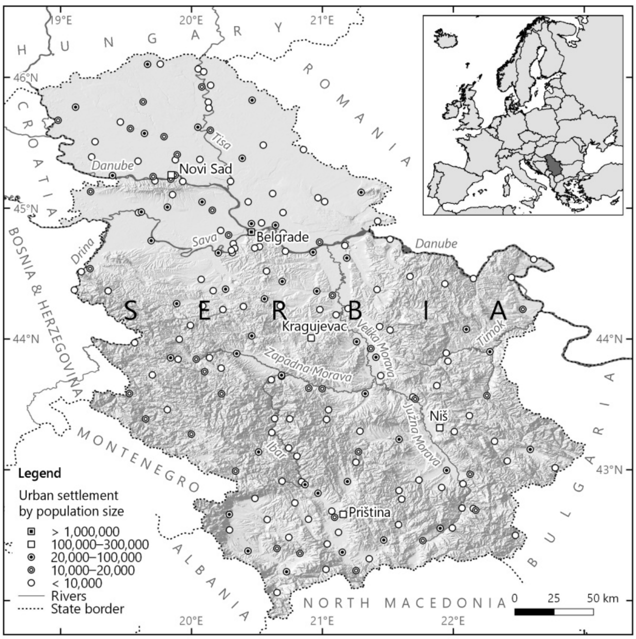

Figure 1. Study area.

(without Kosovo and Metohija[1]). The population is mainly concentrated at lower altitudes in northern and central areas and along the river valleys, such as valleys of Danube, Sava, and Velika Morava rivers. The majority of the population (~60%) lives in urban settlements and the largest number of them is Belgrade, with the population of ~ 1.7 million in the total city area, i.e., ~ 1.2 million in urban city area (Drobnjaković & Spalević, 2017).

Data and methods

The European Regional Estimates (V4.EU.02) dataset (Atmospheric Composition Analysis Group, n.d.) is used to calculate the mean annual values of the total $PM_{2.5}$ (μg/m^3) for the territory of Serbia in the period 2001–2016. It presents the combined geophysical-statistical estimation of $PM_{2.5}$ over Europe using available measurements obtained from the EEA Air Quality e-Reporting system. The estimation of ground-level fine particulate matter ($PM_{2.5}$) in total (μg/m^3) is based on combining Aerosol Optical Depth (AOD) retrievals from the NASA MODIS, MISR, and SeaWIFS instruments with the GEOS-Chem chemical transport model, and subsequently calibrated to regional ground-based observations using Geographically Weighted Regression (GWR) (van Donkelaar et al., 2019). The spatial resolution is 0.01° × 0.01°. Besides the regional aspect of variability, this study explores the changes in $PM_{2.5}$ for different altitudes. Using the European Digital Elevation Model (EU-DEM), version 1.1 (EEA, 2013), several hypsometric zones for the territory of Serbia were singled out (< 200 m, ≥ 200 < 500 m, ≥ 500 < 1000 m, ≥ 1000 < 1500 m, and ≥ 1500 m a.s.l.) and the average mean annual $PM_{2.5}$ is calculated. Digital elevation model has been resampled to fit the grid of $PM_{2.5}$ data to perform a zonal grid statistics report. In order to explore the temporal variability of annual $PM_{2.5}$ values, the linear trend is calculated for the observed period, while for determining the significance of a trend, the Student's t-test is used.

Many urban activities contribute to the increase of PM concentration. This study aims to analyze the variation of $PM_{2.5}$ concentration depending on urban surface size for the territory of Serbia. The Global Human Built-up And Settlement Extent (HBASE) Dataset From Landsat, v1 (2010) (Wang, Huang, Brown de Colstoun, Tilton, & Tan, 2017) is used to obtain the data on the urban extent and "man-made" surfaces with a spatial resolution of 30 m in 2010 as the reference year. For the purpose of this study, urban land cover, i.e., continual built-up surfaces, are categorized into several classes: < 5 km^2, ≥ 5 < 10 km^2, ≥ 10 < 20 km^2, ≥ 20 < 50 km^2, ≥ 50 < 100 km^2, and ≥ 100 km^2. For each class, the average annual $PM_{2.5}$ is calculated for the target year (2010).

The estimation of the population exposure to ambient air pollution is the first step in the impact assessment studies. In order to calculate the population exposure to $PM_{2.5}$, two different datasets were used: UN WPP-Adjusted Population Count, v4.11 (Center for International Earth Science Information Network [CIESIN], 2018a) and UN WPP-Adjusted Population Density, v4.11 (CIESIN, 2018b). The data are available as gridded datasets with spatial resolution of 30 arc-second. The analyses were conducted for 2005, 2010, and 2015 and population count and density were calculated for annual values of $PM_{2.5}$. All the calculations in the study were performed using Quantum Geographical Information System (QGIS) (version 2.18.4) and the System for Automated Geoscientific Analyses Geographical Information System (SAGA GIS) (version 4.0.1, Conrad et al., 2015).

[1]According to Kokotović Kanazir et al. (2017), the estimated number of inhabitants (based on United Nations statistics) in the Republic of Serbia (including Kosovo and Metohia) was 9,854,000 in 2011.

Results and discussion

The variability in the shares of sources that contribute to the emission of $PM_{2.5}$ results in various spatio-temporal patterns. The mean annual values of $PM_{2.5}$ total (2001–2016) for the territory of Serbia vary in the range 13.93 μg/m^3–28.91 μg/m^3. The calculated values are classified into several categories (together with the percentage of territory with these values): < 15 μg/m^3 (3.27%), ≥ 15 < 17.5 μg/m^3 (20.27%), ≥ 17.5 < 20 μg/m^3 (38.41%), ≥ 20 < 22.5 μg/m^3 (36.36%), ≥ 22.5 < 25 μg/m^3 (1.01%), ≥ 25 < 27.5 μg/m^3 (0.64%), and ≥ 27.5 μg/m^3 (0.04%) (Figure 2). The most prominent feature are the isolated "islands" of the highest values of $PM_{2.5}$ (> 22.5 μg/m^3) that are spatially overlapped with the largest urban settlements on the territory of Serbia. Generally, higher values are obtained for the northern part of Serbia (20–22.5 μg/m^3). Central and southern areas show values of 17.5–20.0 μg/m^3, while the lowest ones are in the southeast (15–17.5 μg/m^3) and southwest (< 15 μg/m^3) parts of the country. The analysis of the hypsometric zones and mean annual $PM_{2.5}$ (2001–2016) show a general tendency that, with an increase of altitude, the mean annual $PM_{2.5}$ decreases (Table 1). The average mean annual $PM_{2.5}$ has values of 21.08 μg/m^3 and 16.92 μg/m^3 for the terrain of < 200 m and ≥ 1,500 m, respectively. The minimum and maximum values are similar within the zones with altitudes up to 1,000 m a.s.l., but with the higher difference between them comparing to the terrain with altitude more than 1,000 m a.s.l. This can be explained by the presence of "hot spots" of air pollution, i.e., urban settlements that are mainly situated in lower altitudes. The percentage of the territory of hypsometric zones with specified mean annual $PM_{2.5}$ values is presented on Figure 3. It is shown that the higher percentage of the territory in lower altitude zones is exposed to the higher values of $PM_{2.5}$. This pattern follows the spatial distribution of the population in Serbia.

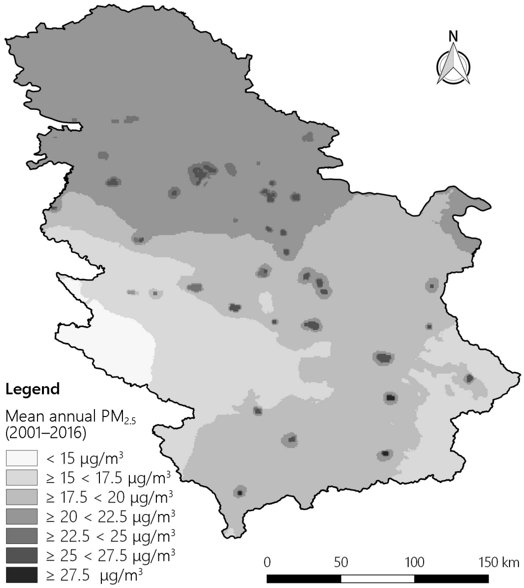

Figure 2. The mean annual values of $PM_{2.5}$ (μg/m^3) in Serbia for period 2001–2016.

Table 1
The average (M), standard deviation (SD), maximum (min), and minimum (max) values of mean annual $PM_{2.5}$ (μg/m^3) for hypsometric zones in Serbia

Hypsometric zones	*M*	*SD*	*min*	*max*
< 200 m	21.08	0.95	15.88	27.38
≥ 200 m < 500 m	18.90	1.32	14.49	28.91
≥ 500 m < 1000 m	17.51	1.26	14.04	27.40
≥ 1000 m < 1500 m	16.21	1.20	13.93	19.30
≥ 1500 m	16.92	0.78	14.49	18.38

The mean annual $PM_{2.5}$ concentration limit established by the European Parliament and the Council of the European Union (2008) is 25 μg/m^3. This is significantly higher than the level considered as safe by WHO, i.e., specified as the guideline value of 10 μg/m^3 (WHO, 2006). The legislation in Serbia is in line with Directive 2008/50/EC and the defined limit for annual $PM_{2.5}$ (Stage 1) is 25 μg/m^3 (Uredba o uslovima za monitoring i zahtevima kvaliteta vazduha, 11/2010, 75/2010, 63/2013). According to the results, it can be concluded that more than 99% of the territory of Serbia has mean annual values under this limit, but comparing to WHO standard, the whole territory is above the safe level. Even comparing to the value of 12 μg/m^3 imposed by the U.S. Environmental Protection Agency for USA (Giannadaki, Giannakis, Pozzer, & Lelieveld, 2018), the whole territory of Serbia has higher mean annual $PM_{2.5}$ concentrations.

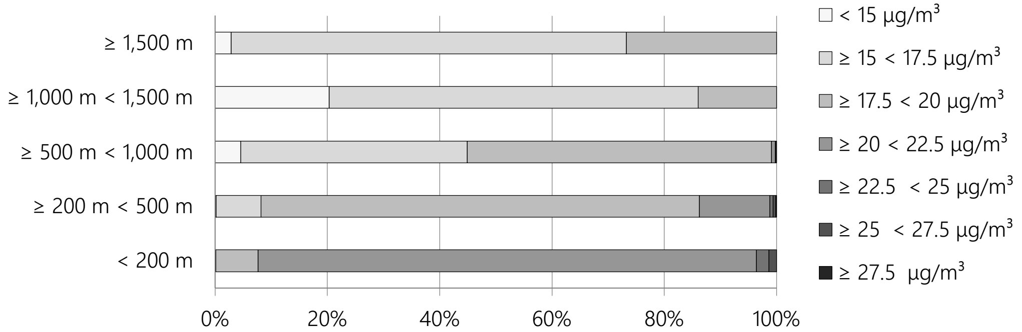

Figure 3. The percentage (%) of the territory of hypsometric zones with specified mean annual values of $PM_{2.5}$ (μg/m^3).

Multiple factors influence the level of $PM_{2.5}$ concentrations in the atmosphere. Kiesewetter and Amann (2014) stated the importance of long-range transport of PM and precursor gases, from Europe-wide transboundary transport to local emissions within an urban area. Also, different source sectors that contribute to emissions vary for different territories. In Serbia, the largest contributors to the emission of gases (precursors of secondary PM) in 2016 were energy production and distribution (49.6% of NO_x and 93% of SO_2) and road traffic (24.3% of NO_x), as well as agriculture (84.7% of NH_3). The district heating plants with a capacity below 50 MW and individual domestic household heating contribute with 21.68% of non-methane volatile organic compounds (NMVOCs) (Lekić, 2018). The contribution of fossil fuels dominates (88%) in primary energy consumption in Serbia (Krunić Lazić, 2018). According to Jovanović (2019), district heating plants with a capacity below 50 MW and individual household heating contribute with 75% in the total emissions of $PM_{2.5}$ in Serbia. In the dwellings without district and central heating (57.2% of the total occupied dwellings in Serbia), wood is mostly used for heating (63.3%), while electricity (15.2%) and coal (13.4%) are used less. In dwellings with central heating (20.6% of the total occupied dwellings in Serbia), wood (46.2%) is predominantly used for heating, followed by coal (25.4%) and gaseous fuels (18.4%), while in dwellings with district heating (22.1% of the total occupied dwellings in Serbia), gaseous fuels (30.4%) and oil fuels (26.1%) are mostly used, followed by coal and wood (21.7% and 17.3%) (Statistical Office of the Republic of Serbia, 2013). The household heating is the leading source of the emissions of $PM_{2.5}$ in many regions in the world (Karagulian et al., 2015; Saarikoski et al., 2008; Samek et al. 2017; Xiao, Ma, Li, & Liu, 2015). The road transport sector contributed with 6%, while the share of industry was 9% in the total $PM_{2.5}$ emissions in 2018 in Serbia (Jovanović, 2019). The relatively small share of industry in the emissions can be explained by the sharp decline in industrial production since the 1990s in Serbia (Hadžić & Zeković, 2019), but on the local level, in the larger industrial centers, it significantly contributes to PM emissions (PM_{10} and $PM_{2.5}$) (e.g., Stevanović, Jovašević-Stojanović, & Jović Stošić, 2016).

The analysis of temporal variability showed a negative trend of annual $PM_{2.5}$ for most of the Serbian territory in the period 2001–2016 (Figure 4). The highest statistically significant negative trend values are obtained for eastern and western parts of the country (in some areas in the east they reach up to –0.37 μg/m^3 per year, $p \leq .05$). The positive trend values are obtained only for small areas in the south that match urban settlements and reach up to 0.19 μg/m^3 per year. These results are in accordance with the findings of studies exploring the spatial distribution and the trends of annual $PM_{2.5}$ using similar datasets and periods. Using satellite data (aerosol optical depth), CTM and ground-measurements, Boys et al. (2014) produced the high-resolution (1° × 1°) global dataset of $PM_{2.5}$ for the period 1998–2012 and singled out four regions with a statistically significant trend: negative trend in Eastern US, and positive in Arabian Peninsula, South Asia, and East Asia. For the region of Southeast Europe (Serbia), they observed the absence of a statistically significant trend or significant negative trend (in the range from –0.25 to –0.50 μg/m^3 per year, α= 1% and α= 5%). Yang et al. (2018) showed an increasing trend of $PM_{2.5}$ along with the urbanization during 1998-2015, but with a variety of evolutionary relations in different countries and regions. They stated that most countries in East, Southeast Asia and South Asia, and some African countries show rapid increase in both urbanization and $PM_{2.5}$ while a decrease of $PM_{2.5}$ concentrations are found in most European and American countries. Van Donkelaar et al. (2019) produced the high-resolution (1 km × 1 km) dataset of the chemical composition of $PM_{2.5}$ for North America and found a decrease in population-weighted $PM_{2.5}$ concentrations since 2000 which can be explained by a reduction in sulfate and organic matter. According to EEA (2019), concerning the period 1990–2017, in EEA-33 region, the emission of NO_x, SO_x, and NMVOCs decreased by 57%, 82%, and 54% (61%, 91%, and 58% in EU-28), respectively. The NH_3 emissions have decreased by 18% (23% in EU-28), but have been continuously increasing since 2014. It is also stated that the three largest sectors (Energy production and distribution, Energy use in industry, and Commercial, institutional and household) have reduced the emissions of SO_x by more than 75% since 1990, but in the EEA-33 region (Southest Europe), the power and heat generation still remain the most significant sources of SO_x contributing to over a half of the current total emissions. Besides the reduction in Energy production and distribution sector, the high impact on NO_x has the decrease in the emissions in road transport sector. Agriculture dominates in the emissions of NH_3 (92% of total emission in EEA-33). The emissions of primary $PM_{2.5}$ decreased by 28% mainly due to the reductions in electricity generation and heavy industrial sectors. The households are still the largest contributor of $PM_{2.5}$ in Southeast Europe. In Serbia, the emissions

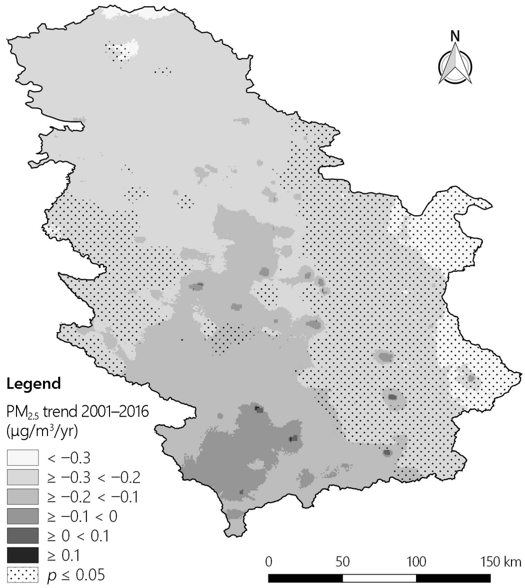

Figure 4. Trend of annual values of $PM_{2.5}$ (μg/m^3 per year) in Serbia for period 2001-2016.

Table 2
The average (M), standard deviation (SD), maximum (min) and minimum (max) of $PM_{2.5}$ (µg/m^3) for urban land cover surfaces for 2010 in Serbia

Classes	*M*	*SD*	*min*	*max*
< 5 km^2	21.12	1.67	15.20	26.00
≥ 5 km^2 < 10 km^2	21.28	1.91	16.10	25.20
≥ 10 km^2 < 20 km^2	22.22	2.11	17.90	28.50
≥ 20 km^2 < 50 km^2	23.25	1.66	18.80	26.20
≥ 50 km^2 < 100 km^2	23.11	1.59	20.00	26.40
≥ 100 km^2	23.33	0.97	22.00	25.30

of NO_x and SO_x showed a slow decrease, the emissions of NMVOCs showed a very slow decrease, while the emissions of NH_3 and PM_{10} did not show any significant change in the period 1990–2016 (Lekić, 2018).

Both coarse and fine PM are present in urban environments. Many studies showed that their proportion varies substantially between cities depending on the local topography, meteorology and specific PM sources (Querol et al, 2008; Rogula-Kozłowska et al., 2014; Perrone et al., 2018). Also, within one urban area, heterogeneous distribution patterns of $PM_{2.5}$ can exist (Cheng & Wang-Li, 2019). The highest mean annual $PM_{2.5}$ values are observed for urban settlements in Serbia. The analysis of $PM_{2.5}$ values and urban land cover for 2010 showed that with an increase of the size of urban land cover, the annual $PM_{2.5}$ increased (Table 2). It is similar for the minimum values, but the maximum values are found in the class ≥ 10 < 20 km^2 (value of 28.50 µg/m^3). In many cities in Serbia, the problem of increased $PM_{2.5}$ concentrations is related to district heating plants and a large share of individual household heating during the winter. This is especially expressed for urban settlements positioned in basins where local morphological and meteorological conditions contribute to the increased pollutant levels. The examples are the cities of Valjevo and Užice in western Serbia, where the observed records are the results of several factors during the winter season such as individual households heating, basin position, poor air ventilation, etc. In the city of Užice, in 2016, daily PM_{10} reached a value of 438 µg/m^3 (Radović & Jovanović, 2017), while in the city of Valjevo, in 2017, daily PM_{10} reached a value of 806 µg/m^3 (the upper limit defined by regulation is 50 µg/m^3) (Radović & Jovanović, 2018). Unfortunately, the measurements of $PM_{2.5}$ do not exist, so it is not possible to make the comparison with this record.

For assessing the impact of air pollution on humans, it is of the greatest importance to determine the population's exposures to the defined pollution levels. Population count and density data are related to the annual values of $PM_{2.5}$ for three years: 2005, 2010, and 2015. The results are presented in Table 3. Certain differences between the observed years exist, but with a clear upward trend in the share of the population exposed to the higher values of $PM_{2.5}$. While the population density in the most polluted areas increases over time, the distribution of the population (in %) varies for the specified $PM_{2.5}$ classes between years. The share of the population exposed to values higher than the nationally established annual safe limit of 25 µg/m^3 increased from 6.65% in 2005 to 11.4% in 2015. Comparing to the WHO limit (10 µg/m^3), the total population in Serbia is exposed to higher values during the observed years. In recent decades, urban settlements in Serbia have recorded population growth (Drobnjaković & Spalević, 2017). This process is accompanied with the increase in traffic activity, household heating, industry and other urban activities that increase air pollution in the urban environment. It is shown that the areas with the highest $PM_{2.5}$ have the highest population density (in 2015, for areas with $PM_{2.5}$ > 27.5 µg/m^3, the average population density is 2,339.28 inhabitants per km^2).

Table 3
The population counts (% of inhabitants) and average population density (inhabitants per km²) for specified values of annual $PM_{2.5}$ (µg/m³) for 2005, 2010, and 2015 in Serbia

$PM_{2.5}$ classes (µg/m³)	2005		2010		2015	
	Population (%)	Population density (inh./km²)	Population (%)	Population density (inh./km²)	Population (%)	Population density (inh./km²)
< 15	0.03	11.24	0.02	9.25	3.35	20.80
≥ 15 < 17.5	8.68	43.44	2.83	32.00	22.90	65.05
≥ 17.5 < 20	27.01	86.34	19.02	57.39	18.50	109.11
≥ 20 < 22.5	14.30	78.74	46.36	98.12	32.65	114.92
≥ 22.5 < 25	43.33	170.51	25.86	362.54	11.20	588.12
≥ 25 < 27.5	6.65	1453.08	5.88	1977.12	9.10	1391.12
≥ 27.5	0	0	0.03	284.13	2.30	2339.28

The air pollution problem is recognized as a global issue and numerous activities have been undertaken through many agendas (i.e., "Transforming our world: the 2030 Agenda for Sustainable Development" (United Nations, 2015)) and action programs (i.e., "Living well, within the limits of our planet" (European Parliament & Council of the European Union, 2013)). Van Donkelaar et al. (2016) found that the global population-weighted annual average $PM_{2.5}$ concentration is 32.6 µg/m³ (1998–2015) and it is driven by high concentrations in several Asian and African regions. The results of this study showed that Serbia is below this worldwide average, but still in the health risk zone, especially concerning the WHO limit. The Report "Health impact of ambient air pollution in Serbia – A call to action" (WHO Regional Office for Europe, 2019) showed that nearly 3,600 premature deaths every year (2010–2015) are attributable to the exposure to $PM_{2.5}$ in 11 cities in Serbia. It is evident that Serbia needs an adequate public policy in mitigating this health risk.

Conclusion

One of the biggest challenges facing the world's population is air pollution problem and finding the solution how to reduce and control the emissions of pollutants in order to prevent negative health effects. Long-term exposure to ambient $PM_{2.5}$ becomes one of the leading global health risk factors. The satellite-derived data products help to overcome the insufficient number of ground-level measurements and give an insight in spatio-temporal variability of $PM_{2.5}$. These datasets provide valuable information for epidemiological studies and health impact assessment. This study analyzes the spatial variability and temporal evolution of total $PM_{2.5}$ and estimates population exposure in Serbia for the period 2001–2016. The main findings are:

- The mean annual $PM_{2.5}$ values (2001–2016) vary in the range 13.93 µg/m³–28.91 µg/m³. The higher values are obtained for the northern part of Serbia (20–22.5 µg/m³). Central and southern areas show values of 17.5–20.0 µg/m³, while the lowest values are recorded on the southeast (15–17.5 µg/m³) and southwest (< 15 µg/m³). In addition, with an increase in altitude the concentration of $PM_{2.5}$ decreases. The spatial pattern of $PM_{2.5}$ in Serbia follows the distribution of the population with the highest values for urban areas (> 22.5 µg/m³).
- For most parts of Serbia, there is a negative trend of annul $PM_{2.5}$, with the highest values in the eastern part of the country (–0.37 µg/m³ per year). This is confirmed by the findings of other studies that are using similar datasets and periods to analyze trends of annual $PM_{2.5}$. In Europe,

a significant reduction in emissions of gases (that are precursors of secondary PM) since 1990, is mainly related to the changes in energy and industry sectors. In Serbia, for the period 1990–2016, the NO_x, SO_x, NH_3, and NMVOCs showed a slight decrease in emissions.

- Despite the negative trend, the problem of air pollution, i.e., $PM_{2.5}$, became a leading challenge in most urban environments mainly due to the emission from household heating, traffic, industry, and other urban activities. It is shown that, in Serbia, with the increase of the size of the urban area, the concentration of $PM_{2.5}$ increase as well. The highest observed concentrations are not always recorded for the largest urban areas due to the influence of many factors (local topography, meteorology, and specific of PM sources). High level of pollutants has been recorded in many cities in Serbia during the winter months, due to activation of large number of individual household heating. In 2018, heating plants with a capacity below 50 MW, and individual domestic household heating contribute with 75% in total emissions of $PM_{2.5}$.
- It is shown that more than 99% of the territory of Serbia has the mean annual values under the national limit established by regulation (25 $\mu m/m^3$), but comparing to WHO standard (10 $\mu m/m^3$), the whole territory is above the safe level. In line with urbanization, there is a clear upward trend in the share of the population exposed to higher values of $PM_{2.5}$. The share of the population exposed to values higher than 25 $\mu g/m^3$ increased from 6.65% in 2005 to 11.4% in 2015. Comparing to the WHO limit, the total population in Serbia is exposed to higher values in all the observed years (2005, 2010, and 2015).

Despite some advances in spatio-temporal resolution of the existing $PM_{2.5}$ datasets, there are still uncertainties mainly due to the insufficient number of ground-level measurements. The larger spatial coverage by ground-level monitoring stations can improve model accuracy as well data about chemical composition. Also, this is important for understanding the small-scale processes, especially in urban environments where the population is exposed to the highest air pollution. The significant efforts must be performed in the area of public policies to reduce emissions and population exposures. This study aims to contribute to scientific knowledge about $PM_{2.5}$ for the territory of Serbia. Further research will focus on the local predictors of $PM_{2.5}$ distribution and episodes with extremely high values of PM.

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