Long-term Characterization of Urban PM₁₀ in Hungary

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ABSTRACT

Over urban areas in Hungary, the annual average PM₁₀ concentrations are not frequently higher than 40 µg m⁻³. Despite the mitigation efforts of the local governments, the annual number of exceedances of the daily limit of 50 µg m⁻³ is higher than what is outlined in EU Directive No 2008/50/EC. The goal of the present study is to assess the characteristics of the temporal (annual, seasonal, daily) variations in PM₁₀ concentrations in selected Hungarian cities with large populations, where most of the exceedances have been reported. The impacts of meteorological conditions on the measured PM₁₀ concentrations and their temporal variations are also evaluated. An important aspect of studying the trends of air pollution is that the tendencies depend not only on the emissions of certain pollutants but also on the meteorological conditions in the area of interest. To analyse emission-related trends, the meteorological signal must be removed from the data series. In this study, the Kolmogorov-Zurbenko (KZ) filter was used for this type of trend separation. Moreover, multiple nonlinear regression analysis was used to find relationships between the PM₁₀ concentration and several meteorological parameters. The goal of this analysis is to estimate the expected daily mean PM₁₀ concentration values. The results of this analysis demonstrate that the regression equation can provide an adequate method for PM pollution forecasting. In addition to the hourly PM₁₀ concentrations and basic meteorological data, global radiation and boundary layer height were considered in the characterization process.

Keywords: PM₁₀, Cold season episode, Regression analysis, Kolmogorov-Zurbenko (KZ) filter

1 INTRODUCTION

Currently, particulate matter (PM) is one of the most critical atmospheric pollutants since it has negative impacts not only on human health but also on environmental issues such as visibility, infrastructure, and ecosystems (Horvath, 1995; Lim *et al.*, 2012; Fang *et al.*, 2013; Kuzmichev and Loboyko, 2016; Tidblad *et al.*, 2017; De Marco *et al.*, 2019).

The primary sources of the particles are highly variable in intensity, space and time, and emitted particles of different sizes and compositions have widely different fates and effects depending on atmospheric conditions. In addition, aerosols can be formed in the atmosphere (secondary aerosols), adding to the immense complexity of the PM issue. It is therefore a challenge to reduce PM emissions (EU Directive, 2016) and the associated risks to human health from exposure to PM. A better understanding of the status of and trends in air quality in Europe is crucial to support European, national, and regional governments in policy-making and implementation, as well as to improve the tools of air quality assessment and management (Guerreiro *et al.*, 2014).

PM is a complex mixture of solid- or liquid-phase primary and secondary particles. Their physical and chemical properties, such as shape, size, solubility, residence time, toxicity, and chemical composition and structure, vary greatly. Among them, the chemical composition and

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particle size are key factors in understanding the behaviour and role of the particles in atmospheric processes. These characteristics impact human health; they control aerosol optical properties and hygroscopicity, playing a substantial role in the atmospheric radiation budget and cloud formation (Seinfeld and Pandis, 2012). Local and regional meteorological factors, including wind speed, wind direction, vertical atmospheric stability, long-range transport, and pollution dispersion, play an important role in determining PM concentrations. For this reason, the analysis of local and regional meteorology is important to better understand the processes that are responsible for the spatial and temporal distribution of PM (Demuzere *et al.*, 2009; Velders and Matthijsen, 2009; Pearce *et al.*, 2011; Barmpadimos, *et al.*, 2012; Chen *et al.*, 2018).

In Europe, the emissions of many primary air pollutants have decreased substantially over the past decades, resulting in improved air quality across the region (European Environment Agency, 2019). Nevertheless, several emission sources remain for which much less progress has been achieved. Examples are biomass burning and residential heating or emissions from agricultural activities, all of which are significant primary sources of particulate matter in Hungary. Non-industrial sources account for more than 50% of total emissions, mostly from residential heating (Kis-Kovács *et al.*, 2019). Resuspended particles from agricultural operations, bare soils, or road traffic are also considered uncontrolled and significant sources. The time series of total emissions by sector (GNFR – gridded nomenclature for reporting) in Hungary between 2007 and 2017 does not show a definitive trend.

In urban areas, residential combustion and traffic-related emissions (exhaust and non-exhaust) are the main primary sources of PM. Particulate matter from roads (resuspended by wind or other anthropogenic activities) significantly contributes to urban PM₁₀, e.g., up to 30% in Veszprém, Hungary (Jancsek-Turóczy *et al.*, 2013). Residential wood combustion is the largest single source of organic aerosols. The aerosol emissions from this source are still significantly underestimated in Europe (van der Gon *et al.*, 2015). According to Karagulian *et al.* (2015), in Central and Eastern Europe, domestic fuel burning (wood, coal and gas) might contribute up to 45% of PM₁₀ emissions (while globally, its share can reach 15%).

PM is a complex mixture of solid- or liquid-phase primary and secondary particles. Their physical and chemical properties, such as shape, size, solubility, residence time, toxicity, and chemical composition and structure, vary greatly. Among them, the chemical composition and particle size are key factors in understanding the behaviour and role of the particles in atmospheric processes. These characteristics impact human health; they control aerosol optical properties and hygroscopicity, playing a substantial role in the atmospheric radiation budget and cloud formation (Seinfeld and Pandis, 2012). Local and regional meteorological factors, including wind speed, wind direction, vertical atmospheric stability, long-range transport, and pollution dispersion, play an important role in determining PM concentrations. For this reason, the analysis of local and regional meteorology is important to better understand the processes that are responsible for the spatial and temporal distribution of PM (Demuzere *et al.*, 2009; Velders and Matthijsen, 2009; Pearce *et al.*, 2011; Barmpadimos *et al.*, 2012; Chen *et al.*, 2018).

According to Barmpadimos *et al.* (2011), in Switzerland, PM₁₀ concentrations are affected by wind gusts, temperature, the precipitation rate and the boundary layer depth. Dunea *et al.* (2015) reported that there were negative correlations between PM₁₀ and temperature and solar radiation, while relative humidity and atmospheric pressure were positively correlated with PM₁₀. Vardoulakis and Kassomenos (2008) found a significant correlation between PM₁₀ and solar radiation during cold seasons, and these correlations became weaker during warm seasons. Most of these papers finally concluded that in addition to changes in anthropogenic emissions, changes in meteorology can be responsible for long-term changes in PM₁₀ concentrations.

In Hungary, three important factors have essential effects on PM₁₀ concentrations: local anthropogenic emissions, long-range transport (sources outside of the country) and meteorological conditions. It has been estimated that the background annual average PM₁₀ mass concentration for continental Europe is $7.0 \pm 4.1 \mu\text{g m}^{-3}$ (Van Dingenen *et al.*, 2004), and the natural contribution to PM can range from 5% to 50% in different European countries. The effects of natural sources on PM₁₀ concentrations are almost negligible in Hungary (Diapouli *et al.*, 2017). The most important local emissions that have significant effects on the PM_{2.5} concentrations at an urban background site are traffic (19%) and biomass burning (24%) (Perrone *et al.*, 2018), while the impacts of sources originating from the transboundary and countryside are significant, more than 30% (Thunis

et al., 2017). The impact of traffic on PM₁₀ is continuous throughout the year; however, the effect of residential heating is considerable only in winter; otherwise, it can be neglected. During spring and fall, depending on the local weather conditions, inhabitants use combustion equipment mostly at night. In winter, the effect of residential heating must be taken into account throughout the day.

In our previous work (Ferenczi and Bozó, 2017), the effect of transboundary sources on the air quality of the greater Budapest area was evaluated. We found that the contribution of long-range (transnational) atmospheric transport to the annual cumulative PM₁₀ concentrations in Hungary was remarkably high (higher than 50%). However, during the winter, late fall and early spring, episodes of very poor air quality (exceedances of PM₁₀ limit concentrations) were always caused by local (regional) emission sources in combination with special and highly unfavourable meteorological conditions—such as extensive cold air cushions—which prevent mixing and dilution of air pollutants. Usually, this type of meteorological condition is frequently associated with low ambient air temperatures, which further increases the use of solid fuels (wood and coal) and leads to an increase in PM₁₀ emissions from domestic heating (Tarrason *et al.*, 2018; Ferenczi *et al.*, 2020). These synoptic conditions favour smog formation, and the effect of long-range transport is negligible compared to the role of local sources and processes. These events are usually terminated by markedly changing meteorological conditions (intensive horizontal and vertical mixing of the air), which restore the importance of long-range transport in determining PM₁₀ concentrations.

The main objectives of the present work are to determine the spatial and temporal variability of the PM₁₀ concentration in Hungarian cities, to separate the effects of emission sources and meteorology on PM₁₀ concentrations, and finally to investigate the relationships between the PM₁₀ level and meteorological parameters to identify the most important parameters that can be used in PM₁₀ prediction.

2 MATERIALS AND METHODS

2.1. Monitoring Sites

Three monitoring stations with significantly different characteristics (population, type of station) were selected for the detailed analysis of three cities, Budapest, Miskolc and Pécs. In these cities, exceedances of PM₁₀ limit concentrations have been reported most often (Fig. 1). The data availability of hourly PM₁₀ concentrations is sufficiently high at all three stations for statistical trend analysis.

At several locations in Budapest (525.1 km², 1,752,286 inhabitants), the monitoring of PM₁₀ with fine temporal resolution started in 2007. The air quality monitoring network of Budapest consists of 12 stations that are classified as traffic (4), industrial (3) or background (5). Among the monitoring sites, the Gilice tér urban background station (located in the SE part of Budapest) was selected for our analysis, which is a standard meteorological and air quality monitoring station providing PM₁₀ concentrations and detailed meteorological observations with good data coverage. This location is in the area of the Marczell György Main Observatory of the Hungarian Meteorological Service. The classification of this air quality monitoring site is suburban with a significant influence from major sources from the greater Budapest area.

Miskolc (236.7 km², 159,000 inhabitants) is represented by the Búza tér station. The air quality monitoring network of Miskolc consists of 3 stations that are categorised as traffic (1) and background (2). The classification of the site is urban traffic with a significant contribution from traffic-related sources. Moreover, the whole city is located in an unfavourable geographical location in the valley of the Sajó River surrounded by the Bükk Mountains. Its special orography contributes to the development of long-lasting (several days up to weeks) and severe air pollution episodes.

In Pécs (162.8 km², 148,000 inhabitants), the selected station (Boszorkány utca) is located in a suburban environment. The air quality monitoring network of Pécs consists of 3 stations that are classified as traffic (1) and background (2). The hourly PM₁₀ data for our complex analysis have been available since 2009. One of the major industrial emission sources in this area is a coal-fuelled power plant equipped with two modern electrostatic precipitators. This development

Fig. 1. Number of exceedances of the threshold value ($50 \mu\text{g m}^{-3}$) for the number of daily averages as a function of the season (green-spring, orange-summer, yellow-fall, blue-winter) (2007–2017). Dotted line indicates the limit value (35 days).

further decreases the PM_{10} emissions in the city. However, compared to Miskolc, the city of Pécs has more favourable orography: the northern part of the city is bordered by the Mecsek Mountains, but the southern side is open and flat.

2.2. Monitoring Methods

The evaluation of the air quality in Budapest, Miskolc, and Pécs is based on data from the Air Quality Monitoring Network of Hungary and meteorological observations from the Hungarian Meteorological Service. Calibration procedures and measurements of standards are used for air data quality control. The instruments and measurement methods fulfil the requirements of the pending EU standards. The concentration of PM_{10} was measured with different types of monitors based on the β -attenuation method. The flow rate of the PM analysers was calibrated twice a year, and the mass measurement was calibrated once a year.

To obtain reliable PM_{10} data, regular systematic quality assurance and quality control, standardized operating procedures and intercomparison measurements were performed. The PM_{10} data used in this study were collected and managed (validated) by the Hungarian Meteorological Service, Air Quality Reference Centre.

2.3. Data Analysis

All monitoring data were subdivided into four periods, spring (1 March–31 May), summer (1 June–31 August), fall (1 September–30 November) and winter (1 December–28 February), to account for the seasonal factors influencing particulate levels. The 1-hour data (PM_{10} concentrations and meteorological parameters) were averaged over 24-hour periods when at least 75% of 1-hour data were available for each time period. The averaged diurnal variations in PM_{10} concentrations at the monitoring stations were determined to build diurnal profiles of PM_{10} emissions factors used in chemical transport models in the future (Menut *et al.*, 2012).

Initially, temporal variation analysis was carried out, then the Kolmogorov-Zurbenko (KZ) filter was used to spectrally decompose the pollutant time series, and finally, multiple regression analysis was performed to find the relationships between PM_{10} concentrations and several

meteorological parameters. In the temporal variation analysis, 1-hour and, in other cases, 24-hour data were used. The KZ filter is suitable for separating both long- and short-term variations in time series. There are many similar filtering techniques, such as the anomaly technique and the wavelet transform, which can be used to separate the short-, seasonal- and long-term components of air quality data. However, the KZ filter has several advantages: it can be applied to a time series even if it includes missing data, it is an established method that has been widely applied, and it is relatively simple to use.

Finally a multiple nonlinear regression analysis was carried out to find relationships between the PM₁₀ concentration and several meteorological parameters. This technique has been widely used for PM₁₀ forecasting especially in urban areas. The statistically significant variables were selected for the PM₁₀ prediction model.

3 RESULTS

3.1. Temporal Variation in the PM₁₀ Concentration

In this section, the temporal variation in PM₁₀ at the three stations is discussed. The annual PM₁₀ mean values were examined in the period from 2007–2017 (2009–2017 in the case of Pécs). Comparing the sites, we can conclude that the PM₁₀ values are generally the highest in Miskolc and lowest in Pécs, while Budapest is characterized by an intermediate level of air pollution. In 2011, PM₁₀ peaked at all stations; not surprisingly, the winter this year was unusually cold; the number of frosty days was the highest (102 days on average) during the entire time period (2007–2017). A local maximum was observed in 2017; this year, two extensive poor air quality episodes (affecting an extended region in Europe (Tarrason *et al.*, 2018)) occurred from 20th to 30th January and from 9th to 17th February 2017. The event in January also included a sea salt episode (Tarrason *et al.*, 2018). Both events were associated with high background PM_{2.5} concentrations and an important contribution from long-range transport (Tarrason *et al.*, 2018). Hungary was also part of both events that had an effect on the annual average PM₁₀ concentrations in 2017.

The number of exceedances of the EU air quality standard for daily average (the threshold of 50 µg m⁻³ for the 24-hour average PM₁₀ mass concentration) is also reported (note: the standard should not be exceeded more than 35 times during a year (EU Directive, 2008)). Unlike the annual average PM₁₀ limits, this threshold was exceeded in almost all years in Budapest and Miskolc. Not surprisingly, most of the exceedances occurred during the winter and fall months (Fig. 1). The data in Fig. 1 show that at all stations, the number of exceedances exhibits a slight downward trend. Only two years, 2011 and 2017, are out of line, which is consistent with our previous explanation.

In Fig. 2, the average diurnal variations in the PM₁₀ concentrations on weekdays and weekends are presented. In summer, the PM₁₀ concentrations tend to be somewhat lower during weekends than during weekdays, while at night, the concentrations are rather similar. At the Miskolc site, the impact of transport is dominated on PM₁₀, since this station is a traffic site as clearly indicated by the morning rush hours on weekdays. At the other two sites, the effect of the morning rush hour on the PM₁₀ concentration is negligible compared to the evening PM₁₀ maximum since these two sites correspond to urban background monitoring sites.

In winter, a significant difference in the PM₁₀ concentration was found when working and weekend days were compared. In Miskolc, the impact of residential heating is comparable to that of traffic emissions (EMEP/CEIP), which is the reason why the effect of morning rush hour is less important. Independent of the seasons, at all stations, an evening peak is typically found probably due to the unfavourable dispersion situation in this part of the day. The evening peak in winter is much higher, presumably as a result of indoor heating. The shift of the morning peak at Miskolc by about one hour from summer to winter can be explained by the shift to daylight savings time in Hungary.

The observations show that in every season, the highest PM₁₀ concentrations can be expected in the morning and evening hours, while the minimum concentrations were measured between 11 and 15 hours. In winter, during this time period, the contribution of residential heating was not remarkable because people spent their time in public institutions fuelled by natural gas (Muntean *et al.*, 2017).

Fig. 2. Diurnal variation in the averaged PM₁₀ concentrations in Budapest (Gilice tér), Miskolc (Búza tér), and Pécs (Boszorkány utca) (averaged over the 11-year period from 2007–2017).

3.2. Spectral Decomposition of Time Series

Local meteorological conditions exert a strong influence over day-to-day variations in PM₁₀ concentrations; therefore, the meteorological signal must be removed for air quality planners and managers to examine the underlying emissions-related trends and make better air quality management decisions for the future. In this part of the study, a Kolmogorov-Zurbenko (KZ) filter was used to spectrally decompose the PM₁₀ time series between 2007 and 2017 into different forcings controlled by different atmospheric processes that influence the measured PM₁₀ concentrations. The Kolmogorov-Zurbenko (KZ) filter can be used to separate the time series of short-term components (influenced by the prevailing meteorological conditions) and baseline components (influenced by the emissions and boundary conditions) (Kang *et al.*, 2013).

The KZ filter is based on Rao and Zurbenko's statement and was described in detail by Wise and Comrie (2005). It is well documented that there is an obvious link between meteorological conditions and the air pollution level. Meteorological variability could overwhelm the effects of changes in pollutant emissions. To properly assess the changes in air concentration due to emission changes or air quality measures, the influence of short-term meteorological variability should be eliminated. Application of the KZ filter was first tested for tropospheric ozone issues, and it was later extended to regional PM and NO₂ problems (Sá *et al.*, 2015). A time series of air quality data can be represented by

$$A(t) = e(t) + S(t) + W(t) \quad (1)$$

where $A(t)$ is the original time series, $e(t)$ is the long-term trend component, $S(t)$ is the seasonal variation, and $W(t)$ is the short-term component. Baseline components are defined as the sum of the long-term and seasonal components.

$$e(t) = KZ_{(365,3)} \quad (2)$$

$$S(t) = KZ_{(15,5)} - KZ_{(365,3)} \quad (3)$$

$$W(t) = A(t) - KZ_{(15,5)}. \quad (4)$$

The KZ filter is a low-pass filter derived from repeated iterations of a moving average. In our study, the daily mean PM₁₀ concentration values were used. $KZ_{(15,5)}$ denotes the filter with a 15-day length and five iterations, and $KZ_{(365,3)}$ denotes the filter with a 365-day length and three iterations. The moving average for a $KZ_{(m,p)}$ filter (a filter with window length m and p iterations according to Sá *et al.* (2015)) is defined by

$$Y_i = \frac{1}{m} \sum_{j=-k}^k A_{i+j} \quad (5)$$

where k is the number of values included on each side of the targeted value, the window length is $m = 2k + 1$, and A is the input time series. The separation of the original air quality time series into the three components (long-term, seasonal, and short-term) for the daily average PM₁₀ is presented in Fig. 3. The short-term component is attributable to weather and short-term fluctuations in precursor emissions (random effect), the seasonal component is a result of changes, for example, in the boundary layer height (effects that have typical seasonal variability), and the long-term trend results from changes in overall emissions, atmospheric transport, and climate.

Fig. 3. Decomposition of a daily average PM₁₀ time series by the Kolmogorov-Zurbenko (KZ) filter into long-term, seasonal, and short-term components.

Table 1. Contribution of each component to the total variance in the original PM₁₀ concentration.

	Budapest	Miskolc	Pécs
Long-term component	1%	1%	6%
Seasonal component	32%	45%	33%
Short-term component	67%	54%	61%

The effect of the local weather conditions is reflected in the short-term component. These effects are random and can change from place to place, but they may have a significant impact on the daily variability in PM₁₀ concentrations. The effect of this component is the most important among the three components during the winter months for all three Hungarian cities. The short-term variations in Budapest and Pécs exhibit relative maxima in January 2011 and a secondary maximum in January 2017 (Fig. 3), which can be explained by a strong atmospheric inversion over the whole Carpathian Basin. In January 2017 an extreme maximum of the short-term component can be observed in Miskolc. This study also confirms that poor air quality situations in Hungary are generally connected to special meteorological conditions.

In the case of the seasonal component, we found a strong relationship with the boundary layer height and the temperature since these two parameters show typical seasonality. In the case of a low boundary layer height, the mixing properties in the lower atmosphere are very poor and cause the accumulation of PM. When the temperature is below 0°C, the emission from residential heating increases.

The long-term trend results are related to changes in emissions. There are large differences between the long-term components of the three examined cities. No significant decrease in the long-term component concentration values occurs in Budapest. In the case of Miskolc and Pécs, the decreasing trend can be explained by the implementation of several measures regarding PM₁₀ levels. In Miskolc, the cement industry was phased out in 2011 with the aim of reducing the level of pollutant release. This study also demonstrates the need for more local measures to improve the air quality in these cities.

To understand the contribution of each temporal component to the original daily average PM₁₀ concentration value, the variance in each generated time series was determined, and their contributions to the total variance in the original data were calculated (Table 1).

3.3. Relationship between PM₁₀ and Meteorological Parameters

The importance of the short-term component, containing the day-to-day variability due to weather, demonstrated that the meteorological parameters affect the PM₁₀ concentrations in Hungary. To investigate the relationship between PM₁₀ concentrations and meteorological variables, first, the correlation coefficients between daily meteorological variables such as temperature, wind speed, relative humidity, atmospheric pressure, global radiation, and boundary layer height, and daily PM₁₀ were determined (Table 2).

Although the relationship between meteorological parameters and PM₁₀ is rather complex, we found some strong relationships between them. Positive (RH, p, and GR) and negative (T, WS, and PBL) correlations were found between several meteorological parameters and PM₁₀ values for the whole year. The seasonality of the correlation between PM₁₀ and the meteorological parameters was analysed separately, and large differences were found between the seasons. See Table 2 for the correlation values.

The highest negative values of the correlation coefficient can be observed with temperature, wind speed, and boundary layer height in the winter period. Significant seasonality can be observed in the case of temperature and relative humidity. In summer, the correlation with temperature resulted in a positive value, while in other seasons, a negative value of the correlation coefficient was observed. This suggests that in summer, heat might lead to poor air quality due to increasing concentrations of secondary PM₁₀. The correlation with temperature changed from positive to negative (from summer to autumn, respectively). The explanation of the special behaviour of temperature can be connected with the seasonal variability of the usage of solid fuel (wood and coal). In summer, there is no residential combustion, while the intensity of heating is the greatest in winter when the usage of solid fuel is determinative in Hungary.

Table 2. Correlation coefficients between PM₁₀ and several meteorological parameters.

	Spring	Summer	Fall	Winter	Year
Budapest					
temperature (°C)	−0.01	0.68	−0.18	−0.49	−0.38
wind speed (m s ^{−1})	−0.33	−0.31	−0.42	−0.58	−0.44
relative humidity (%)	−0.19	−0.32	0.15	0.26	0.25
pressure (hPa)	0.21	0.01	0.34	0.30	0.32
global radiation (J cm ^{−2})	−0.02	0.26	−0.09	0.01	0.28
boundary layer height (m)	−0.28	0.02	−0.55	−0.48	−0.50
Pécs					
temperature (°C)	0.00	0.69	−0.23	−0.50	−0.37
wind speed (m s ^{−1})	−0.29	−0.22	−0.37	−0.47	−0.33
relative humidity (%)	−0.12	−0.29	0.20	0.20	0.24
pressure (hPa)	0.19	−0.09	0.23	0.26	0.24
global radiation (J cm ^{−2})	0.00	0.29	−0.15	0.05	−0.26
boundary layer height (m)	−0.32	−0.03	−0.49	−0.41	−0.47
Miskolc					
temperature (°C)	−0.13	0.56	−0.23	−0.42	−0.48
wind speed (m s ^{−1})	−0.12	0.02	−0.37	−0.37	−0.26
relative humidity (%)	−0.14	−0.19	0.20	0.19	0.30
pressure (hPa)	0.16	0.08	0.23	0.18	0.24
global radiation (J cm ^{−2})	−0.02	−0.09	−0.15	0.12	−0.07
boundary layer height (m)	−0.33	−0.04	−0.49	−0.44	−0.56

In the case of relative humidity, the situation was opposite in summer and spring, with a negative correlation coefficient, while in the other times of the year, a positive correlation coefficient was found. The moderate negative correlation suggested that in spring and summer, the high relative humidity was likely the result of precipitation; therefore, the PM₁₀ concentration was reduced by wet deposition. On the one hand, the strong wind flushes the fine fraction of the PM out of the system, and on the other hand, it can increase the coarse fraction concentration, as a result of the more intense suspension of particles from ground surfaces. (Chaloulakou *et al.*, 2003; Kukkonen *et al.*, 2005; Zhang *et al.*, 2018). A negative correlation between PM₁₀ and boundary layer height likely resulted in significantly low dispersion and dilution of air pollutants. This fact demonstrates that meteorological conditions play an important role in the development of air pollution episodes.

Vardoulakis and Kassomenos (2008) obtained similar relationships and found correlation coefficients in the case of temperature (−0.24) and wind speed (−0.55) in the Birmingham Centre during cold conditions.

Multiple linear regression analysis has been used in the field of air pollution by several authors (Gupta and Christopher, 2009; Vlachogianni *et al.*, 2011; Lin *et al.*, 2012; Garcia *et al.*, 2016). In our work, a regression method was applied for only winter because the strongest correlation between meteorological variables (temperature, wind speed, and boundary layer height) and PM₁₀ was observed in this season, and the highest PM₁₀ concentration level occurred in winter, which we wanted to predict.

Based on the results of the determination of the correlation coefficients, many meteorological parameters were selected for the regression analysis. Table 3 shows the obtained R² values for the three most important meteorological parameters. In the case of temperature, the relationship was linear, while the boundary layer height and wind speed resulted in much better R² values when a logarithmic relation was supposed. These results indicated that nonlinear multiple regression should be applied to predict the PM₁₀ concentration.

The primary aim of our regression analysis was to determine the meteorological situations in which the PM₁₀ concentration was expected to exceed the 50 µg m^{−3} concentration threshold. The studies presented so far helped us to determine the most important meteorological factors that had an essential effect on the PM₁₀ concentration in the winter period. In the development of the method, we assumed that the emissions were constant each day in winter and that the

Table 3. The R^2 values for several meteorological parameters in winter.

	Budapest	Pécs	Miskolc
temperature (°C) – linear	0.24	0.25	0.17
boundary layer height (m) – linear	0.24	0.16	0.19
boundary layer height (m) – logarithmic	0.38	0.24	0.30
wind speed (m s ⁻¹) – linear	0.34	0.22	0.13
wind speed (m s ⁻¹) – logarithmic	0.49	0.29	0.22

actual PM₁₀ concentration was controlled by the weather. Unfortunately, we did not have exact information about how the emission of PM₁₀ from domestic heating changed from day to day. We only assumed that this effect could be driven by the air temperature, but we could not determine its measure. This was the reason why we did not take into account the effect of emission changes in this method.

As a result of the analysis of the correlation coefficients, it was possible to predict the daily average PM₁₀ concentration. In our case, the dependent variable that we wanted to predict was the PM₁₀ concentration, and the independent variables were the temperature (T), wind speed (WS), and boundary layer height (PBL). The data analysis suggested that the best result could be obtained by fitting the nonlinear model to the meteorological variables. Several multiplicative regression models were tested to find the best-fitting model. Based on the evaluations of different models, the proposed regression formula was the following:

$$\ln[\text{PM}_{10}] \approx \alpha_0 + \alpha_1 \ln[\text{PBL}] + \alpha_2 \ln[\text{WS}] + \alpha_3 [T] \quad (6)$$

The summarized results of the regression (Table 4) confirmed that the proposed model could adequately predict the PM₁₀ values. The overall test statistics indicated the significance of our model, i.e., high F statistics for 3 variables, and given degrees of freedom along with the zero p-values for the null hypothesis test indicated that all the model coefficients were equal to zero.

From the regression formula (Eq. (6)), the equation that predicts the daily average PM₁₀ concentration is:

$$\text{PM}_{10} \approx \exp(\alpha_0 + \alpha_1 \ln[\text{PBL}] + \alpha_2 \ln[\text{WS}] + \alpha_3 [T]) \quad (7)$$

where α_0 α_1 α_2 α_3 are the multiple regression coefficients, T is the temperature (°C), PBL is the boundary layer height (m), and WS is the wind speed (m s⁻¹).

In Table 5, we summarized the corresponding parameters of the prediction in Eq. (7) for the three sites studied in this paper. To evaluate the equation performance, we tested it on an independent database (December 1, 2018–February 28, 2019). During the validation of the regression equation, the predicted PM₁₀ values were compared with the corresponding PM₁₀

Table 4. Summary of the evaluation of the regression.

	Budapest	Pécs	Miskolc
residual standard error of estimates	0.38	0.42	0.41
R^2	0.58	0.49	0.43
adjusted R^2	0.57	0.49	0.43
F statistic	440.6 (3, df = 974)	260.8 (3, df = 799)	246.9 (3, df = 977)
p-value	< 2.2e ⁻¹⁶	< 2.2e ⁻¹⁶	< 2e ⁻¹⁶

Table 5. Multiple regression coefficients and R^2 values for predicting the PM₁₀ concentration.

	α_0	α_1	α_2	α_3	R^2
Budapest	5.19	−0.22	−0.53	−0.04	0.57
Pécs	5.27	−0.28	−0.32	−0.05	0.49
Miskolc	5.98	−0.38	−0.05	−0.04	0.43

Fig. 4. Observed and predicted daily average PM₁₀ concentrations at the Budapest, Pécs, and Miskolc stations during the winter period.

measurements. Time series of the observed and predicted PM₁₀ concentrations for the Budapest, Miskolc, and Pécs stations are presented in Fig. 4. The temporal variations in the curves are similar for most of the predicted and observed values. We recognize that extremely high observed daily mean concentrations were underpredicted in most of the cases with this method.

Several statistical scores, such as bias (BIAS), mean absolute error (MAE), root mean square error (RMSE) and R^2 , were determined to evaluate the regression equation. The second group of scores focuses on the prediction of PM₁₀ exceedances at the threshold ($50 \mu\text{g m}^{-3}$) and includes indices based on a contingency table. These indices are the probability of detection (POD) and false alarm ratio (FAR). Table 6 shows the results of the evaluation statistics.

The classical statistical scores (BIAS, MAE, RMSE) indicate that the regression equation provided the best results in Pécs. In Budapest, the determination coefficient (R^2) was the highest ($R^2 = 0.48$), while in Pécs and Miskolc, it was low but acceptable (0.34 and 0.25, respectively). The POD and FAR values were determined for the threshold value of $50 \mu\text{g m}^{-3}$. These indicators were the best in Miskolc. The result of the validation showed a mixed picture, and it would be very difficult

Table 6. Evaluation statistics for forecasting.

Evaluation Statistics	Budapest	Pécs	Miskolc
RMSE	19.69	14.77	26.44
BIAS	-7.79	0.62	-12.26
MAE	13.56	11.44	19.58
R^2	0.48	0.34	0.25
POD = $A/(A + B)^*$	0.32	0.31	0.63
FAR = $C/(C + A)^*$	0.11	0.33	0.10

* A, B and C are the number of forecasted and observed exceedances of the limit value, number of observed exceedances but not forecasted, and number of forecasted exceedances but not observed, respectively.

to determine the station with the best results from the regression equation. Our method exhibited the best result for Miskolc when the goal was to predict whether the PM₁₀ concentration would exceed the 50 µg m⁻³ concentration threshold limit.

Finally, it must be mentioned that in the case of Budapest, the locations of the meteorological instruments and air quality monitors are the same. Unfortunately, the situations in Pécs and Miskolc are different, and the meteorological station is 500–1000 m from the air quality monitoring station. This fact could influence the results of the multiple regression analysis.

4 CONCLUSION

In this paper, the effects of meteorological parameters on PM₁₀ concentrations and their temporal behaviours were examined using monitoring data from different cities in Hungary. Several statistical indicators (average, temporal variation, correlation, regression) were determined to assess the characteristics of the PM₁₀ pollution levels in these cities.

The yearly, monthly, weekly, and daily variations in the PM₁₀ time series were determined using the data from three large cities in Hungary where the air quality is problematic. In the case of yearly averages, no limit value exceedances (40 µg m⁻³) were detected after 2011. However, analysing the daily average, the most limit value exceedances could be expected in winter and fall. There were large differences between the cities, and the air quality varied between moderate and poor in both Pécs and Miskolc. There were large variances year after year, which could be controlled by the meteorological conditions. Essential differences were not found between the characteristics of the weekly variations. In summer, the daily variation in PM₁₀ was very similar in Budapest and Pécs, but in Miskolc, we found a characteristic peak in the morning that could be attributed to local traffic. In winter, the daily variation in PM₁₀ from every station had the same features. We assumed that in the case of Miskolc, the effect of residential combustion was higher than the effect of traffic, so the peak originating from morning traffic could not be identified. We concluded that no significant variability in the average PM₁₀ concentrations was found throughout a week. In each city, there was a slight increase from Monday to Friday, and then a pronounced decline was observed during the weekend. A difference between the mean weekly concentration values in the individual cities was clearly observable.

Variations in the concentration levels of PM₁₀ could be decomposed to a baseline component of pollution created by emission sources and to other components controlled by meteorological conditions. The Kolmogorov-Zurbenko (KZ) filter is a useful tool for the investigation of the interactions between emissions, meteorological conditions, and air quality levels. The PM₁₀ long-term component has the lowest contribution (only 1–6%) to the variance observed in the original data. On the other hand, the short-term component has the highest contribution to the total variance in the original data. In Budapest, the contribution of the short-term component to the total variance in the original data is the highest and is more than 10% higher than that in Miskolc. The contribution of the seasonal component is the highest in Miskolc, approximately 10% higher than that in Budapest.

We also analysed the evolution of urban PM₁₀ concentrations in terms of the relevant meteorological parameters. Correlation analysis was carried out to identify the most important meteorological variables influencing the PM₁₀ concentration in Hungary. As a result of this analysis, the highest PM₁₀ concentrations were clearly related to the decreasing air temperature, lowering PBL height, and weakening wind speed during winter. The effect of temperature on the PM₁₀ concentration was likely to be very complex. Low temperature and poor vertical mixing could be observed together in winter; at the same time, low temperature induced an increase in the usage of solid fuel, which resulted in increased PM₁₀ emissions from domestic heating.

Multiple regression analysis was used to determine the connection between the concentration of PM₁₀ and specific meteorological parameters: temperature, planetary boundary layer height, and wind speed. The results of this study showed that the relationship between PM₁₀ and each meteorological parameter was acceptable (the R² values in the regression formula were 0.57, 0.49, and 0.43), and the model that was built could be used for PM₁₀ prediction. Our method did not take into account the daily changes in PM₁₀ emissions. We assumed that the emissions were constant on each day of the winter season. After model validation, it could be stated that our

constructed statistical model could be effectively used to decide whether the daily mean PM₁₀ values were expected to exceed the 50 µg m⁻³ threshold value.

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