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Analysis of the dependence of the observed urban air pollution extremes in the vicinity of coal fuelled power plants on combined effects of anthropogenic and meteorological drivers

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ABSTRACT

In this paper we assessed effects of changes of meteorological drivers, taken from datasets of observational records and modelling outputs, and human-made pollution, derived from records of energy production, on the mainly wintertime extreme observed values of urban particulate matter (PM) concentrations in the relative vicinity of coal fuelled thermoelectric power plants (TPPs) in Montenegro and Serbia. We used wavelet transform analysis, together with the dependency analysis and analysis of averages of climatic conditions, to study temporal dynamics of urban air pollution extremes in the vicinity of TPPs, the coincidence of their changes with observed levels of SO_2 and NO_2 concentrations in the air, and dependence of PM changes on several possible meteorological and anthropogenic drivers. We found that PM variations in urban areas are most probably caused by PM-SO_2/NO_2 coincidences that appear after a 2- to 3-h time lags needed for transformation of SO_2/NO_2 TPP emissions into PM particles, if pollution is caused by TPP emissions alone. When other causes of PM variations than the TPP production exist, we found that PM-SO_2/NO_2 correlations appear at time ranges from several hours to several days. In our analysis only the changes in the planetary boundary layer height (PBLH) coincided with the drive to extremes in PM values, at PBLH levels lower than 300*m*. Following these findings, we suggested that PM extremes in our sample could be viewed as preconditioned compound events, where TPP and urban heating emissions provide preconditions for PM extremes and PBLH serves as a major meteorological driver to such events.

1. Introduction

Particulate matter (PM) is a major environmental risk factor for human health to which the World Health Organization's (WHO) last estimate attributed 4.2 million excess deaths worldwide in 2017 (WHO, 2022; World Health Organization, 2006). Of those,

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industrial air pollution and air pollution from the energy sector majorly contribute to the increase and severity of respiratory diseases and connected mortality, leading to one million excess annual deaths (27.3% of global annual mortality) according to the latest estimate of the Global Burden of Disease (Institute for Health Metrics and Evaluation, 2020). This egregious health preconditioning will only intensify complications from other diseases or potential harmful effects from yet unknown respiratory illnesses, as was already the case with Covid-19 (X. Wu et al., 2020).

PMs are complex chemical mixtures of liquid droplets and solid particles of various origins that differ in size and chemical composition (Jimenez et al., 2009; Masselot et al., 2021). Some PM particles or their constituent parts are naturally present in the atmosphere, whereas others are produced by anthropogenic sources. Human activities contribute to the direct emissions of particles containing organic carbon (OC) and black carbon (BC), and to the secondary reactions involving gaseous sulphur dioxide (SO_2), nitric oxide (NO) and nitrogen dioxide (NO_2), ammonia (NH_3), and volatile organic compounds (VOCs) that lead to the formation of atmospheric coarse PM of less than 10 μm (PM_{10}) in diameter, a group that includes fine PM, health-damaging particles with a diameter less than 2.5 μm ($PM_{2.5}$) (McDuffie et al., 2020). Current estimates associate 78% of global SO_2 emissions and 60% of NO_x emissions with the energy production (McDuffie et al., 2020). Of those, 63% of current total global SO_2 emissions and around 30% of NO_x emissions are estimated to emanate for coal-fired power plants (McDuffie et al., 2020); on the regional level, as an example, emissions of SO_2 from such power plants accounted for 63% of the anthropogenic source total in the United States at the end of the last century (Brock et al., 2002). Other important sources of human-made air pollution are energy sector and industrial combustion of oil and gas and international shipping for NO_x emissions, on-road transport and residential combustion for NO_x, OC and BC emissions, mainly agriculture and partly waste for NH_3 emissions, and mixture of different sectors' contributions for VOC variability (McDuffie et al., 2020).

Global SO_2 and to some extent NO_x emissions have declined during the last two decades, majorly due to the emission control policies in industry and in energy sectors, as well as due to the continued reductions in on-road transport emissions in Europe and in North America. In Europe, total decrease of SO_2 emissions is estimated at a factor of 6.9, for the period 1979–2017, while NO_x emissions declined by the estimated factor of 2, mostly due to the restrictive vehicle emission standards in the period from 1992 (EEA, 2022b; McDuffie et al., 2020). Since air pollution is a cross-border phenomenon, trends in the future SO_2 and NO_x emissions will mainly reflect activities in current rapidly growing world regions and/or regions that still do not have all the appropriate climate and environmental strategies and regulations implemented. On a European level some of such remaining causes of SO_2 and NO_x secondary PM pollution are still insufficiently regulated coal-fuelled thermoelectric power plants (TPPs) that are major sources of electricity generation in Western Balkans (Belis et al., 2019). Here, the electric power generation is characterized by a high share of fossil fuels, especially coal. This dependency on mainly lignite-fired power generation (Vasquez et al., 2018) is the emission scenario with worrying future implications (Hewitt et al., 2021).

Out of 18 Western Balkans TPPs (CEE Bankwatch Network, 2019), in this paper we were interested in pollution effects to nearby urban areas of the Montenegrin condensation TPP Pljevlja and Serbian TPP Nikola Tesla (in what follows TENT). TPP Pljevlja was put into operation in 1982, initially designed to work as a power plant block of 210 MW (Government of Montenegro, 2015). The altitude of the TPP Pljevlja is 760*m*, while the height of its chimney is 250*m* so that its outlet exceeds 1000*m* above the sea level. The most important mineral raw material - fuel for the operation of the TPP Pljevlja, is coal from the Pljevlja basin, which belongs to the brown lignite group with calorific value that ranges from 3000 to 3700 kcal/kg (Stefanovic et al., 2019). This coal contains about 0.6–0.9% of sulphur (depending on the deposit), about 18% of ash, 34% of moisture and about 30% of carbon (Government of Montenegro, 2015). Electricity generation of TPP Pljevlja is dominant in the total realized electricity production in Montenegro; it amounted for 55% of total production in 2011 and 46% in 2021 (REGAGEN, 2022).

Serbian TPP TENT consists of two branches – TENT A and TENT B, constructed some 15 km apart, and put into operation in 1970 (TENT A; now operating with 1766 MW of installed power) and 1983 (TENT B; now having 1300 MW of installed power) (EPS, 2022a, 2022b). Electricity generation from coal constitutes 54% of Serbian total installed power plants capacities (Nikolic and Filipović, 2020), with TPP TENT covering most of this share, around 50% of total power production (EPS, 2022b). Coal for the TPP TENT is supplied from its own mines located in the vicinity of the TPP, and from the procured reserves from the neighbouring underground coal mines. TPP TENT complex has three chimneys of different heights: of total of 150*m*, 220*m* (TENT A), and 280*m* (TENT B) (Nikezić et al., 2017). The coal fired in TPP TENT is lignite with about 48% of moisture, 21% of carbon, 0.4% of nitrogen, 0.2% of burnable sulphur, and 19% of ash (Stevanovic et al., 2019).

According to the Centre for Research on Energy and Clean Air (CREA) and CEE Bankwatch Network briefing paper on Western Balkan TPP pollution in the period 2015–2019 (CEE Bankwatch Network, 2019), Serbia was the highest emitting SO_2 European country in 2015 and in 2019, releasing 320000 tonnes of SO_2 in 2019. Of those, TPP TENT released 172100 tonnes in 2019, at the time more than total emissions of Poland (88500 tonnes) and Germany (79200 tonnes), the two highest EU polluters, together. According to the same report, Montenegrin SO_2 emissions doubled between 2015 and 2019, from 22400 tonnes to 46600 tonnes, bringing it to sixth place of European SO_2 polluters. Finally, TPP Pljevlja was ranked fourth of the top five NO_x European polluters, with the emissions of 3.2 tonnes of NO_x per GWh. In this paper, TPP Pljevlja and TPP TENT (Belgrade) were taken as relevant examples of power plants in the Western Balkans; due to their large capacities and significant health, environmental, and economic impacts, these power plants are representative of pollution effects of outdated coal-fired power plants. Understanding their impacts may help develop further policies and strategies to reduce pollution, improve public health, and achieve sustainable development in the region.

It was very recently shown in the large global multi-city study of air pollution that the two components representing the largest fraction of urban PM particles are generally SO_2 and NO_x, which were both linked to the fossil fuel burning (Masselot et al., 2021). It has also already been shown from the observational studies of coal-fired TPPs in the south-eastern United States (Brock et al., 2002) that PM particles are present at the edges of some power plant plumes within 2 h of emission, with detectable increases in particle volume with increasing plume age, viewed as signs of continued gas-to-particle conversion, associated with TPPs that emit substantial quantities of SO_2. The same study found that the SO_2-rich energy and industrial emitters were larger sources of PM pollution than the other sources in the nearby urban areas. The fraction of TPP emissions that constitutes PM pollution depends on and can be modified by climatic, meteorological, or environmental conditions (Masselot et al., 2021).

In that manner, the planetary boundary layer hight (PBLH) can critically influence concentrations of surface PM, especially during the episodes with unusually low PBLH that can lead to unusually high PM concentration. These episodes with excessive high PM concentration can be the result of a positive feedback loop between PBLH and aerosols concentration or of the massive secondary PM production within the boundary layer (Petäjä et al., 2016). Both processes ultimately lead to a nonlinear relation between PBLH and surface PM concentration; there exist recent attempts to, using direct observations, estimate exact inverse function between these two variables (Su et al., 2018). It was additionally shown that even with significant decrease in pollutant emissions the feedback loop and the production of secondary aerosols can cause pollution levels similarly high to those from high emissions of primary pollutants and low PBLH (Su et al., 2020). Other surface meteorological variables such as wind, temperature, relative humidity, surface pressure and precipitation can also play significant role in terms of concentration of surface PM but the influence can vary in terms of season and region (Chen et al., 2020; Singh et al., 2021). In terms of the process of secondary PM production humidity, temperature and surface radiation can have specific role, and the influence of these variables can be different for different seasons (Mishra et al., 2023). Several studies have shown that during haze pollution events secondary aerosols can contribute to total concentrations up to 77% of organic $PM_{2.5}$ (Huang et al., 2014). On the other hand, it appears that PBLH can be considered as a single most important meteorological driver for the surface pollution dynamics (Su et al., 2020), probably because it integrates all other surface variables in a single measure of overall state of the planetary boundary layer and vertical turbulent mixing within it. Typical situations with shallow and stable planetary boundary layer and low PBLH are during the wintertime, with low incoming solar radiation and low winds, when sources of turbulent kinetic energy production, wind shear and buoyancy forces are low or even absent.

Finally, emissions from traffic should always be considered as one of the significant sources of air pollution in the cities (Pant and Harrison, 2013). In addition to direct vehicle emissions of PM, secondary pollutants that are partly formed from traffic emissions are also present in the urban air (H. Wu et al., 2021), thus elevating its total concentration of PM. Traffic-related air pollution also includes a non-combustion emissions such as road dust and tire wear (Fussell et al., 2022). The periods of total lockdown in 2020, during global pandemic od Covid-19, were especially interesting for investigation of the contribution of traffic emissions to total levels of concentrations of PM in cities, since during that period emissions from vehicles, both direct (gasses and particles) and indirect (dust and tire wear) were almost absent, or on a very low level (Ruberti et al., 2020).

In this paper we were interested to use meteorological observational records and modelling outputs in combination with records of energy production and city road traffic to investigate in more detail origins of mainly wintertime extreme observed values of PM concentrations in urban clusters and large urban areas in the relative vicinity of coal fuelled TPPs TENT and Pljevlja. We were interested to understand their compound anthropological and meteorological drivers and to examine and quantify characteristic times of PM composition around TPPs. Our additional interest was to examine other urban anthropogenic drivers of PM emissions, particularly coal, oil and gas burning for residential heating and vehicle emissions. Our intent was to add to the body of observational and modelling studies that aim at providing information on PM chemical composition and spread to assist public health preparedness and policies as well as continued reduction of SO_2 and NO_x emissions from coal fuelled TPPs.

This paper is organized as follows: in the next section (Section 2) we provide essential information on the data that we used for our analysis, which is followed by the short description of the used analysis methods. In Section 3 we provide our results and finish the paper in Section 4 with the discussion of our findings and a recommendation for their use to aid public health interventions.

2. Data and Methods

2.1. Data

Meteorological variables used in this paper were provided from ERA5 dataset (Hersbach et al., 2018). Boundary layer height (PBLH) and temperature at 2*m* above surface (T2m) were given on regular latitude-longitude grid with horizontal resolution of 0.25 x 0.25°. Hourly data of those variables were used. ERA5 is the fifth generation ECMWF reanalysis for the global climate and weather for the past 4 to 7 decades. Reanalysis combines model data with observations from across the world into a globally complete and consistent dataset using the laws of physics (Copernicus, 2018). Hourly and daily observational meteorological records for the town of Pljevlja (Montenegro) and Belgrade (Serbia) were taken from the Institute of Hydrometeorology and Seismology of Montenegro (ZHMS, 2022) and the Republic Hydrometeorological Service of Serbia (RHMZS, 2022) respectively, for the period 2011–2021.

Unverified real-time hourly records of the city of Belgrade PM, SO_2 and NO_2 for the period 2010–2021 are publicly available and were downloaded from the Serbian Environmental Protection Agency (SEPA) website (SEPA, 2022). The station "Novi Beograd" (New Belgrade; at 44° 48′ N and 20° 24′ E) was a representative station that we used for Belgrade. It is located in an urban area and belongs to the group of background stations. The data set related to the town of Pljevlja air quality includes the unverified real-time hourly raw data of PM, SO_2 and NO_2 concentrations, which refers to the period from 2014 to 2021 and were provided by the Environment Protection Agency of Montenegro (EPA Montenegro, 2022). In this paper we used "Pljevlja-Gagovica imanje" measuring station (43° 21′ N and 19° 20' E), because it is a representative station measuring background pollution in the TPP Pljevlja nearby urban area, northern air quality zone. Based on the quality and the richness of available datasets we chose to analyse $PM_{2.5}$ data for Belgrade and PM_{10} records for Pljevlja. In those, the Belgrade $PM_{2.5}$ dataset is missing the entire 2017–2018 winter (heating) season. In addition, our Pljevlja SO_2 and NO_2 records were flagged for the indistinct values by the Montenegrin EPA, and we excluded those values from our analysis. In the case of Pljevlja PM10 data we did not have such markers provided, and since unverified Pljevlja PM10 dataset included some strikingly high values, we decided to use only concentrations of $PM_{10} < 300$ mg/m^3 for our analysis, to avoid those records and seemingly artificial data repetitions that were accompanying them.

The distances between two TPPs TENT A and TENT B and "Novi Beograd" station are around 25 km and 35 km respectively. Both TPPs are located on the Sava riverbank west of Belgrade, with no significant topographical features between them and the city of Belgrade, nor any artificial obstacle. Both locations belong to the periphery of the low land Pannonian Plain. According to the last official census from 2011, the city (Eurostat, 2018) of Belgrade has 1.68 million inhabitants (City of Belgrade, 2011). The city is the capital of Serbia and is thus also the centre of multiple administrative and industrial governing bodies and institutions. The relative position of the city of Belgrade and TPPs can be considered unfavourable, since the second most dominant cluster in a city wind rose is the one that includes W, WNW and NW directions (please see the wind rose of Belgrade in Fig. 1), indicating that direct transport of pollutants can be considered frequent.

The "Pljevlja-Gagovica imanje" station is located at around 5 km west of TPP Pljevlja, with no significant topographical features between them; the TPP and the town belong to the same municipality. According to the last (2011) Montenegrin census, the town (Eurostat, 2018) or an urban cluster (US Census Bureau, 2010) of Pljevlja has 30786 inhabitants (MONSTAT, 2011). Even if it is a rather small town, Pljevlja is surrounded by the industry that supports its TPP – coal mine and ash and slag dumps, that heavily pollute its environment (Government of Montenegro, 2015). The town of Pljevlja wind rose is also included in Fig. 1.

In our analysis, we used the European Network of Transmission System Operators (ENTSO-E) Transparency Platform hourly electricity generation per production type country data (ENTSO-e, 2022). These records are calculated as hourly averages of all available instantaneous net generation output values on each market time unit, or if those are not available (that is, no real-time measurement devices exist) the estimates are given instead (ENTSO-e, 2022). The records for electricity generation from coal were available from 2017 for Serbia and from 2018 for Montenegro. We also used data obtained from TPP Pljevlja on the generator-created electricity and working hours per day, available for the period 2014–2021. Such records from TPP TENT were not available to us.

For consideration of dependences of recorded PM concentrations on the urban traffic, we used daily values from the automatic traffic counters from New Belgrade, available for the period 2018–2019. These data were provided to us by the metropolitan Office for Transport and Traffic (City of Belgrade, 2022b). The automatic traffic counters are sensors that are imbedded into the upper layers of city roads that monitor different aspects of road traffic. They provide, among other, hourly numbers of cars that pass above them, from which total daily traffic numbers (numbers of cars) are calculated. The information is always provided from a pair of sensors, to cover the vehicle flow in two directions (City of Belgrade, 2022b).

In this paper, we considered heating season to span from October 1 to April 30 (City of Belgrade, 2022a).

2.2. Wavelet transformation method

We used wavelet transformation (WT) as a part of our data analysis. WT was developed as an alternative to the Fourier trans-

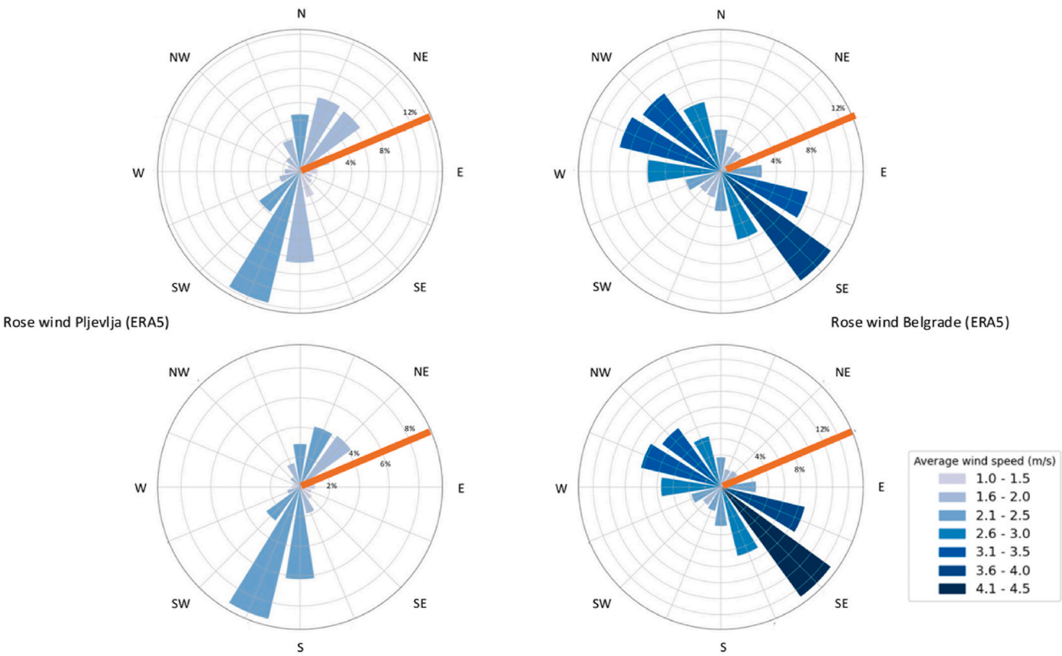

Fig. 1. Wind rose for the town of Pljevlja (left) and the city of Belgrade (right), given as annual (top) and seasonal (OCT-DEC and JAN-APR; down) ERA5 averages for the period 1981–2010.

formation (FT) of signals or discrete data that partially overcomes the FT limitations to locate signal changes (decomposition) in real time (or space) as well as in frequency (Addison, 2002; Torrence and Compo, 1998; Wilczok, 2000). WT performs a two-dimensional time/space (in this paper we were interested into the time decomposition, so will proceed to mention only this component in what follows) and temporal scale decomposition with a group of functions constructed by expanding by time scale and translating along the real time of a specifically chosen original wavelet function (Astaf'eva, 1996); this allows for visualization of local (temporal) components of the analysed signal. When these two-dimensional signal decompositions are integrated over real time, global signal characteristics are calculated (Torrence and Compo, 1998). Those are analogous and mathematically comparable to corresponding FT decompositions (Perrier et al., 1995). In this paper we calculated global wavelet power spectra (WTS), to examine characteristic time intervals of changes in our data. As Fourier power spectra, WTSs are defined as local wavelet power spectra $E(a,b) = |W(a,b)|^2$, where $W(a,b)$ are values of the wavelet coefficients for a given signal at the time scale (equivalent to frequency in Fourier decompositions) a and real time b, that are integrated, or in discrete case summated, over real time (Addison, 2002; Astaf'eva, 1996; Torrence and Compo, 1998). Additionally, as a part of the analysis of this paper we calculated the local wavelet cross-correlation functions (lcWTS), which are, as in Fourier decompositions, defined as $CE_{x,y}(a,b) = E_x^*(a,b)E_y(a,b)$, where E_x and E_y are local WTSs of signals (records) x and y respectively and (*) is complex conjugation (Addison, 2002, 2018). Since $CE_{x,y}$ are by this definition complex functions, we used their absolute values for data analysis. We calculated lcWTSs in this paper to be able to examine where in real time coincidences (cross-correlations) between our variables of interest occur, in addition to their values or characteristic times. We calculated global wavelet cross-correlation spectra (cWTS) by integrating lcWTSs over the real time.

Values of local and global wavelet coefficients (of both auto- and cross-correlation spectra) depend by definition both on the signal and the analysing wavelet used (Perrier et al., 1995). Thus, only the resulting spectra for the very similar datasets analysed by the same wavelet functions can be compared also in terms of the values of their wavelet coefficients. In other cases, to be able to compare different wavelet power spectra, it is usually recommendable to find a common normalization, so that wavelet spectra could be viewed as relative to some norm (such as, for example, white or red noise) (Torrence and Compo, 1998). As for the wavelet spectral shapes, it

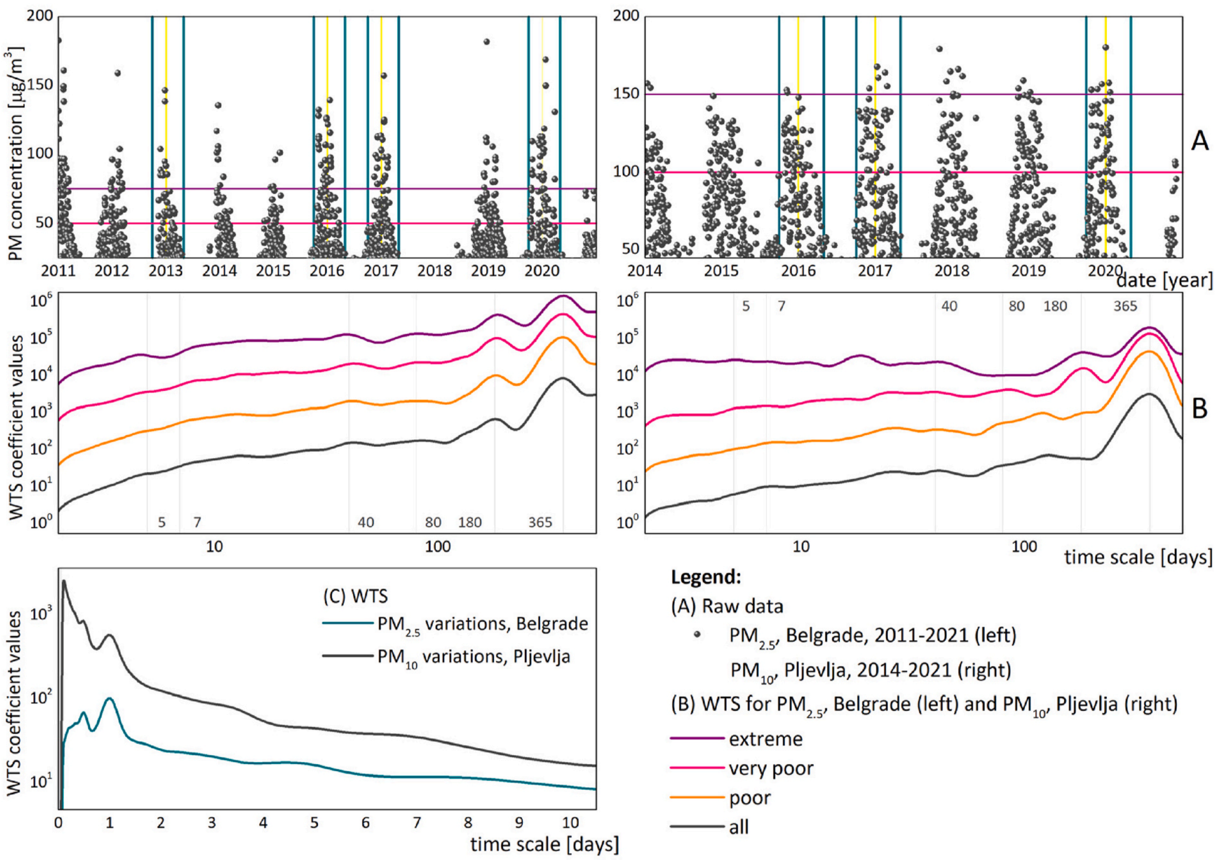

Fig. 2. (A) Raw $PM_{2.5}$ data from Belgrade (left) and PM_{10} data from Pljevlja (right), for the records that exceed WHO recommended values. Horizontal lines mark beginnings (October 1) and ends (April 30) of the heating season (teal) and beginnings of the year (January 1; yellow) and serve as visual guides. Horizontal lines mark regions of EEA classification of 'poor' air quality (below pink line) and 'very poor' and 'extremely poor' PM levels (below and above purple line, respectively). (B) WTS functions of the entire PM records for Belgrade (left) and Pljevlja (right), together with WTS functions for records with different levels of EEA 'poor' extracted, given in a log-log presentation. Vertical lines mark peaks in WTS spectra, given in days, and serve as visual guides. (C) Log-linear WTS functions of the hourly PM variations for Belgrade and Pljevlja.

has been proven (Perrier et al., 1995) that those corroborate results given by the Fourier spectral analysis, with an additional insight into the contribution to the total energy content at the local level (around the specific point x in time or space). Finally, due to the finite area below the analysing wavelet functions, the slopes of wavelet spectra can, in addition to pointing to a certain kind of harmonic behaviour, also signal existence of singularities in the signal (Torrence and Compo, 1998). In this paper we were interested in the shapes of local and global wavelet spectra and were using values of their coefficients in an absolute form, as guides to where significant changes in the signals appear.

To obtain statistically significant results and avoid finite size effects on WT statistics, we calculated WTSs and (l)cWTSs between the time scales of $a = 1$ and $a = N/10$ (Koscielny-Bunde et al., 2006), where N is the total number of data points in a series. To test the significance of peaks in WTS spectra we used significance test of Torrence and Compo (1998) against the long-range autocorrelated noise that corresponds to each of our time series. We modelled these noises following the procedure described in (Lennartz and Bunde, 2011) and compared their WT power spectra against of WTSs of our time series.

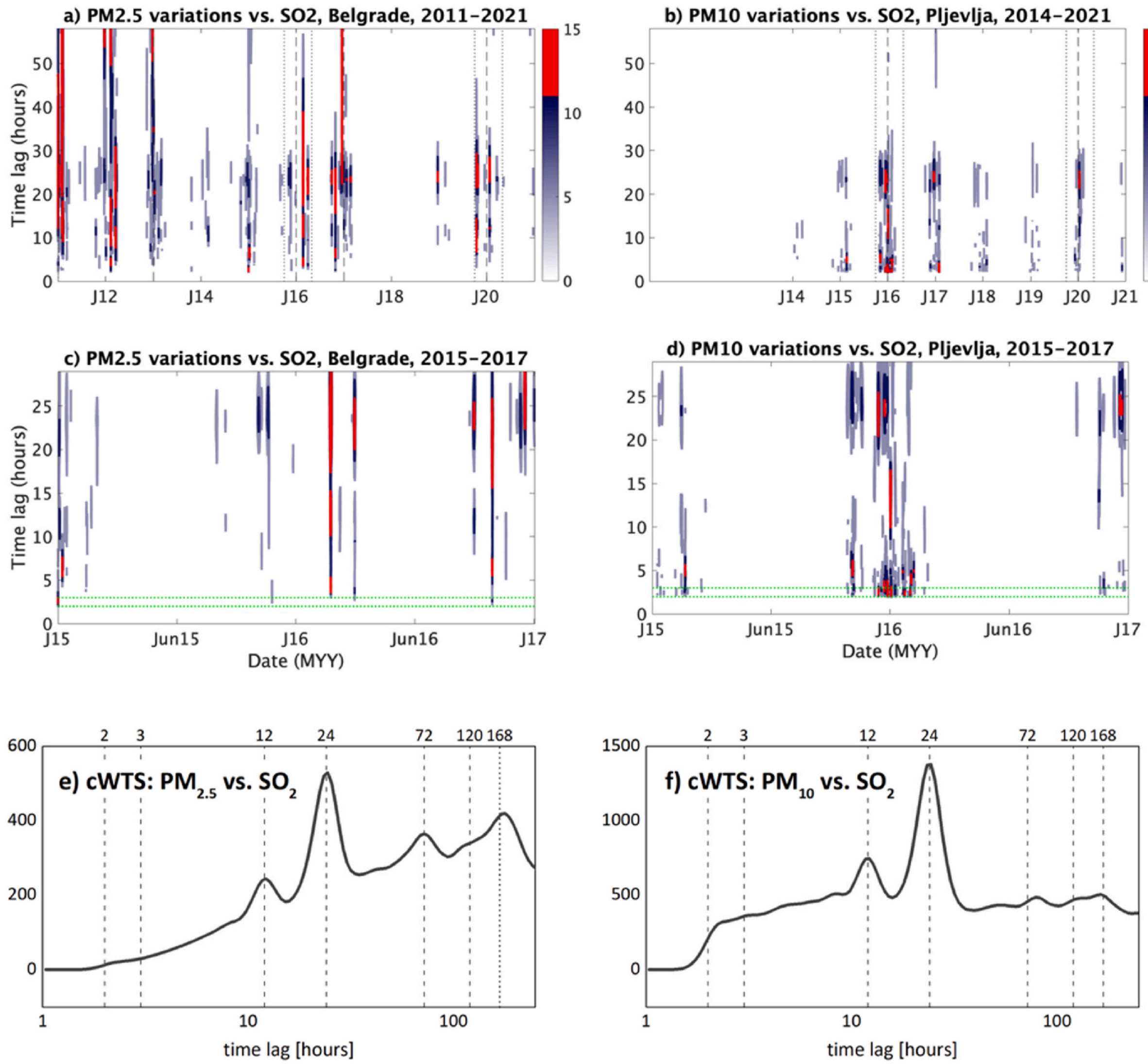

Fig. 3. (a) and (b): Local wavelet cross-correlations between recorded PM levels and SO_2 concentrations in Belgrade (a) and Pljevlja (b), for the entire recording periods. Vertical lines mark beginnings (October 1) and ends (April 30) of heating seasons (dotted grey lines) and year starts (January 1; dashed grey lines) and serve as visual guides. Colorbars code values of lcWTS coefficients. (c) and (d): Detailed view of lcWTSs given in (a) and (b) for the heating season centered around the year 2016 and a 24-h time lag. Horizontal green doted lines mark time lags of 2 and 3 h. (e) and (f): Global wavelet cross-correlation spectra between recorded PM levels and SO_2 concentrations in Belgrade (e) and Pljevlja (f), for the entire recording periods. Vertical lines mark 2, 3, 12, and 24-h and 3, 5, and 7-day time lags, and serve as visual guides.

3. Results

In Fig. 2A we present raw data of daily averages of the observational records of $PM_{2.5}$ concentrations in Belgrade, Serbia, for the period 2011–2021, and PM_{10} concentrations in Pljevlja, Montenegro, for the period 2014–2021. Raw data are given in mg/m^3 of air volume, only for records that were above the recommended WHO levels of 15 mg/m^3 for a $PM_{2.5}$ and 45 mg/m^3 for a PM_{10} daily averages (WHO, 2022; World Health Organization, 2006). It can be seen from Fig. 2A that the values of the quality of air above the WHO Air Quality Guidelines, which are considered to be of a various degrees of 'poor' by the European Environmental Agency (EEA) (EEA, 2022a), were recorded almost exclusively inside the heating season periods in Belgrade, and also during the summer months for some years in Pljevlja. These accounted for 33% of the recording period days for Belgrade (out of which 12% in the 'very poor' EEA air quality range of above 50 mg/m^3 and 5% in the 'extremely poor' EEA range of above 75 mg/m^3) with maximum concentration of 185 mg/m^3 recorded on January 1, 2011, and 38% of recording period days for Pljevlja (with 11% above EEA 'very poor' value of 100 mg/m^3 and 1% above the EEA 'extremely poor' value of 150 mg/m^3) and maximum concentrations limited by us to 300 mg/m^3 for the purpose of analyses of this paper (please see Data and Methods above).

We give the WTS spectra for daily records of $PM_{2.5}$ in Belgrade and PM_{10} in Pljevlja in Fig. 2B, separately for all records and for data with various levels of EEA 'poor' extracted. It is visible from Fig. 2B that for Belgrade the WTS spectra for the different categories of air quality data are very similar, with visible annual, semi-annual, and around 40-day variations, and slightly more pronounced short-range 5-day and 7-day peaks for the 'extremely poor' group. The 5-day variation is usually associated with the human everyday activity variations of a working week, and can generally mean that those variations result from the difference of PM concentrations during the working week in relation to weekends, or from the difference of PM concentrations at the beginning and at the end of the working week (for example, PM levels on Mondays in relation to Fridays) (Stratimirović et al., 2018; Stratimirovic et al., 2021). The appearance of a 5-day (working week) peak in energy production was already encountered by our group in our analysis of the data related to the work of hydrological power plants (Stratimirovic et al., 2021). A 7-day peak reflects weekly variations in data (such as, for example, a probable repeated change in production on Mondays). This is different for Pljevlja, where the WTS spectrum of the 'extremely poor' group is different from other spectra, with visibly elevated slope in the small-time scales area by the existence of a

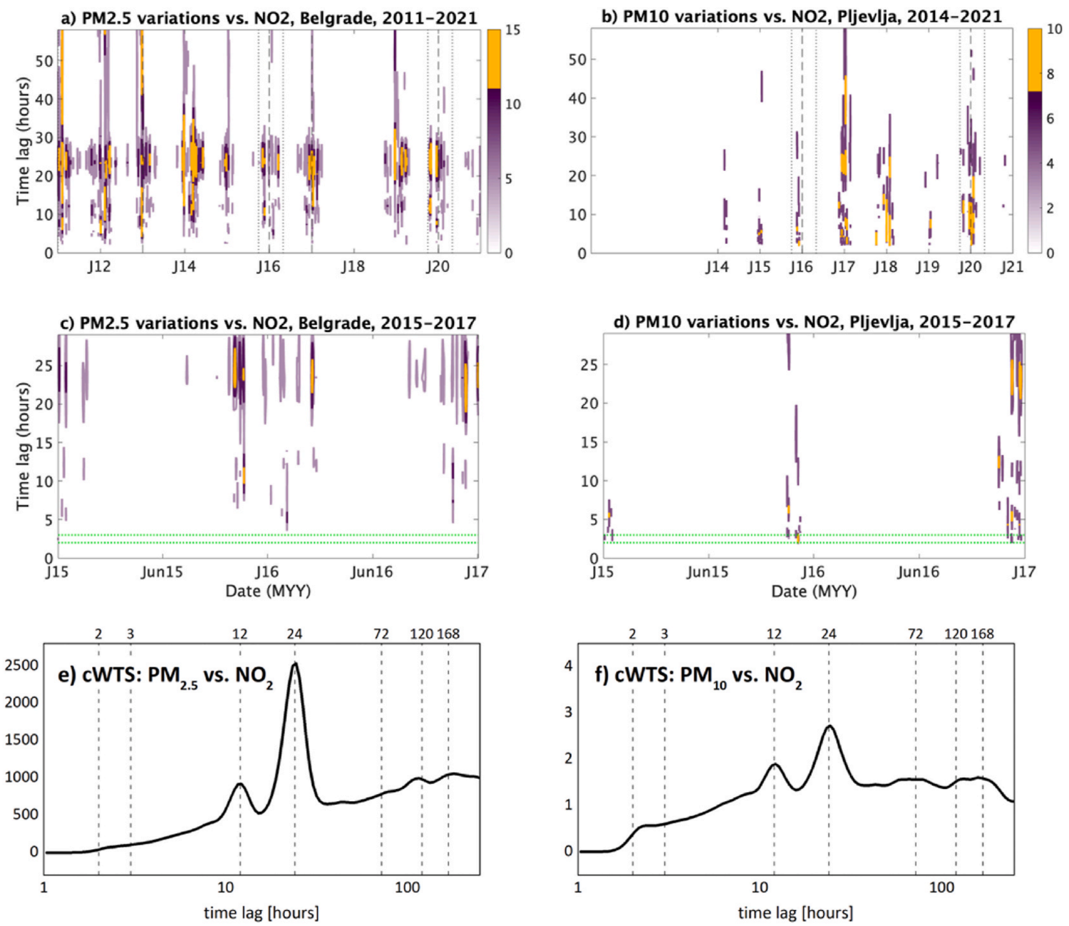

Fig. 4. Wavelet cross-correlations between recorded PM levels and NO_2 concentrations in Belgrade (a, c, e) and Pljevlja (b, d, f), presented in the same manner as in Fig. 3.

prominent peak at around 2–3 days, in addition to a working week and a one-week peaks. In addition, even if WTS spectra for the other PM concentration groups are similar, it is visible that those have differently pronounced peaks, that is that those are differently driven by various external influences, which was not the case for Belgrade. Finally, in Fig. 2C we display WTS spectra for the hourly PM variations, calculated as series of increments $\Delta PM_i = PM_{i+1} - PM_i$ ($i = 1 \ldots N - 1$, N the number of data points in a series) of the original series of hourly PM concentrations in Belgrade and in Pljevlja. It is visible from Fig. 2C that those display visible peak at a 5-day interval, as already seen in Fig. 2B, together with a prominent 12-h and 24-h peaks for Belgrade, and a 3–4 days peak, together with a visible 12-h, 24-h, and a prominent less than 12-h (around 3-h) peaks for Pljevlja. Potential sources of these high frequency variations of ΔPM_i were of interest of our analysis in the rest of the paper.

To this end we analysed WT cross-corelations of time series of hourly PM variations in relation to observed SO_2 and NO_2 concentrations, recorded at the same recording sites (and during the same corresponding time periods) in Belgrade and in Pljevlja. We present our results in Fig. 3, for PM-SO_2 wavelet cross-correlations, and in Fig. 4, for PM-NO_2 (l)cWTSs. It is visible from Fig. 3a and 4a how local cross-correlations between levels of PM and SO_2 and NO_2 are more immediate (have shorter time lags) in Pljevlja than in Belgrade, with significant lcWTS values appearing at several hours to 12 and 24 h in Pljevlja, and continuously at 12 and 24 h and at time lags of several days, in Belgrade. To examine this more closely, in Fig. 3b and 4b we provide detailed views of PM-SO_2/NO_2 lcWTSs for the heating season centered around the year 2016, and for the 24-h time lags, while in Fig. 3c and 4c we give their corresponding global cross-correlation functions for the whole recording period. It is visible from Fig. 3b and c and 4b and 4c how 2–3-h time lags prominently appear in (l)cWTSs in Pljevlja and are not present in PM-SO_2/NO_2 cross-correlations in Belgrade. This probably reflects the time needed for TPP emissions to turn into secondary sourced PM at the TPP site. It also appears from Figs. 3 and 4 that the cross-corelation patterns at all the short time ranges are more complex for the PM-SO_2 lcWTS in Pljevlja, and for the PM-NO_2 lcWTS in Belgrade, which may be an indication of a presence of other sources of NO_2 pollution that affect PM levels in that area.

It is also visible from Fig. 3a and c and 4a and 4c how cross-correlations (coincidences) between PM hourly variations and concentrations of SO_2 and NO_2 decay faster for Pljevlja. To further illustrate this, in Fig. 5 we present their lcWTS values at 3 day (72 h) and 5 day (120 h) time lags for the period 2014–2021, where we have data from both towns, in a form of overlapping coloured area charts. It can be seen from Fig. 5 how values of lcWTS coefficients are comparable at 3-day lags in Belgrade and in Pljevlja, for the overlapping makes different coloured areas almost indistinguishable. The area charts become visually separated for a 5-day lags, where lcWTS coefficient values decrease for Pljevlja and results for Belgrade are more prominent. This is particularly visible for years 2015, 2016, and 2019, and serves as an additional illustration of the effect of faster decay of PM – SO_2/NO_2 cross-correlations in Pljevlja that are already seen in graphs in Fig. 3a), b), 4a), and 4b). Since Fig. 5 is just an additional illustration of the effects already seen in Figs. 3 and 4, this visualization may be used as corroboration of the hypotheses that sources of SO_2/NO_2 pollution in Belgrade are more complex and/or more remote than those in Pljevlja. Finally, it is visible from Figs. 3–5 that the highest values of lcWTS coefficients appear in or around the month of January of each year, when recorded PM extremes are also high in both Belgrade and Pljevlja.

We examined three different possible influencing factors for these changes in PM levels: 1) the SO2 and NO2 pollution from TPP production near the two towns, as a electricity production induced driver, 2) the changes in PBLH levels, as meteorological regulator, 3) the changes in T2m also as meteorological regulator, and specifically as a proxy for presence of cold or warm weather, that corelates with pollution emissions from communal and individual heating. Finally, we used specific situation of COVID lockdown in Belgrade to examine contribution from on-road transportation to elevated pollution. In the absence of records of TPP TENT daily or hourly production, while having in mind the total share of this TPP in Serbian energy production, we chose to use publicly available ENTSO-E Serbian total energy production from coal as a proxy measurement for this TPP production. We checked the validity of this choice in the case of TPP Pljevlja and ENTSO-E Montenegrin total energy production from coal by calculating lcWTS between those two datasets

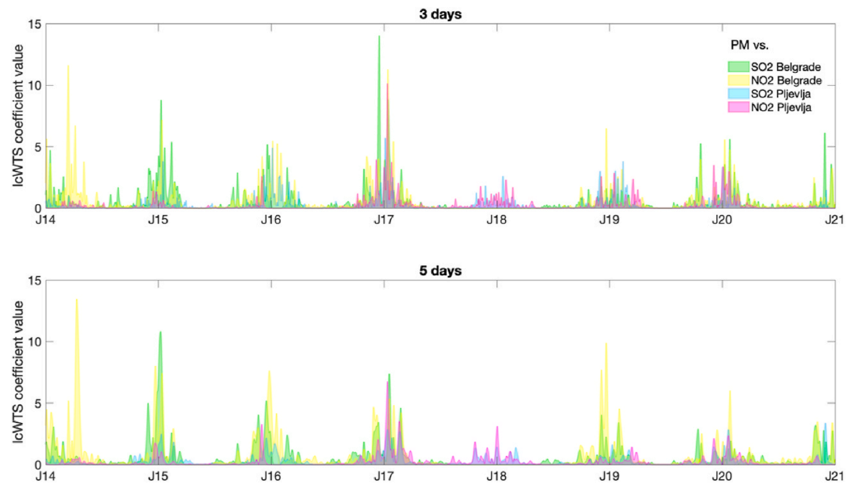

Fig. 5. Local wavelet cross-correlations spectra for coincidences between PM hourly variations and SO_2 and NO_2 concentrations in Belgrade and Pljevlja for a 3-day time lag (up) and a 5-day time lag (down) intervals.

and SO_2 and NO_2 concentrations and got virtually the same results. Those are presented in Fig. 6.

Dependences between PM variations from changes of these variables are depicted in Fig. 7, in a form of dependency (X-Y) plots. It is visible from Fig. 7 that none of these relationships are linear for either Belgrade or Pljevlja. In Belgrade, PM variations show very weak correlations with the TPP production (that is, energy production from coal) and weak anticorrelations with T2m in high and low ends of X-Y graphs respectively, showing that extreme PM values are coinciding with the highest energy production and with the lowest temperatures of the heating season, but are otherwise evenly distributed across wide ranges of different values of the TPP production and T2m. In contrast, the PM variations show clear anticorrelation with the PBLH levels in the low end of the dependency graph, with high PM levels clearly coinciding with the low PBLH values. This clear association terminates at PBLH values of about 300*m*; importantly, this association relates only to all the various values of 'poor' PM quality in Belgrade. In Pljevlja only the PM-PBLH relationship holds to an extent but is much shorter (holds for lower PBLH values of up to around 200*m*) and is visibly broaderly scattered across the X-Y plot area, while the PM-TPP and PM-T2m relations are completely lost. This is probably the reflection of the fact that the various levels of 'poor' PM values are seen in Pljevlja also outside of the heating season.

To try to examine separately urban transportation emissions as PM extremes drivers and possibly disaggregate those from communal heating sources in Belgrade, we followed the idea of (Ruberti et al., 2020) to examine air pollution during the Covid-19 total lockdown of the spring of 2020, when there was complete or substantial reduction of city road traffic, but not communal heating, during longer periods of time. We complemented these with dependency analysis of the Belgrade PM records from the number of automatically counted vehicles of the city traffic at the same recording site. The results are given in Fig. 8, for: 1) the $PM_{2.5}$ daily concentrations in the JAN-APR 2020 period compared to their previous 10-year (2010–2019) average, to enable examination of the differences in air pollution during the Covid-19 lockdown period, compared with the previous 10-year period, and 2) for recorded $PM_{2.5}$ daily concentrations against recorded daily traffic numbers, for the period 2018–2019, to examine if there was indeed visible dependence between the two, at least for the period for which we had data. It can be seen from Fig. 8 that Covid-19 lockdown PM levels remained similar to their 10-year average values in the significant absence of road transport emissions, while recorded PM data showed no visible correlation with the recorded number of urban vehicles.

Following findings presented in Figs. 7 and 8, in Fig. 9 we provide raw PBLH records for Belgrade and Pljevlja, together with maps of spatial composites (averages) of meteorological spatial fields for differences of PBLH levels during the heating season (JAN-APR and OCT-DEC) and average PBLH levels for each year, from 2014 to 2021 (period for which we have data from both towns), over the entire area of Western Balkans that encompasses both Montenegro and Serbia. Spatial composites are calculated as differences between the PBLH average values for the heating season and the PBLH average values for the two years that encompass this heating season. Raw data are given only for PBLH values that were equal or lower than 300*m* in Belgrade and 200*m* in Pljevlja, as distinguished as transition from anticorrelated to non-correlated dependence in Fig. 7C and D. It is visible from Fig. 9 that heating seasons are characterized by spatially homogeneous anomalies in the PBLH levels across the region, indicating that large-scale weather patterns may affect and favour aggregated urban PM extremes that we analysed.

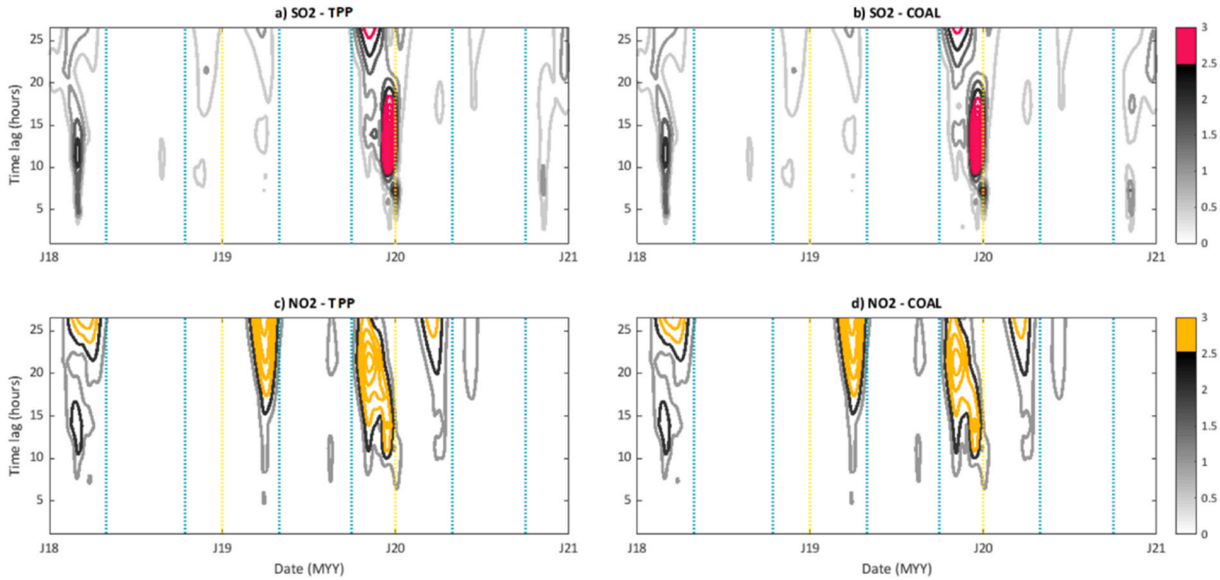

Fig. 6. Comparison of the lcWTS results for the SO_2 – TPP (a) and SO_2 – national (Montenegrin) energy production from coal (b), and NO_2 – TPP (c) and NO_2 – national energy production from coal (d), for the daily averages data for Pljevlja, for the period 2018–2021. Vertical blue and yellow dashed lines give the boundaries of the heating season and year starts, respectively.

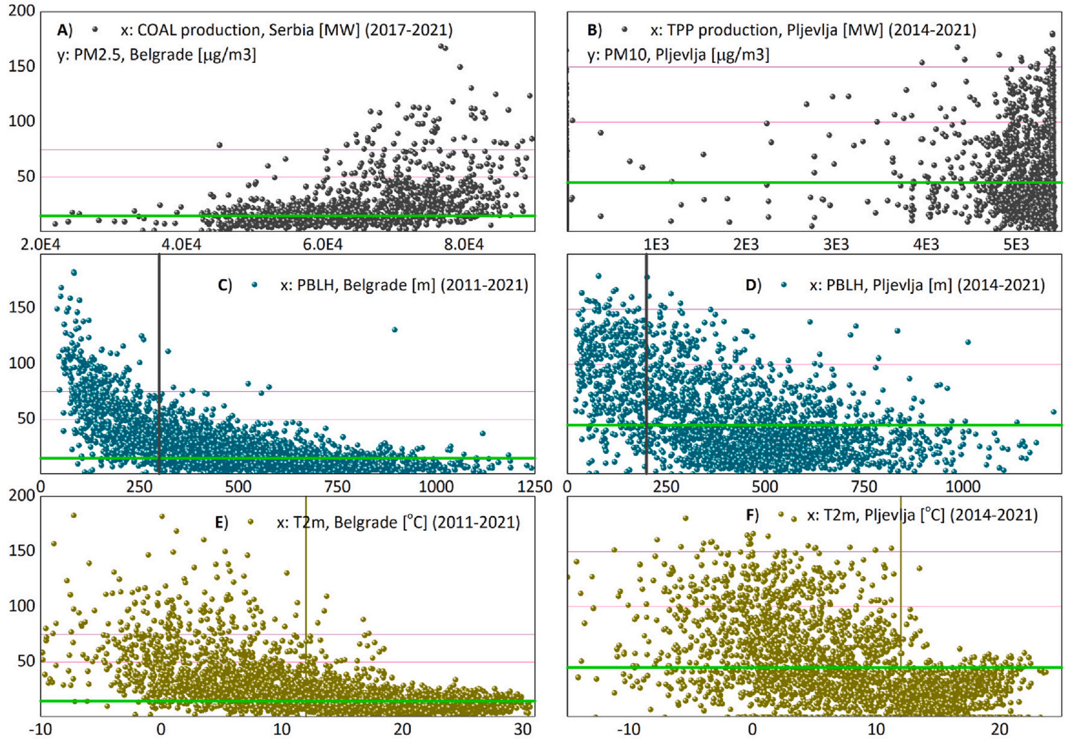

Fig. 7. Dependency plots of (A) and (B) PM-TPP (or energy) production, (C) and (D) PM-PBLH levels, and (E) and (F) PM-T2m variability in Belgrade (left column) and Pljevlja (right column). Horizontal lines mark region ends for WHO recommended PM levels (green tick line), and EEA 'poor' (pink line) and 'very poor' (purple line) PM levels. Vertical lines mark 300*m* and 200*m* PBLH level respectively in (C) and (D), and 12 °C in (E) and (F).

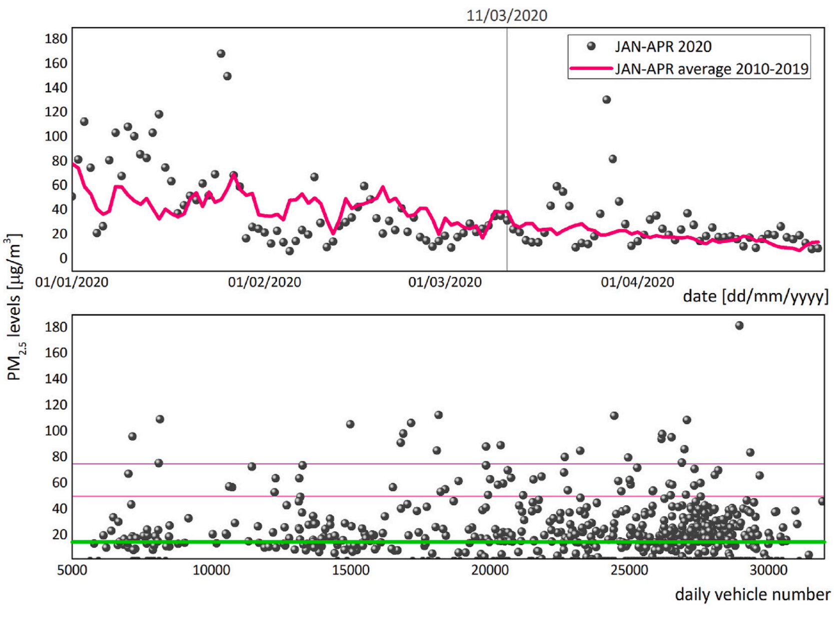

Fig. 8. (up) $PM_{2.5}$ daily concentrations during the JAN-APR 2020 period (dark grey dots) compared to the JAN-APR $PM_{2.5}$ daily concentration averages for the preceding 10-year period (pink line). (down) Dependency plot of the daily PM-traffic number correlations for Belgrade, in the period 2018–2019. Horizontal line marks region ends for WHO recommended PM levels (green tick line), and EEA 'poor' (pink line) and 'very poor' (purple line) PM levels.

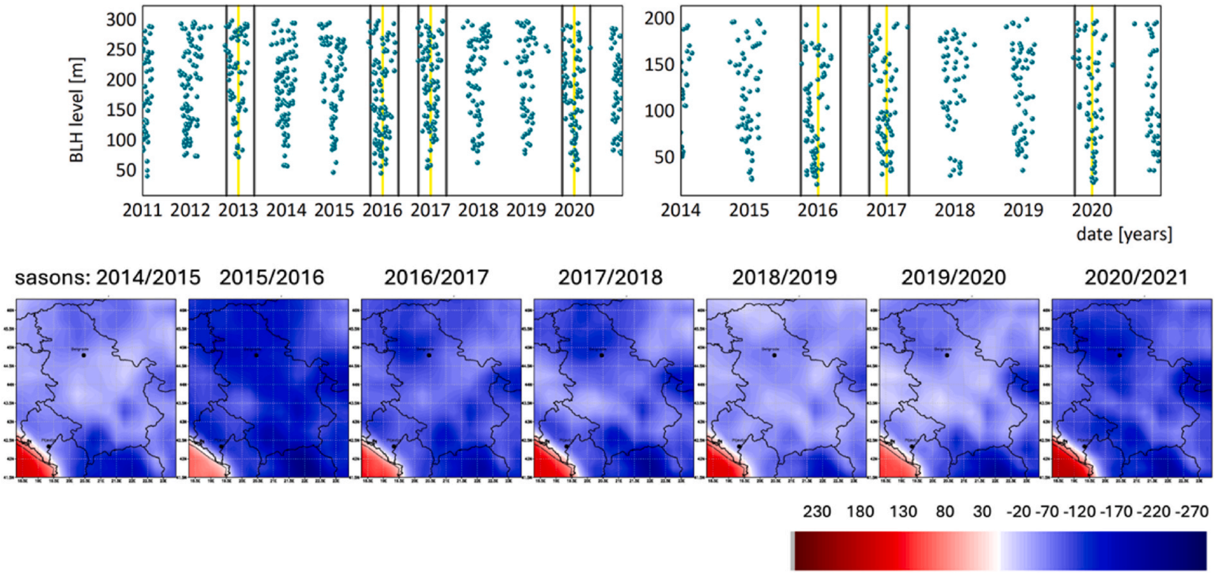

Fig. 9. Raw data of PBLH records for Belgrade (upper raw left) and Pljevlja (upper raw right), with spatial composites of the difference between the average PBLH level during the heating season and the average for the given year (colorbar in *m*, lower row). Raw PBLH data for Belgrade are given for the period 2011–2021, while the raw PBLH records for Pljevlja and PBLH spatial composites are given for the period 2014–2021. Vertical lines in upper row graphs are given as visual guides, and mark beginnings (October 1) and ends (April 30) of the heating seasons (dark grey lines) and year starts (January 1; yellow lines) for several years. Locations of Belgrade and Pljevlja (red dots) are given in the lower row maps.

4. Discussion

In this paper we performed time series analysis of the observed and modelled meteorological data and energy production records from the two TPPs that are major sources of energy production in two countries in Western Balkans, to examine combined human-made and climatic drivers of air pollution extremes in nearby urban areas. To this end we firstly preformed WT analysis of daily averaged PM records from the city of Belgrade, viewed as a large urban area, in the vicinity of TPP Nikola Tesla in Serbia and from the town of Pljevlja, viewed as a small urban area or an urban cluster, in the very near vicinity of TPP Pljevlja in Montenegro. We showed that the extreme PM values in Belgrade appear only during the wintertime, or more precisely during the OCT-DEC and JAN-APR heating season, and that the WTS functions of those extremes follow the shape and slope of the WTS function of the all-PM values records. Those display characteristic times of autocorrelations (peaks) at 40 days, 80 days, half a year and at annual intervals, at the high time scales, and peaks at working week duration and a week duration (5 and 7 days) in the small time scales area. Working week and one week peaks are usually seen and interpreted as signs of human activity (Stratimirović et al., 2018), including in the energy sector (Stratimirovic et al., 2021), and in our Belgrade sample those are more pronounced for the WTS of PM extremes than for the all-PM data time series, showing that external sources of these variations are more dominant for PM extremes. They can be related to both energy production weekly regimes at TPP TENT and weekly regimes of fuel combustion for heating, combined with urban vehicle emissions in Belgrade. The WTS of recorded hourly PM Belgrade data shows two more prominent peaks in the very small-time scales area, at 12-h and at 24-h intervals, probably reflecting daytime and nigh time differences in PM values (Brock et al., 2002).

In slight contrast to Belgrade data, extreme values of daily PM averages in Pljevlja appear also outside the OCT-DEC and JAN-APR heating season period, while WTS functions for their extremes present with the different slope than for all-PM records. Wavelet spectra for PM extremes show visibly more prominent small time scales peaks at 5-day, 7-day, and even less than 5-day intervals that change WTS slopes. This suggests that weekly, or even shorter (weekend), human-related activities almost completely dominate extreme PM daily dynamics in Pljevlja. Furthermore, WTS of the PM hourly variations show the existence of a dominant 2- to 3-h peak, that may reflect the time needed for TPP emissions to turn into PM particles in the close vicinity of the TPP Pljevlja. This result is in line with previous observations from plumes of the coal fuelled power plants in the United States (Brock et al., 2002).

The local and global wavelet cross-correlation analysis that we conducted between the hourly PM variations and recorded hourly levels of SO_2 and NO_2 in two towns furthermore confirmed that the very short PM WTS variations in Pljevlja are most probably caused by PM-SO_2 and to some extent PM-NO_2 coincidences after a 2- to 3-h time lag needed for transformation of SO_2/NO_x TPP emissions into PM particles around TPP Pljevlja. The PM-SO_2/NO_2 local and global cross-correlation WT spectra for Pljevlja overall show quite a simple structure – in addition to 2–3 h changes, only other visible coincidences appear at time lags at approximately 12 and 24 h, after which time scales lcWTS and cWTS coefficients decline visibly. This is not the case for Belgrade, where both PM-SO_2/NO_2 (l)cWTSs display much richer temporal structure, with (l)cWTS coefficients appearing at time ranges from several hours to several days, probably reflecting effects of different other causes on PM variations than the TPP production alone. Temporal diversity of the time scale PM-SO_2/NO_2 local coincidences may also include the time needed for TPP TENT PM emissions to reach Belgrade under relatively stable weather conditions (Brock et al., 2002), which is not the case in Pljevlja.

The difference between the relative simplicity of Pljevlja lcWTSs and more complex corresponding functions for Belgrade is particularly visible for the PM-NO_2 cross-correlations. Since possible other emission sources in Belgrade appear inside the heating season exclusively, those are probably related to other energy sources, including, or most probably, fuel burning for residential and industry heating. Our examination of daily PM Belgrade concentrations during the JAN-APR 2020 period in that regard confirmed the results of the recent regional study of the emissions during Italian Covid-19 total lockdown (Ruberti et al., 2020), which included some industries closure and significant reduction of the on-road transportation. This study found that heating household systems rather than road traffic are probably the main sources of high levels of the NO_2 pollutant in cities with high population densities that leads to massive use of domestic heating systems and significant number of cars per km^2 and called for the reconsideration of urban containment measures concerning the blocking of traffic, claiming that those may not be sufficient to lower the concentration peaks of NO_2 in large urban areas or highly industrialized sites (Ruberti et al., 2020). Our findings from Belgrade PM-city road traffic dependency analysis provide an additional data-based evidence to that claim for our data were, unlike in studies of Covid-19 lockdown, collected in times of normal, including peak daily or seasonal on-road city traffic.

Our results that concern dependences between the observed PM values and their possible anthropogenic (TPP production and T2m as a proxy measure for communal and individual heating) and meteorological (PBLH) drivers showed that TPPs TENT and Pljevlja, even if they are major sources of harmful emissions and thus major sources of PM concentrations in the two towns, are probably not meaningful sources of PM variations, especially in Pljevlja. These TPPs are inert sources of energy production that emit continually and have no significant hourly or daily changes in production and emissions, other than those that can be related to the quality of used coal, which are probably minor in this context. Significant effects of TPPs to PM changes could be investigated only during times of power plants' total or significant closures, due to different reasons such as repair, accidents, or production plans, using the event-based statistical approaches (Donges et al., 2016). From our findings it appears that similar conclusions as for TPP drivers could be drawn for industrial and residential heating as important sources of emissions that lead to high PM concentrations – those do not seem to influence short-term PM variations, also especially in Pljevlja. In our sample PBLH was the only variable that visibly influenced PM changes in Belgrade particularly, and to extent also in Pljevlja. In both towns in our analysis changes in PBLH coincided with the drive to extremes in PM values. Following these findings PM extremes in our sample could be viewed as preconditioned compound events (Bevacqua et al., 2021; Zscheischler et al., 2020), where TPP and urban heating emissions provide preconditions for PM extremes and PBLH serves as at least one meteorological driver to such events. PBLH was reported before as a key driver of wintertime PM extremes across the United States (Porter et al., 2015), or as the modulator of the urban summertime PM variations in China (Miao et al., 2017). Other possible meteorological drivers that influence air quality sensitivity to meteorology and that are often analysed include humidity (specific and relative), wind speed, precipitation, atmospheric pressure, cloud fraction and radiation (Chen et al., 2020). Since we did not have access to these variables, due to the very strict policies of the meteorological services responsible for the meteorological observations, in this work we limited ourselves to only one directly measured variable. Following this fact analysis of the impact of other drivers remain to be investigated in the future research, by other groups or by us. It additionally remains to be examined whether these other meteorological drivers act as dominant or minor concurrent (with PBLH) drivers of PM extremes in such cases (Bevacqua et al., 2021), to guide their future statistical modelling.

Regarding the longer characteristic time intervals of PM (including extreme PM) WTS variability in Belgrade and Pljevlja of around 40 and 80 days, those could arise as effects of both human-made and climatic drivers. We encountered the 40-day cycle in our previous work that related to the effects of damming to changes of river natural cycles (Stratimirovic et al., 2021), where we argued that possible climatic sources of such variation could be the two climatic modes of oscillation with periods near 48 and 23 days that exists in the Northern hemisphere (Ghil and Mo, 1991), or a 35- to 40-day oscillation, characterized by blocking structure over Eurasia continent (Zhang et al., 1997), along with intraseasonal oscillation in tropics such as the Madden–Julian Oscillation. On the other hand, the longer intervals of 40 and 80 days might result from forecasts, planning, or even separation (Varotsos et al., 2014) between the acute events that are used by TPP operators to adjust the operation of TPPs in the overall hydro-thermal energy coordination, with possibility to include variable renewable energy sources (Stratimirovic et al., 2021).

In our previous work, using the South African health, climate and environmental dataset, we showed that the change in instances of hospital admissions for respiratory diseases, including pneumonia and asthma, follow the change in PM levels after a time period of 10–15 days (Kapwata et al., 2021). Further work is needed to confirm this time estimate; nevertheless, in the case it is used as an approximate temporal parameter that links air quality changes with their health outcomes, our findings here may candidate PBLH as an important indicator of PM changes that lead to extreme PM variations. Information of PBLH changes could then be used as a measure to guide early warning systems and predictions of occurrence of PM extremes that inform public health preparedness and responses while continuity and level of the TPP energy production and communal (heating) wintertime pollution remains unchanged (JICA, 2022; Spinoni et al., 2018). It can also provide a better focus on time periods for respiratory disease and other health outcomes change monitoring, to further inform public health, energy sector and public policy interventions.

5. Conclusions

In this paper we were interested to better understand combined anthropogenic and meteorological drivers of urban PM extremes in the vicinity of thermoelectric power plants (TPPs) in the Western Balkans. To this end we used wavelet transform analysis, combined with the dependency analyses, to investigate relations between the TPP energy production levels or alternatively coal production for energy use records, SO2 and NO2 pollution levels, and temperature and planetary boundary layer height (PBLH) values with recorded levels of PM2.5 and PM10 particles in the air of cities of Belgrade (Serbia) and Pljevlja (Montenegro). The TPPs in the vicinity of these cities were used as examples of the outdated, but still active, high polluters in the Western Balkans.

We confirmed strong correlations between SO2 and NO2 TPP emissions and changes of urban PM values and provided a time estimate of 2–3 h needed for transformation of SO2/NO2 TPP emissions into PM particles in towns in the vicinity of TPPs. In our sample this was more prominent for the city of Pljevlja, which is a much smaller urban area than the city of Belgrade, with a TPP much closer to the city centre.

We found that in both our case studies, and especially in the city of Belgrade, temperature and the planetary boundary layer height (PBLH) exuberated the effects of the TPP pollution. We provided an estimate of critical PBLH height in this context to be 300m and lower, for our sample. These PBLH values coincide with the duration of the heating season, when other emission sources contribute to the PM extremes in both towns. We examined dependence of the elevated levels of urban PM on the on-road traffic and showed that it may not be the main additional source of complex urban emissions.

Based on our results, we suggested that PM extremes in our sample could be viewed as preconditioned compound events, where TPP and urban heating emissions provide preconditions for PM extremes, with PBLH as a major contributing meteorological driver. The study highlights the implications for the Western Balkan (and potentially countries with similar climatology) public health systems, emphasizing the need for efforts to control wintertime emissions. The critical role of PBLH could provide this region with an early warning system for extreme urban pollution and thus assist public health preparedness and messaging. Future studies should include examination of other meteorological and environmental drivers of the extreme pollution in the vicinity of TPPs, to better understand, model, and forecast its onset.

CRediT authorship contribution statement

Ana Gardašević: Writing – review & editing, Writing – original draft, Visualization, Formal analysis, Data curation. **Neda Aleksandrov:** Writing – review & editing, Writing – original draft, Visualization, Formal analysis, Data curation. **Ilija Batas-Bjelić:** Writing – review & editing, Conceptualization. **Ivan Bulatović:** Writing – review & editing, Supervision, Resources, Conceptualization. **Vladimir Djurdjević:** Writing – review & editing, Writing – original draft, Supervision, Resources, Methodology, Investigation, Funding acquisition, Conceptualization. **Suzana Blesić:** Writing – review & editing, Writing – original draft, Visualization, Validation, Supervision, Methodology, Investigation, Funding acquisition, Formal analysis, Conceptualization.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Data availability

Data will be made available on request.

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