Mann–Kendall trends and air quality assessment of PM_{10} and $PM_{2.5}$ in three urban areas of northeast Romania, 2017-2024

Tendances Mann-Kendall et évaluation de la qualité de l'air pour les PM_{10} et $PM_{2.5}$ dans trois zones urbaines du nord-est de la Roumanie, 2017-2024

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ABSTRACT: From 2017 to 2024, PM_{10} and $PM_{2.5}$ concentrations were analyzed at four monitoring stations (Suceava, Iași, and Botoșani) using the Mann–Kendall test and Sen's slope estimator to identify monotonic trends. Significant decreases in PM_{10} were observed at BT-1 (−1.8 µg/m³/year) and IS-2 (−1.1 µg/m³/year), while SV-1 and SV-2 showed non-significant declines. For $PM_{2.5}$, IS-2 exhibited a significant decreasing trend from 2017 to 2024 (−2.9 µg/m³/year), which was not significant over 2009–2024. Extended analysis of $PM_{2.5}$ from 2009 to 2024 revealed clearer reductions in March–April at SV-1, whereas summer and autumn months showed no significant trends. At BT-1, $PM_{2.5}$ concentrations were generally stable, except for a slight increase in June. Hourly PM_{10} concentrations displayed distinct diurnal and seasonal patterns, with peaks in the morning and evening, lower values around midday, higher levels in late autumn and winter, and generally lower concentrations on weekends. In 2024, PM_{10} and $PM_{2.5}$ levels remained below the limits set for human health protection, but meeting the stricter standards of Directive (EU) 2024/2881 will require further efforts.

KEY WORDS: PM_{10}, $PM_{2.5}$, Mann–Kendall test and Sen's slope estimator, Diurnal variation, Urban air quality.

RÉSUMÉ : De 2017 à 2024, les concentrations de PM_{10} et $PM_{2.5}$ ont été analysées dans quatre stations (Suceava, Iași et Botoșani) avec le test de Mann-Kendall et la pente de Sen. Des baisses significatives de PM_{10} ont été observées à BT-1 (−1,8 µg/m³/an) et IS-2 (−1,1 µg/m³/an), tandis que SV-1 et SV-2 ont montré des diminutions non significatives. Pour les $PM_{2.5}$, IS-2 a présenté une baisse significative entre 2017 et 2024 (−2,9 µg/m³/an), non significative sur 2009–2024. L'analyse 2009–2024 des $PM_{2.5}$ a révélé des réductions en mars-avril à SV-1, les mois d'été et d'automne restant stables. À BT-1, les $PM_{2.5}$ étaient généralement stables, sauf une légère hausse en juin. Les PM_{10} horaires montraient des variations diurnes et saisonnières, avec des pics matin et soir, des valeurs plus basses vers midi, des niveaux plus élevés fin d'automne et hiver, et des concentrations généralement plus faibles le week-end. En 2024, les niveaux de PM_{10} et de $PM_{2,5}$ sont restés en dessous des limites fixées pour la protection de la santé humaine, mais le respect des normes plus strictes prévues par la Directive (UE) 2024/2881 nécessitera des efforts supplémentaires.

MOTS CLÉS : PM_{10}, $PM_{2.5}$, Test de Mann–Kendall et estimateur de pente de Sen, variation diurne, qualité de l'air urbain.

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1. Introduction

Air pollution is a major environmental and public health concern worldwide, particularly in urban areas. , Particulate matter with an aerodynamic diameters of 10 µm (PM_{10}) and 2.5 µm ($PM_{2.5}$) is especially harmful, because these particles can penetrate deep into the respiratory system, causingadverse health effects (Mebrahtu et al., 2023; Mohebbichamkhorami et al., 2020; Pini et al., 2021).

In Europe, a large proportion of the population is still exposed to urban $PM_{2.5}$ levels above the WHO recommended level, despite a gradual decline in emissions over recent decades (EEA, 2025). According to recent reports from the European Environment Agency, over 90% of the urban population in the EU is exposed to $PM_{2.5}$ concentrations that exceed WHO guidelines. High concentrations of PM_{10} and $PM_{2.5}$ persist, especially in Central and Eastern Europe, where solid fuels are still commonly used for heating (European Environment Agency, 2025).

Romania is among the member states where particle concentrations can exceed daily limit values for PM_{10} and WHO guideline values for $PM_{2.5}$, especially in urban areas and during the cold season. This is due to residential heating with solid fuels and traffic (Z. Bodor, 2020; European Environment Agency, 2025).

Recent studies based on data from the national air quality monitoring network indicate the need for trend analyses and integrated air quality assessments. These studies show episodes of significant PM_{10} pollution in several Romanian cities, including those in the eastern part of the country (Z. Bodor, 2020).

The northeastern region of Romania, which includes the cities of Suceava, Iași, and Botoșani, is affected by several sources of particulate matter, including urban traffic, residential heating with solid fuels, industrial activities, and the long-range transport of pollutants from other regions. Previous studies by Nistor et al., Lazurcă and Mihăilă, Drăgoi et al., and Sfîcă et al. have shown that PM_{10} levels peak at sites influenced by traffic and industry in Iași and Suceava and are strongly enhanced in winter under stable atmospheric conditions. This finding emphasizes the combined role of local emissions, meteorology, and urban morphology in causing exceedances (Dragoi et al., 2023; Lazurca, 2015; Nistor et al., 2020; Nistor et al., 2023; Sfîcă et al., 2018).

Temperature plays an important role in the dispersion or accumulation of pollutants. It influences chemical reactions in the atmosphere and emission levels (Basemera et al., 2025). For example, low temperature during the cold season increase fossil fuel consumption for residential heating, leading to higher particulate emissions.

As a result,the evolution of PM concentrations in urban areas in northeastern Romania is strongly influenced by traffic intensity and the regional continental climate, characterized by hot summers and cold winters with frequent episodes of atmospheric stability during the cold season. These conditions favour pollutant accumulation and the occurrence of pollution episodes, particularly in winter(Sfîcă et al., 2018).

Analyzing long-term trends in PM_{10} and $PM_{2.5}$ makes it possible to identify significant changes in pollution levels and to evaluate the effectiveness of emission reduction policies. Non-parametric statistical methods, such as the Mann–Kendall (MK) test and the Theil–Sen (TSE) slope estimator, are widely used to analyze highly variable, non-normally distributed atmospheric time series data and therefore represent appropriate tools for investigating particulate matter trends in this region (Chaudhuri & Dutta, 2014; Gocic & Trajkovic, 2013; Jaiswal et al., 2018; Mahesh & Sivakumar, 2025; Mohammad et al., 2022; Sen et al., 2019; Umoh et al., 2022).

In addition to multi-year trend analysis, assessing daily and seasonal variability of particulate matter concentrations is important for identifying patterns specific to urban environments. In cities such as Suceava, Iași and Botoșani, hourly variations in PM_{10} variations are closely linked to traffic intensity, daily population activity and atmospheric dispersion conditions. Seasonal variability reflects the the additional impact of residential heating in winter and the enhanced resuspension of particulate matter during the warmer months. Analyzing the hourly distribution of PM_{10} over several years helps to identify critical exposure intervals and to better constrain the dominant sources of urban pollution.Long-term Mann–Kendall trend analysis for 2017–2024 shows significant decreases in PM_{10} and $PM_{2.5}$ at the urban background station in Iași and in PM_{10} at the urban background station in Botoșani, while the two Suceava stations generally exhibit non-significant or only weak seasonal trends, indicating that recent improvements are spatially heterogeneous and more pronounced for $PM_{2.5}$ than for PM_{10}.

In Romania, ambient air quality is regulated by Law No. 104/2011, as amended, which transposes Directive 2008/50/EC on ambient air quality and cleaner air for Europe and Directive 2004/107/EC concerning arsenic, cadmium, mercury, nickel, and polycyclic aromatic hydrocarbons in ambient air.

For PM_{10}, Law No. 104/2011 establishes, for the protection of human health, a daily limit value of 50 μg/m³, not to be exceeded more than 35 times per calendar year, and an annual limit value of 40 μg/m³. For $PM_{2.5}$, the law establishes only an annual limit value of 25 μg/m³.

In 2024, Directive (EU) 2024/2881 on ambient air quality and cleaner air for Europe was adopted, with transposition into Romanian national legislation required by December 2026. This directive establishes more stringent air quality standards.

Considering these aspects, this article aims to analyze the evolution of urban air quality in the municipalities of Suceava, Iași, and Botoșani between 2017 and 2024 by: (i) evaluating the annual and monthly statistical trends of PM_{10}, $PM_{2.5}$ concentrations and air temperature using the Mann–Kendall test and the Theil–Sen estimator; (ii) investigating diurnal and seasonal variations in hourly PM_{10} concentrations; and (iii) assessment of the compliance of PM_{10} and $PM_{2.5}$ concentrations with the limit values established by national and European legislation for the protection of human health.

The results provide relevant information for assessing air quality and supporting strategies to reduce atmospheric pollution.

2. Methods

2.1. Data source

This study uses PM_{10} and $PM_{2.5}$ concentration data for the period 2017 to 2024. The data were obtained from three urban background stations and one industrial station located in the urban area, all belonging to the National Air Quality Monitoring Network (RNMCA)(Figure 1 and Table 1).

Trends in PM_{10} and $PM_{2.5}$ concentrations, as well as the number of exceedances of the daily limit value for PM_{10}, were analyzed based on daily concentrations measured using the gravimetric reference method, in accordance with Law No. 104/2011 on ambient air quality. Additionally, the evolution of annual mean PM_{10} and $PM_{2.5}$ concentrations recorded at the monitoring stations within the study area was evaluated against the corresponding annual limit value.

In this study, we only considered those years for which the national air-quality website validated the annual mean concentrations, i.e. years that fulfil the quality and aggregation requirements of Annex 4 of Law no. 104/2011 and Annex I of Directive 2008/50/EC (including the minimum 90% annual data capture for daily gravimetric PM_{10} and $PM_{2.5}$ measurements, excluding maintenance periods).

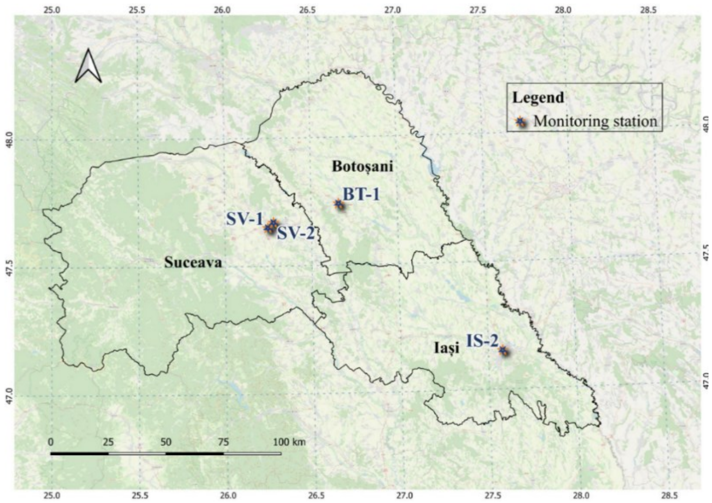

Figure 1 Geographical locations of the four study monitoring stations.

Table 1 Location of air quality monitoring stations used in this study.

Site	Latitude	Longitude	Altitude
SV-1	47.649259°N	26.249009°E	376 m
SV-2	47.668825°N	26.281403°E	289 m
BT-1	47.739945° N	26.658999°E	167 m
IS-2	47.150951°N	27.581920°E	42 m

The diurnal variability of PM_{10} concentrations was assessed using hourly data obtained by the nephelometric method from the same RNMCA stations. Currently, there is no defined reference gravimetric method for hourly PM_{10} measurements. Therefore, these data are considered indicative and are used solely to characterize typical diurnal cycles, not to assess compliance with legal limit values. Consequently, we did not apply the annual data-capture criteria used for daily gravimetric data, since the purpose of the nephelometric hourly series is to describe the shape of the diurnal cycle rather than to provide reference-quality concentration value.The data are publicly available on the national air quality platform https://www.calitateaer.ro (accessed on 12 December 2025. Python was used to process the data.

2.2. Trend analysis methods

To detect significant trends in particulate matter PM_{10} and $PM_{2.5}$ concentrations, both parametric and non-parametric tests are used. In this study, two non-parametric methods were applied: the Mann-Kendall test and Sen's slope estimator. To ensure statistical rigor, only years that met the minimum data capture criteria were retained. One advantage of these non-parametric tests is that they can process data series with gaps (missing years) without compromising the validity of the results (Gilbert, 1987).

2.2.1. Mann–Kendall (MK) Trend Test

The Mann-Kendall (MK) test is a non-parametric method used to identify monotonic trends (increasing or decreasing) in time series data, by comparing the relative magnitudes of observations without relying on absolute values (Gilbert, 1987).

The MK test starts from the null hypothesis H_0, which states that there is no monotonic trend (the observations x_i are independent and randomly ordered in time), in contrast to the alternative hypothesis H_1, which assumes the presence of an increasing or decreasing monotonic trend. The data, treated as an ordered time series, are compared in pairs: if a later value is greater than an earlier one, the statistic S is increased by 1, if it is smaller, S is decreased by 1. The net result of all these comparisons gives the final value of S, which is negative for a decreasing trend (S<0), zero for no trend (S=0), and positive for an increasing trend (S>0) (Mann, 1945; Kendall, 1975; Gilbert, 1987).

The MK test principal statistic S is calculated using the following equations.

$$S = \sum_{i=1}^{n-1} \sum_{j=i+1}^{n} sgn(x_j - x_i) \quad (1)$$

$$sgn(x_j - x_i) = \begin{cases} +1 \ if \ x_j - x_i > 0 \\ 0 \ if \ x_j - x_i = 0 \\ -1 \ if \ x_j - x_i < 0 \end{cases} \quad (2)$$

where x_j and x_i are the annual values in different years j and i, j>i, respectively.

The variance, Var(S), measures the expected dispersion of the S statistic under the null hypothesis. It quantifies the variability that could arise from simple random fluctuations (coincidence) in the absence of a real trend.

The variance is computed as:

$$Var(S) = \frac{n(n-1)(2n+5) - \sum_{i=1}^{m} t_i(t_i-1)(2t_i+5)}{18} \quad (3)$$

where n is the number of years with available data, m is the number of tied groups and t_i denotes the number of ties of extent i. A tied group is a set of sample data having the same value. Since the concentration values for PM_{10} and $PM_{2.5}$ in the analyzed period were distinct for each year, the number of tied groups (m) was zero, and the variance formula was applied in its simplified form.

Although exact distribution tables for S are often used for small sample sizes (n<10), the standard normal test statistic Z_S was computed in this study using a continuity correction (as shown in Equation 4). This approach is widely implemented in statistical software to provide a standardized measure of trend significance across all monitoring stations (Gilbert, 1987).

$$Z_S = \begin{cases} \frac{S-1}{\sqrt{Var(S)}}, \ if \ S > 0 \\ 0 \ , \quad if \quad S = 0 \\ \frac{S+1}{\sqrt{Var(S)}}, \ if \ S < 0 \end{cases} \quad (4)$$

Positive Z_S values indicate an increasing trend, while negative Z_S values indicate a decreasing trend. In this study, the null hypothesis H_0 is rejected and a significant trend exists if $|Z_S|>Z_{1-\alpha/2}$, where $Z_{1-\alpha/2}$ is the critical value obtained from the standard normal distribution table. Two significance levels were considered for the analysis: α=0.05 (corresponding to a 95% confidence level) and α=0.01 (99% confidence level). At the 5% significance level, the null hypothesis of no trend is rejected if $|Z_S|>1.96$, while at the 1% significance level, it is rejected if $|Z_S|>2.576$ (Gilbert, 1987).

Probability (p-value) was calculated based on the standard normal distribution. For a two-tailed test, it represents the probability of obtaining a Z statistic as extreme as the one observed, assuming the null hypothesis (H_0) is true. If the p-value is less than the significance level α=0.05, H_0 is rejected. Rejecting H_0 indicates that a significant trend exists in the time series, while accepting H_0 indicates that no trend was detected. Upon rejecting the null hypothesis, the result is considered statistically significant (Gilbert, 1987; Karmeshu, 2012).

2.2.2. Theil–Sen Slope Estimator (TSE)

To quantify the magnitude of monotonic trends in annual mean concentrations, Sen's slope estimator (Sen, 1968; Wilcox, 2001) was applied in combination with the Mann–Kendall test. Sen's slope was computed as the median of all pairwise slopes between data points in each time series, defined as the change in concentration divided by the time difference between two years. Negative Sen's slope values indicate a decreasing trend in pollutant levels over time, while positive values indicate an increasing trend. For each station and pollutant, Sen's slope was reported together with the Mann–Kendall p-value, and trends were considered statistically significant only when $p < 0.05$. (Bosch et al., 2025; Kushwaha et al., 2025; Shiferaw et al., 2023; Yadav, et al., 2025).

The trend analysis for PM_{10} and $PM_{2.5}$ and temperature was performed using the Mann–Kendall test and Sen's slope applied to annual mean PM concentrations and annual mean temperatures, as well as to monthly mean concentrations to investigate seasonal patterns in the monotonic trends. The statistical significance of the trends was evaluated at a confidence level of 95% (α=0.05), where the null hypothesis (H_0) of no trend was rejected if the calculated p-value was less than 0.05 (Gilbert, 1987).

3. Results and discussion

3.1. Statistical Trend Analysis of PM and Temperature Data

This chapter presents the assessment of temporal variations in PM and temperature, employing non-parametric statistical methods to identify the presence, direction, and magnitude of long-term trends. Temperature was included in the trend analysis to provide a broader environmental context, as it plays a critical role in the variability of particulate matter. Variations in temperature directly lead to changes in residential heating demands and atmospheric dispersion conditions, such as thermal inversions. These changes can result in significant fluctuations in PM_{10} and $PM_{2.5}$ concentrations.

3.1.1. Annual Trend Analysis of PM_{10} and $PM_{2.5}$ concentrations

For PM_{10} over 2017–2024, statistically significant decreasing trends were detected at BT-1 and IS-2, with Sen's slope estimates of about −1.8 and −1.1 µg/m³ per year, respectively. At SV-1 and SV-2, negative Sen's slopes indicated slight decreases. However, the Mann–Kendall test revealed no significant monotonic trend ($p > 0.05$), classifying these series as having no significant trend.

For $PM_{2.5}$, for period 2017-2024, a statistically significant decreasing trend was observed only at IS-2, with Sen's slope indicating a reduction of about 2.9 µg/m³ per year over the study period, while BT-1 and SV-1 indicated no significant monotonic trend according to the Mann–Kendall test ($p > 0.05$). Although a negative slope of concentrations was estimated at BT-1 and SV-1, this trend is not statistically significant ($p > 0.05$). Therefore, these series were classified as having no significant trend.

At $PM_{2.5}$, the analysis period was extended to the 2009–2024 interval due to the need for a more robust dataset. Shorter periods can be sensitive to extreme annual variations, which can mask real underlying trends.

While no significant trend was observed at the SV-1 station for the 2017–2024 period (p=0.133), the long-term analysis (2009–2024) indicated a statistically significant decreasing trend (p=0.012). These results demonstrate that improving air quality at this location is a long-term process that remains undetectable within a shorter six-to-eight-year timeframe due to interannual variability. Therefore, extending the analysis period was necessary to capture the underlying monotonic trend that would have otherwise been obscured by short-term fluctuations.

At the IS-2 station, although a statistically significant decreasing trend was observed during the period 2017–2024 (p = 0.024, Z = −2.875), this significance was not maintained over the extended period 2009–2024 (p = 0.443). The results suggest that the reduction in pollution at the IS-2 station does not represent a long-term linear process, but rather a recent phenomenon, likely driven by the implementation of measures outlined in the air quality plans (Drăgoi et al., 2025).

At the BT-1 station, an analysis of the period from 2009 to 2024 indicated a continued absence of a significant trend (p = 0.710), which is consistent with the short-term findings. The persistence of high p-values indicates that $PM_{2.5}$ concentrations at this location have remained relatively stable over the 15-year period without a statistically significant change in direction, despite isolated annual fluctuations (Table 2).

Table 2 Results of the Mann–Kendall test and Sen's slope estimator for annual trends of PM_{10} and $PM_{2.5}$ concentrations.

Station	Period	n	S	Z	p-value	Significant trend (α = 0.05)	Sen's slope (μg/m³/year)
				PM_{10}			
SV-1	2017-2024	8	-12	-1.360	0.174	No significant trend	−0.53
SV-2	2017-2021	6	-2	-0.245	0.807	No significant trend	−0.15
BT-1	2017-2024	8	-18	-2.100	0.035	Yes (Decreasing trend)	−1.75
IS-2	2017-2024	8	-22	-2.590	0.009	Yes (Decreasing trend)	−1.08
				$PM_{2.5}$			
SV-1	2017-2024	6	-9	-1.503	0.133	No significant trend	-0.89
BT-1	2017-2024	3	-1	0	1.0	No significant trend	-0.55
IS-2	2017-2024	6	-13	-2.254	0.024	Yes (Decreasing trend)	-2.88
SV-1	2009-2024	10	-29	-2.504	0.012	Yes (Decreasing trend)	-0.84
BT-1	2009-2024	8	-4	-0.371	0.711	No significant trend	-0.21
IS-2	2009-2024	14	-15	-0.766	0.443	No significant trend	-0.27

3.1.2. Monthly Trend Analysis of PM_{10} and $PM_{2.5}$

During the period from 2017 to 2024, station BT-1 showed statistically significant decreasing trends in PM_{10} only in April and August. The p-values were close to 0.02 and 0.035, and the Sen's slope estimates were approximately −2.3 and −3.1 μg/m³ per year, respectively. These results suggest consistent reductions in PM_{10} concentrations during these two months throughout the study period. In contrast, for all other months, p-values exceed 0.05, suggesting no significant trends. However, several months indicated small negative Sen's slopes, indicating weak, though not significant, decreases.

For IS-2, June is the only month between 2017 and 2024 with a statistically significant decreasing trend (p = 0.019 and Sen's slope = −1.3 μg/m³ per year). The remaining months during this period show moderate negative slopes, but no significant monotonic trend (p > 0.05).

At SV-1 and SV-2, none of the months between 2017 and 2024 exhibit statistically significant trends. This implies that monthly PM_{10} variability from 2017 to 2024 is dominated by interannual fluctuations rather than systematic changes (Table 3).

At station IS-2, monthly $PM_{2.5}$ concentrations showed statistically significant decreases in February, March, April, and June, with Sen's slopes ranging from −2.25 to −5.09 μg/m³/year during the period from 2017 to 2024. The remaining months exhibited no significant trends.

Table 3 Results of the Mann–Kendall test and Sen's slope estimator for monthly trends of PM_{10} concentrations.

Station	Month	n	S	Z	p-value	Significant trend (α = 0.05)	Sen slope (µg/m³/year)
BT-1	January	8	-16	-1.86	0.063	No significant trend	-3.07
	February	8	-14	-1.61	0.108	No significant trend	-1.38
	March	8	-8	-0.87	0.386	No significant trend	-1.47
	April	8	-20	-2.35	0.019	Yes (Decreasing trend)	-2.32
	May	8	0	0.00	1.000	No significant trend	-0.05
	June	8	-12	-1.36	0.174	No significant trend	-1.03
	July	8	-6	-0.62	0.536	No significant trend	-0.80
	August	8	-18	-2.10	0.035	Yes (Decreasing trend)	-3.08
	September	8	-2	-0.12	0.902	No significant trend	-0.47
	October	8	-4	-0.37	0.711	No significant trend	-0.29
	November	8	-14	-1.61	0.108	No significant trend	-1.81
	December	8	-12	-1.36	0.174	No significant trend	-1.91
IS-2	January	7	-9	-1.20	0.230	No significant trend	-2.16
	February	8	-14	-1.61	0.108	No significant trend	-2.47
	March	8	-12	-1.36	0.174	No significant trend	-1.78
	April	8	-8	-0.87	0.386	No significant trend	-0.92
	May	8	-8	-0.87	0.386	No significant trend	-1.52
	June	8	-20	-2.35	0.019	Yes (Decreasing trend)	-1.29
	July	8	-8	-0.87	0.386	No significant trend	-0.74
	August	8	-6	-0.62	0.536	No significant trend	-0.89
	September	8	-8	-0.87	0.386	No significant trend	-0.48
	October	8	-12	-1.36	0.174	No significant trend	-0.75
	November	8	-2	-0.12	0.902	No significant trend	-0.34
	December	8	-14	-1.61	0.108	No significant trend	-1.29
SV-1	January	8	-10	-1.11	0.266	No significant trend	-1.49
	February	8	-10	-1.11	0.266	No significant trend	-1.15
	March	8	-16	-1.86	0.063	No significant trend	-0.95
	April	8	2	0.12	0.902	No significant trend	0.06
	May	8	0	0.00	1.000	No significant trend	0.05
	June	8	-4	-0.37	0.711	No significant trend	-0.14
	July	8	4	0.37	0.711	No significant trend	0.10
	August	8	-4	-0.37	0.711	No significant trend	-0.39
	September	8	0	0.00	1.000	No significant trend	0.07
	October	8	-8	-0.87	0.386	No significant trend	-1.19
	November	8	0	0.00	1.000	No significant trend	-0.12
	December	8	-4	-0.37	0.711	No significant trend	-0.71
SV-2	January	6	-3	-0.38	0.707	No significant trend	-3.11
	February	6	-7	-1.13	0.260	No significant trend	-1.18

Station	Month	n	S	Z	p-value	Significant trend (α = 0.05)	Sen slope (μg/m³/year)
	March	6	-3	-0.38	0.707	No significant trend	-0.78
	April	6	3	0.38	0.707	No significant trend	0.65
	May	6	3	0.38	0.707	No significant trend	0.20
	June	6	3	0.38	0.707	No significant trend	0.48
	July	6	-3	-0.38	0.707	No significant trend	-0.80
	August	6	-11	-1.88	0.060	No significant trend	-2.56
	September	5	0	0.00	1.000	No significant trend	-0.32
	October	5	-2	-0.24	0.806	No significant trend	-1.53
	November	5	2	0.24	0.806	No significant trend	1.19
	December	5	-4	-0.73	0.462	No significant trend	-1.27

Statistically significant decreasing trends in $PM_{2.5}$ at station SV-1 were observed only in March and April ($p = 0.024$), with Sen's slope estimates of approximately −2.5 and −1.2 μg/m³ per year, respectively. These results suggest a decrease in $PM_{2.5}$ concentrations at spring throughout the study period. In all other months, p-values exceed 0.05, and the time series are classified as exhibiting no significant trend. However, most months show slightly negative Sen's slopes (approximately −0.3 to −1 μg/m³ per year), suggesting weak, non-significant downward tendencies.

There was insufficient data from station BT-1 during the common analysis period (2017–2024) to allow for estimation of the $PM_{2.5}$ trend (Table 4).

Table 4 Results of the Mann–Kendall test and Sen's slope estimator for monthly trends of $PM_{2.5}$ concentrations, 2017-2024.

Station	Month	n	S	Z	p-value	Significant trend (α = 0.05)	Sen slope (μg/m³/year)
IS-2	January	6	-11	-1.88	0.060	No significant trend	-5.80
	February	6	-13	-2.25	0.024	Yes (Decreasing trend)	-5.09
	March	6	-13	-2.25	0.024	Yes (Decreasing trend)	-5.03
	April	6	-13	-2.25	0.024	Yes (Decreasing trend)	-4.18
	May	6	-9	-1.50	0.133	No significant trend	-2.90
	June	6	-13	-2.25	0.024	Yes (Decreasing trend)	-2.25
	July	6	-11	-1.88	0.060	No significant trend	-1.25
	August	6	-9	-1.50	0.133	No significant trend	-1.24
	September	6	-9	-1.50	0.133	No significant trend	-0.85
	October	6	-7	-1.13	0.260	No significant trend	-2.43
	November	6	-11	-1.88	0.060	No significant trend	-1.33
	December	6	-9	-1.50	0.133	No significant trend	-3.01
SV-1	January	5	-4	-0.73	0.462	No significant trend	-1.94
	February	6	-5	-0.75	0.452	No significant trend	-0.55
	March	6	-13	-2.25	0.024	Yes (Decreasing trend)	-2.48
	April	6	-13	-2.25	0.024	Yes (Decreasing trend)	-1.18
	May	6	-1	0.00	1.000	No significant trend	-0.59

Station	Month	n	S	Z	p-value	Significant trend (α = 0.05)	Sen slope (μg/m³/year)
	June	6	-7	-1.13	0.260	No significant trend	-0.87
	July	6	-5	-0.75	0.452	No significant trend	-0.26
	August	5	-2	-0.24	0.806	No significant trend	-0.40
	September	6	3	0.38	0.707	No significant trend	0.49
	October	6	-1	0.00	1.000	No significant trend	0.00
	November	6	-1	0.00	1.000	No significant trend	-0.36
	December	6	-5	-0.75	0.452	No significant trend	-0.93

At station BT-1, $PM_{2.5}$ concentrations from 2009 to 2024 showed no significant trends for most months. The only exception was June, which exhibited a slight but statistically significant increase ($p = 0.035$; Sen's slope = 0.52 μg/m³ per year), while other months displayed small slopes (−1 to +0.3 μg/m³ per year), indicating variability dominated by interannual fluctuations rather than systematic long-term changes.

At station IS-2, monthly $PM_{2.5}$ concentrations were generally stable over the same period, with a statistically significant decrease observed only in April, likely reflecting seasonal effects or changes in emission sources during spring.

At station SV-1, the number of months with significant decreases increased over 2009–2024. January, March, April, and December exhibited significant downward trends ($p < 0.05$), with Sen's slopes ranging from −1.4 to −2.2 μg/m³ per year, indicating clearer reductions during the cold season and spring. In contrast, summer and autumn months (May–November) showed no significant trends and only small negative slopes, suggesting no substantial decreases during these months (Table 5).

Table 5 Results of the Mann–Kendall test and Sen's slope estimator for monthly trends of $PM_{2.5}$ concentrations, 2009-2024.

	Month	n	S	Z	p-value	Significant trend (α = 0.05)	Sen slope (μg/m³/year)
BT-1	January	8	-2	-0.12	0.902	No significant trend	-0.37
	February	8	4	0.37	0.711	No significant trend	0.29
	March	8	-8	-0.87	0.386	No significant trend	-1.01
	April	8	-4	-0.37	0.711	No significant trend	-0.17
	May	8	0	0.00	1.000	No significant trend	-0.02
	June	8	18	2.10	0.035	Yes (Increasing trend)	0.52
	July	7	1	0.00	1.000	No significant trend	0.31
	August	7	-13	-1.80	0.072	No significant trend	-1.29
	September	7	1	0.00	1.000	No significant trend	0.10
	October	8	2	0.12	0.902	No significant trend	0.12
	November	8	-2	-0.12	0.902	No significant trend	-0.10
	December	8	-8	-0.87	0.386	No significant trend	-1.12
IS-2	January	14	-13	-0.66	0.511	No significant trend	-0.38
	February	14	3	0.11	0.913	No significant trend	0.10
	March	14	-11	-0.55	0.584	No significant trend	-0.58
	April	14	-47	-2.52	0.012	Yes (Decreasing trend)	-0.82

	Month	n	S	Z	p-value	Significant trend (α = 0.05)	Sen slope (μg/m³/year)
	May	14	-15	-0.77	0.443	No significant trend	-0.15
	June	14	-11	-0.55	0.584	No significant trend	-0.11
	July	14	-7	-0.33	0.743	No significant trend	-0.06
	August	14	-7	-0.33	0.743	No significant trend	-0.17
	September	14	-21	-1.09	0.274	No significant trend	-0.30
	October	14	5	0.22	0.827	No significant trend	0.03
	November	13	-8	-0.43	0.669	No significant trend	-0.25
	December	13	-8	-0.43	0.669	No significant trend	-0.35
SV-1	January	9	-22	-2.19	0.029	Yes (Decreasing trend)	-1.99
	February	10	-15	-1.25	0.210	No significant trend	-0.75
	March	10	-23	-1.97	0.049	Yes (Decreasing trend)	-1.68
	April	10	-35	-3.04	0.002	Yes (Decreasing trend)	-1.36
	May	10	-17	-1.43	0.152	No significant trend	-0.38
	June	10	-15	-1.25	0.210	No significant trend	-0.31
	July	10	-3	-0.18	0.858	No significant trend	-0.01
	August	9	-14	-1.36	0.175	No significant trend	-0.82
	September	10	-9	-0.72	0.474	No significant trend	-0.55
	October	10	-15	-1.25	0.210	No significant trend	-0.70
	November	10	-11	-0.89	0.371	No significant trend	-0.79
	December	10	-29	-2.50	0.012	Yes (Decreasing trend)	-2.20

Similar to the results obtained in northeastern Romania, recent studies from several European urban areas have reported significant downward trends in PM_{10} and $PM_{2.5}$ concentrations. These trends are attributed to improvements in air quality policies and the modernization of heating and transportation sources (Andersen et al., 2025; Chen et al., 2024; Garcia-Marles et al., 2024).

3.1.3. Annual Trend Analysis of temperatures

Analysis of annual mean temperature trends across all monitoring stations (SV-1, SV-2, BT-1, and IS-2) reveals no statistically significant monotonic trends, with p-values consistently exceeding the 0.05 significance level. Although Sen's slope estimates suggest slight positive tendencies at SV-1, BT-1, and IS-2 (approximately 0.19–0.23°C/year), these increases are not statistically significant. The relative stability of local thermal conditions is important for understanding the particulate matter analysis. It suggests that the significant decreasing trends in PM_{10} and $PM_{2.5}$ at stations such as BT-1 and IS-2 are more likely due to reductions in anthropogenic emissions or improved environmental management than to substantially warmer winters or major changes in the regional climate (Table 6).

Table 6 Results of the Mann–Kendall test and Sen's slope estimator for annual trends of temperature, 2017-2024.

Station	n	S	Z	p-value	Significant trend (α = 0.05)	Sen's slope (μg/m³/year)
SV-1	8	8	0.87	0.387	No significant trend	0.19
SV-2	5	-2	-0.24	0.807	No significant trend	-0.07
BT-1	8	14	1.61	0.108	No significant trend	0.23
IS-2	8	13	1.50	0.135	No significant trend	0.19

3.2. Diurnal and Seasonal Variations of hourly PM_{10} concentrations, 2017 - 2024

Examining diurnal variations helps to identify daily patterns of PM_{10} concentrations and the influence of short-term emission sources, such as traffic or residential heating, which are not captured by long-term trend analysis.

Hourly data for the $PM_{2.5}$ fraction is only available at the IS-2 station, and only starting in February 2022. This substantially limits the spatial coverage required for a robust analysis of diurnal and seasonal variations. Consequently, this subsection focuses on hourly PM_{10} concentrations, for which continuous and comparable time series are available for all three urban areas from 2017 to 2024. $PM_{2.5}$ concentrations are expected to exhibit similar diurnal and seasonal patterns to PM_{10}, consistent with the seasonal variation of the $PM_{2.5}/PM_{10}$ ratio reported in a previous study (Drăgoi (Oniu) et al., 2025).

The analysis of hourly PM_{10} concentrations from 2017 to 2024 shows a bimodal diurnal cycle with morning (around 8–9 a.m.) and evening (around 8–11 p.m.) peaks. These peaks are more pronounced at IS-2 and SV-2 and less pronounced at SV-1 and BT-1 (Figures 2 and 3). The morning peaks mainly coincide with increased traffic and, during the cold season, residential heating. The evening peaks reflect evening traffic, residential activities, and the nocturnal decrease in planetary boundary layer height. This decrease favors the accumulation of particles near the ground. The stronger peaks at IS-2 and SV-2 suggest a greater influence of local sources, such as traffic at IS-2 and industrial activity and traffic at SV-2. This is consistent with studies that report higher concentrations and more pronounced diurnal amplitudes at urban sites with intense traffic or industrial activity compared to urban background stations where regional contributions dominate (Asma et al., 2022; Bathmanabhan et al., 2010; Eskandari et al., 2020; Kassomenos et al., 2014; Ravish & Kashyap, 2025) as well as the evolution of the atmospheric boundary layer, which determines pollutant dispersion (Kim & Kim, 2020; Liu et al., 2014;).

Figure 2 further illustrates the combined seasonal and diurnal variation at all four stations.

At IS-2, high hourly PM_{10} values were observed in February–March between 7:00 and 11:00 a.m., as well as in the evening (20:00–23:00) during winter and early spring, reflecting the influence of evening traffic and residential activities under conditions unfavorable for dispersion.

At SV-2, an industrial-type urban station, the highest hourly concentrations occurred in late autumn and winter, mainly in the evening and night hours (around 20:00–23:00), highlighting the additional contribution of industrial emissions and heating systems to elevated PM_{10} episodes.

Urban background stations SV-1 and BT-1 showed generally lower concentrations and a smoother seasonal pattern, with a smaller contrast between low-background periods in summer and high-episode periods in late autumn and winter evenings.

At all stations, minimum concentrations occurred during summer between 11:00 and 16:00, when the planetary boundary layer is maximal and pollutant dispersion is most efficient. Elevated concentrations in the cold months, particularly in the evening, are consistent with a reduced mixing layer height and stable atmospheric conditions that favor the accumulation of pollutants near the ground.

Overall, these multi-year averages indicate that the diurnal and seasonal structure of PM_{10} is temporally robust, reflecting a relatively stable regime of anthropogenic activities and meteorological conditions characteristic of the studied urban areas.

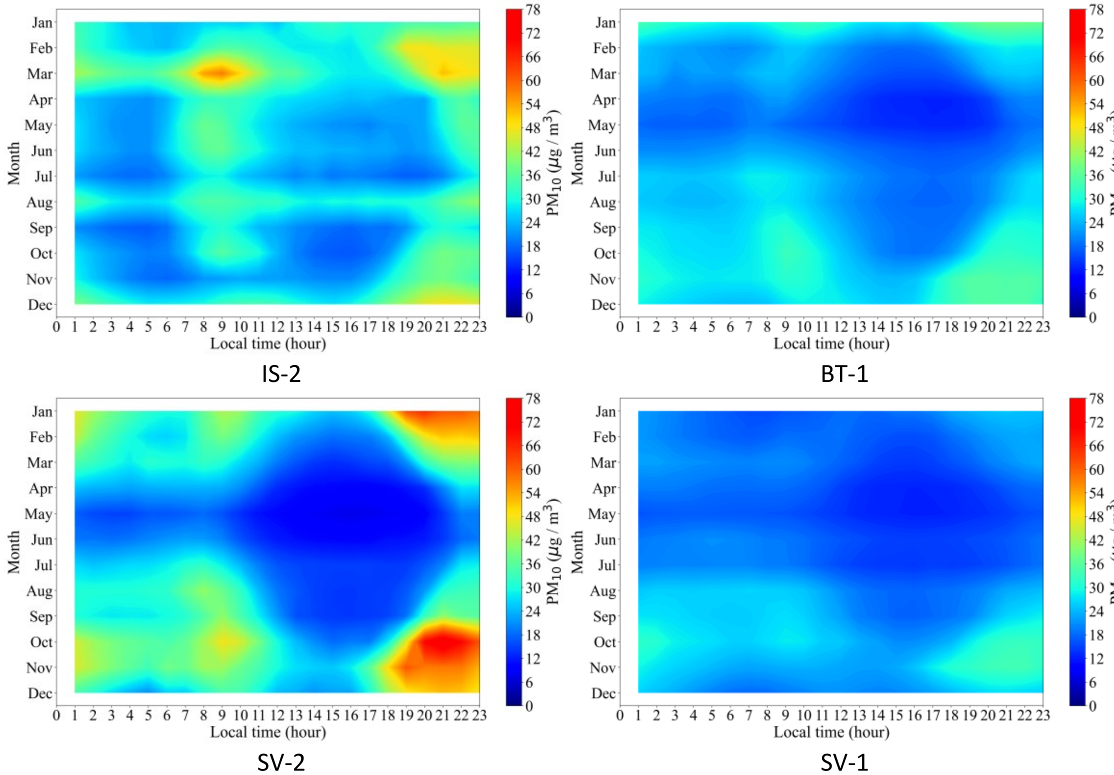

Figure 2 Monthly mean hourly PM_{10} concentrations in the study area, 2017–2024.

The multi-year (2017–2024) average of hourly PM_{10} concentrations shows a pronounced diurnal pattern and clear distinctions between weekdays and weekends at all four stations (BT-1, IS-2, SV-1, and SV-2). During the nighttime hours (1:00–6:00 a.m.), values are slightly higher on weekends, indicating a relatively greater contribution from residential sources and nighttime activities, such as the use of heating systems and the occasional burning of biomass. These activities remain active under very stable and shallow boundary layer conditions that favor particle accumulation. After 6:00 a.m., mean hourly concentrations on weekends consistently remain below weekday concentrations, particularly during peak periods (7:00–10:00 a.m. and 21:00–23:00), reflecting a significant reduction in traffic to public institutions, schools, and offices. Consequently, there is a decrease in PM_{10} emissions associated with road transport. Road transport is recognized as the main driver of diurnal peaks in European urban environments (Adler et al., 2023; Girotti et al., 2025; Vecchi et al., 2007) .

Diurnal minima occur between 11:00 a.m. and 4:00 p.m. at all stations. This coincides with the development of the convective boundary layer and enhanced vertical mixing. Thus, differences in emissions between weekdays and weekends are reflected almost linearly in lower weekend concentrations. This pattern aligns with literature on the "weekend effect" for PM_{10} and other primary pollutants. A pronounced peak reappears at night, between 21:00 and 23:00, especially at SV-2. Mean hourly concentrations during weekdays exceed 40 μg/m³ at SV-2, suggesting a synergy between return traffic, increased residential heating, and the re-establishment of nocturnal atmospheric stability. On weekends, a similar peak is observed, albeit with smaller amplitude, which confirms the dominant role of daily mobility in controlling PM_{10} peaks (Adame et al., 2014; Adler et al., 2023; Bathmanabhan et al., 2010).

When compared across stations, SV-2 shows the highest concentrations and the largest diurnal amplitude. This is due to its proximity to heavily traffic roads and/or areas with dense residential heating. In contrast, SV-1 consistently shows lower levels, which is consistent with an urban background or peri-urban station. BT-1 and IS-2 show intermediate levels with well-defined "two-peak" profiles. Slightly higher values at IS-2 suggest stronger local emission pressure from traffic and/or commercial activities, consistent with similar studies in medium-sized urban areas. Overall, the figure depicts a robust diurnal PM_{10} pattern (Figure 3).

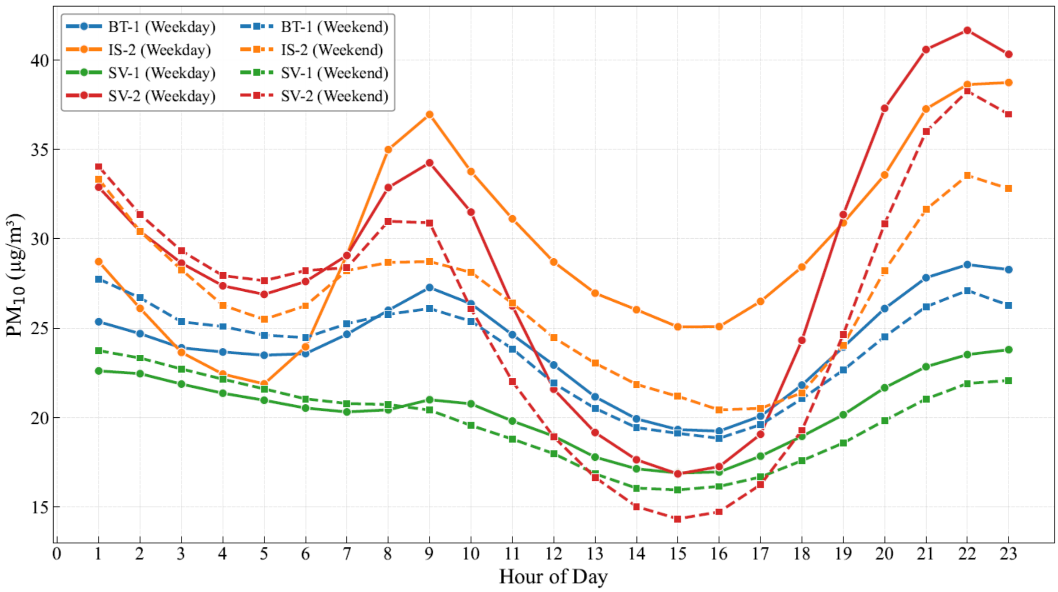

Figure 3 Diurnal variation of PM_{10} concentrations on weekdays and weekends, 2017–2024.

Variations in traffic and residential sources, modulated by the evolution of the atmospheric boundary layer, explain the differences between weekdays and weekends, as well as the spatial contrasts between stations. This contributes to our understanding of particulate exposure mechanisms in urban areas of the region (Adler et al., 2023; García et al., 2018; Hilly et al., 2025; Otmani et al., 2024).

3.3. Assesment of compliance of PM_{10} and $PM_{2.5}$ concentrations with limit values

During the analyzed period, the daily PM_{10} limit value for human health (50 µg/m³, not to be exceeded more than 35 times per calendar year) was reached at SV-2 in 2018 – 2019 and exceeded at IS-2 from 2017 to 2019, reaching the limit value again in 2020. At BT-1 and SV-1, the daily exceedances was never reached.

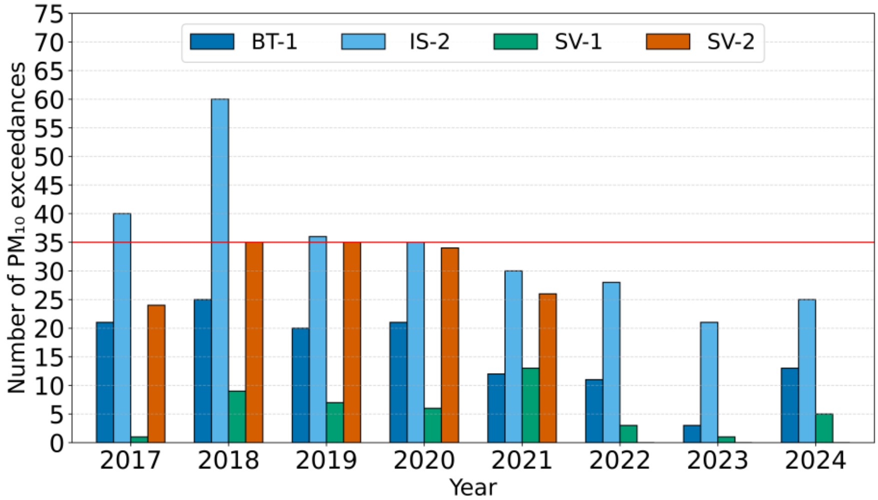

Figure 4 Evolution of the number of daily PM_{10} exceedances (2017–2024) - (The red horizontal line: the maximum number of exceedances per calendar year, Law 104/2011).

From 2017 to 2024, the annual mean PM_{10} concentrations were generally below the applicable annual limit value of 40 μg/m³. Compliance with the future 20 μg/m³ limit value established by Directive (EU) 2024/2881 was observed only at SV-1 in 2022–2023 (Figure 5). The provisions of this new directive are scheduled to be implemented into Romanian legislation by December 2026 (Figure 5).

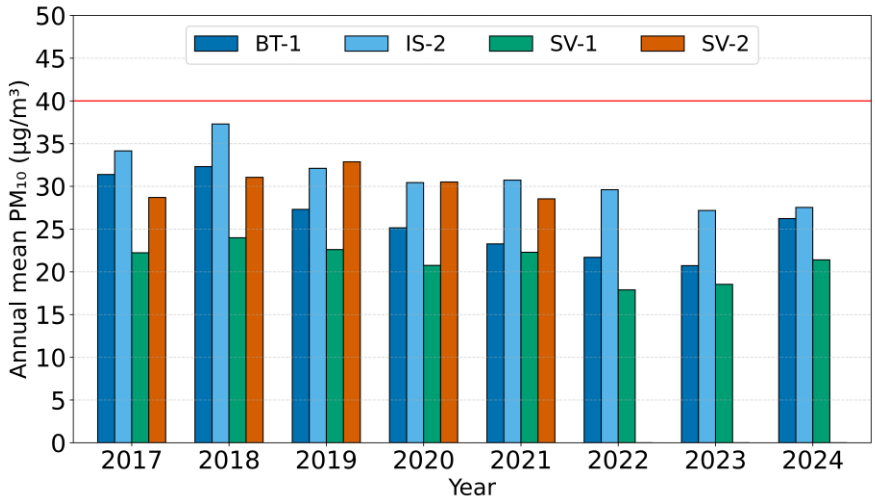

Figure 5 Temporal evolution of annual mean PM_{10} levels in relation to the annual limit value, 2017–2024 (Red line: annual limit value, Law 104/2011).

During the analyzed period, the annual mean $PM_{2.5}$ concentrations were generally below the applicable annual limit value of 25 μg/m³, except at IS-2 in 2017–2018 (Figure 6). However, even in these years, the concentrations remained above 10 μg/m³, which is the limit value established by Directive (EU) 2024/2881.

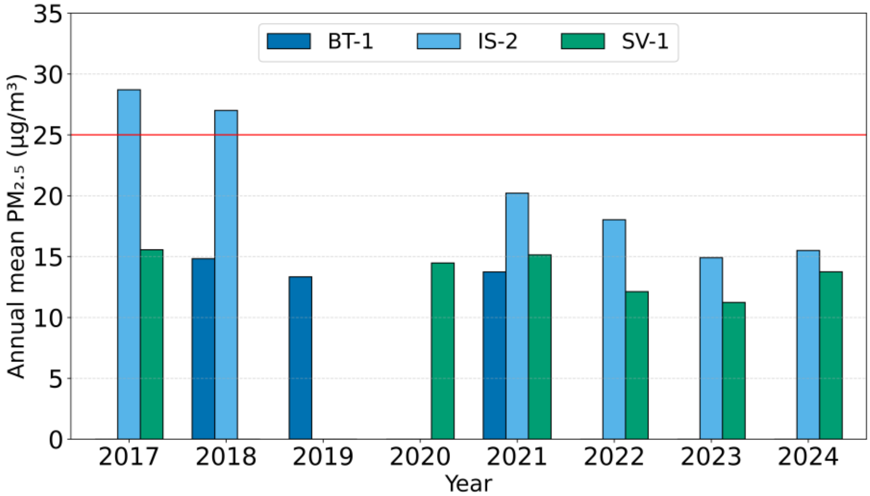

Figure 6 Temporal evolution of annual mean $PM_{2.5}$ levels in relation to the annual limit value, 2017–2024 (Red line: annual limit value, Law 104/2011).

4. Conclusion

Long-term analysis of PM_{10} and $PM_{2.5}$ concentrations at the four monitoring stations indicates that annual trends showed significant decreases in PM_{10} at BT-1 and IS-2, and in $PM_{2.5}$ at IS-2, while SV-1 and SV-2 exhibited non-significant trends.

PM_{10} showed significant declines only at BT-1 (April and August) and IS-2 (June), while other months and stations displayed small, non-significant negative slopes, reflecting interannual variability rather than systematic change.

$PM_{2.5}$ exhibited more pronounced decreases at IS-2 (February–April and June) and SV-1 (March–April), whereas BT-1 remained largely stable, except for a slight increase in June over 2009–2024. Summer and autumn months generally showed no significant trends.

Overall, reductions in particulate matter are seasonal and site-specific, influenced by emission sources and meteorological conditions. These findings suggest that while some improvements in air quality are evident, systematic year-round decreases are not consistent, highlighting the need for targeted, seasonally adjusted mitigation measures.

Diurnal cycles were clearly bimodal at all stations, with morning and evening peaks and midday lows. Weekend concentrations were generally lower than on weekdays, reflecting consistent temporal patterns. Seasonally, the highest concentrations occurred in late autumn and winter and the lowest occurred in the summer months. SV-2, the industrial urban station, consistently recorded the highest concentrations and largest diurnal amplitudes, followed by IS-2. The urban background stations, SV-1 and BT-1, showed lower and smoother profiles. Overall, these findings demonstrate that PM concentrations in urban areas are governed by combined diurnal, seasonal, and spatial dynamics and that long-term monitoring is important to detect meaningful trends and assess exposure patterns.

In 2024, PM_{10} and $PM_{2.5}$ levels remained below health-based limits, but meeting the stricter standards of Directive (EU) 2024/2881 will require further efforts.

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