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PHD THESIS ABSTRACT

*Analysis of PM₁₀ and PM_{2.5} Concentration Variability and Development of
Air Quality Indicators in Relation to European Air Quality Standards*

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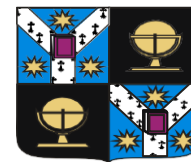
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Abbreviations

cRMSD	centered Root Mean Square Deviation
cRMSDN	centered Root Mean Square Deviation Normalised
CAMS	Copernicus Atmosphere Monitoring Service
CO	Carbon monoxide
CV	Coefficient of variance
COD	Coefficient of divergence
DE	Number of daily exceedances
EEA	European Environment Agency
EC	Excess Concentration
EI	Episode Index
LV	Limit value
MMAF	Ministry of the Environment, Water, and Forests (Ministerul Mediului, Apelor și Pădurilor)
NO_x	Nitrogen oxides
NO₂	Nitrogen dioxide
NO	Nitric oxide
P75	75th percentile
P90,4	90.4th percentile
PM	Particulate matter
PM₁₀	Particulate matter with an aerodynamic diameter $\leq 10 \mu\text{m}$
PM_{2.5}	Particulate matter with an aerodynamic diameter $\leq 2.5 \mu\text{m}$
PM_{2.5-10}	Particulate matter with diameters between $2.5 \mu\text{m}$ and $10 \mu\text{m}$.
R	Pearson Correlation Coefficient
R²	Coefficient of Determination
MAE	Mean Absolute Error
RMSE	Root Mean Square Error
RNMCA	National Air Quality Monitoring Network
SO₂	Sulfur Dioxide
TLI	Toxic Load Index
UE	European Union
UNECE	United Nations Economic Commission for Europe
U.S. EPA	U.S. Environmental Protection Agency
WHO	World Health Organization

Introduction

Particulate matter is currently considered the most reliable indicator of the health effects of ambient air pollution and is also regarded as one of the most harmful air pollutants (Burnett et al., 2014; Costa et al., 2014). Among these particles, fractions with an aerodynamic diameter below 10 μm (PM_{10}) and below 2.5 μm ($\text{PM}_{2.5}$) are particularly relevant in terms of their impact on human health. Numerous epidemiological studies have demonstrated their association with an increased incidence of respiratory and cardiovascular diseases, lung cancer, and premature mortality (EEA 2020; EEA, 2025).

Research Hypothesis

The central hypothesis of this thesis is that the indicators established under Directive (EU) 2024/2881 do not fully capture population exposure or the severity of particulate pollution episodes in relation to limit values and alert thresholds. The complementary indicators proposed in this study: *Excess Concentration (EC)*, *Episode Index (EI)*, and *Toxic Load Index (TLI)*, integrate information on concentration variability, the $\text{PM}_{2.5}/\text{PM}_{10}$ ratio, and exceedances of regulatory limit values. These indicators are expected to provide a more comprehensive characterization of ambient air quality and a more informative assessment of the actual gap between observed pollution levels and the air quality objectives established under Directive (EU) 2024/2881.

Research Aim

The overall aim of this thesis is to advance the understanding of particulate matter pollution through an analysis of the temporal and spatial variability of PM_{10} and $\text{PM}_{2.5}$ concentrations and the $\text{PM}_{2.5}/\text{PM}_{10}$ ratio, while also developing complementary indicators for characterizing population exposure in the context of the transition from Directive 2008/50/EC to Directive (EU) 2024/2881.

To achieve this aim, the following specific objectives are pursued:

- a) to analyse the temporal variability and trends in PM_{10} and $\text{PM}_{2.5}$ concentrations and to quantify medium-term trends using the Mann-Kendall test and Sen's slope estimator, in order to characterize the direction and magnitude of recent changes in particulate matter concentrations;
- b) to investigate the relationships between PM_{10} , $\text{PM}_{2.5}$, and gaseous pollutants (NO_2 , SO_2 , CO, and benzene) using correlation analysis, diurnal variation analysis, and regression models, with the aim of identifying common emission sources and patterns of multi-pollutant exposure;
- c) to investigate the spatio-temporal variability of the $\text{PM}_{2.5}/\text{PM}_{10}$ ratio as an indicator of particulate fraction characteristics and potential source influences;
- d) to assess the performance and potential of satellite-derived products for estimating PM_{10} and $\text{PM}_{2.5}$ concentrations by comparing satellite-based estimates with *in situ* measurements;
- e) to assess PM_{10} and $\text{PM}_{2.5}$ concentrations against the daily and annual limit values established under the current regulatory framework and the requirements introduced by the new European air quality legislation;
- f) to develop, substantiate, and justify the complementary indicators *Excess Concentration (EC)*, *Episode Index (EI)*, and *Toxic Load Index (TLI)*;

g) to apply and extend the proposed indicators at local and national scales in order to assess their applicability and relevance for characterizing population exposure and particulate matter pollution episodes.

Research Methodology

The research methodology integrates several types of analytical tools and approaches: statistical analysis of PM₁₀ and PM_{2.5} time series at daily resolution (and, where available, at hourly resolution), calculation and interpretation of the PM_{2.5}/PM₁₀ ratio as an indicator of particulate fraction structure, assessment of the performance and capacity of satellite-derived products to provide estimates of particulate matter concentrations, comparison of satellite-based estimates with *in situ* measurements using standard performance metrics, assessment of the data series against the requirements of the current and forthcoming regulatory frameworks, development and application of the *EC*, *EI*, and *TLI* indicators; sensitivity analysis of the classification thresholds and extension of the indicators to the national scale using data from the National Air Quality Monitoring Network (RNMCA).

The study focuses on three cities in north-eastern Romania: Suceava, Iași, and Botoșani, for which air quality data from four monitoring stations within the RNMCA are analysed: three urban background stations and one industrial station located in an urban area.

The main analysis of the variability and trends in PM₁₀ and PM_{2.5} concentrations is based on data covering the 2017-2024 period. The variability of the PM_{2.5}/PM₁₀ ratio was assessed for 2019-2023, in accordance with the initial research design. The complementary exposure indicators were defined and calculated for the 2019-2024 period, while the analysis of satellite data was conducted for 2024. The indicators were subsequently assessed at the national scale using records from RNMCA monitoring stations where PM₁₀ and PM_{2.5} concentrations were measured concurrently, covering the 2015-2025 period.

The dataset used in this study presents certain limitations, primarily due to the variable degree of completeness across years and monitoring stations. This limitation may affect the representativeness of the analyses from both temporal and spatial perspectives. In addition, variability in meteorological conditions influences the dispersion and accumulation of atmospheric pollutants and may result in interannual variations that are independent of changes in emission sources. Therefore, the results should be interpreted in light of these sources of uncertainty and the specific characteristics of the dataset.

Chapter 1. Theoretical Framework and State of the Art

1.1. Conceptual Framework

Particulate matter is one of the most important and harmful atmospheric pollutants and a major indicator of the adverse health effects associated with ambient air pollution (Burnett et al., 2014; Costa et al., 2014; EEA, 2020; EEA, 2025).

Particulate matter consists of a complex mixture of solid and liquid particles originating from both natural and anthropogenic sources. These particles can persist in the atmosphere and may be transported over considerable distances (Seinfeld, 2016; WHO, 2006; WHO, 2021). They are primarily classified according to their aerodynamic diameter, with the most relevant fractions being PM₁₀, PM_{2.5}, and ultrafine particles.

Particle size, chemical composition, hygroscopic properties, and optical characteristics influence their atmospheric behaviour, deposition within the respiratory system, and effects on human health and ecosystems (U.S. EPA, 2019; Directiva (UE) 2024/2881; WHO, 2006).

In the atmosphere, particulate matter undergoes continuous physical and chemical transformation processes and is removed through gravitational settling, dry deposition onto surfaces, and wet deposition associated with precipitation. Depending on their origin, particles can be classified as primary, when emitted directly into the atmosphere, or secondary, when formed through chemical reactions involving gaseous precursors such as NH₃, SO₂, and NO_x. Emissions of these precursor gases contribute to the formation of PM₁₀ and PM_{2.5}, a process that is generally more pronounced during the cold season as a result of increased heating-related emissions (Harrison et al., 1997; Seinfeld, 2016; Jimenez et al., 2009; Putaud et al., 2010).

The composition of particulate matter may include nitrates, sulfates, ammonium, and chlorides, as well as elemental carbon, heavy metals, and toxic organic compounds, some of which have carcinogenic properties (Liu et al., 2020; Belis et al., 2013; Hopke et al., 2020; Seinfeld, 2016; Volná and Blažek, 2024). In terms of their sources, particulate matter originates from both natural sources, such as mineral dust, sea spray, pollen, spores, wildfires, and volcanic activity, and anthropogenic sources (Carslaw et al., 2010; Marelli, 2007; WHO, 2006).

Mineral matter is an important component of atmospheric particulate matter and contributes particularly to PM₁₀ concentrations. Silicates, carbonates, oxides, and phosphates are predominantly found in the coarse PM_{2.5-10} fraction, although mineral particles generated by abrasion processes may also contribute to the fine PM_{2.5} fraction (Querol et al., 2001; Putaud et al., 2004; Querol et al., 2004).

Among the most important anthropogenic sources of particulate matter are road traffic, fuel combustion in residential, energy, commercial, and institutional facilities, as well as various industrial processes (EEA, 2019; Gualtieri et al., 2015; Park et al., 2018).

The adverse effects of particulate matter on human health depend on particle size and concentration, chemical composition, duration of exposure, and individual susceptibility (Dettori et al., 2021; Park et al., 2018; Philip et al., 2017).

Coarse particles are predominantly deposited in the upper respiratory tract and are associated with conditions such as asthma and chronic obstructive pulmonary disease, as well as with increased hospital admissions for respiratory (Ai et al., 2019; Mohebbichamkhorami et al., 2020) and cardiovascular diseases (Cheng et al., 2021; Miller, 2022).. Due to their smaller size, PM_{2.5} particles can penetrate more deeply into the respiratory system, reaching the alveolar region, where they may induce inflammatory processes and toxic effects.

Exposure to PM_{2.5} is associated with an increased risk of mortality, respiratory diseases (Liu et al., 2019; Park et al., 2021; Pražnikar and Pražnikar, 2012), cardiovascular diseases, cancer (Pražnikar and Pražnikar, 2012; Taj et al., 2021), and stroke (Committee on the Medical Effects of Air Pollutants, 2011; WHO, 2021). There is also evidence of potential effects on the nervous system (Ke et al., 2022). Episodes characterized by very high particulate matter concentrations, such as those associated with wildfires, dust storms, or industrial emissions, may cause acute health effects and increase hospital admissions. Even modest reductions in PM_{2.5} concentrations may contribute significantly to reducing mortality and improving population health (Oudin et al., 2024).

1.2. International Methodological Approaches

In recent years, numerous international studies have used extended time series of measurements from fixed monitoring stations to characterize the evolution of PM₁₀ and PM_{2.5} concentrations, assess compliance with air quality standards, and identify long-term trends. These analyses have been linked to emission reduction measures and changes in urban behaviour and mobility patterns, including decreases in annual mean concentrations observed after 2020 in the context of COVID-19-related restrictions and the implementation of sustainable urban mobility measures (Mecca et al., 2024; Pala et al., 2022).

Some studies have shown that, although annual mean PM₁₀ concentrations in urban agglomerations across Central Europe are often below 40 µg/m³, the number of exceedances of the daily limit value of 50 µg/m³ continues to exceed the level permitted under European legislation. These findings highlight the importance of detailed analyses of the temporal variability of PM₁₀ concentrations and the influence of meteorological conditions on pollution episodes (Ferenczi et al., 2021).

One category of studies is based on data from ground-based monitoring networks, in which PM₁₀ and PM_{2.5} time series are analysed using linear or multiple regression and time-series models. These approaches are used to assess the influence of meteorological parameters, local emission sources, and seasonality on variations in particulate matter concentrations, as well as the relationships between particulate matter and other gaseous pollutants (Ganguly et al., 2019; Girotti et al., 2025; Loganathan et al., 2022; Manevska et al., 2025). These studies also frequently include long-term trend analyses, using the Mann-Kendall test to assess the statistical significance of trends and Sen's slope estimator to quantify their magnitude (Bosch et al., 2025; Mahesh and Sivakumar, 2025; Sen et al., 2019; Umoh et al., 2022).

Recent studies have employed multiple nonlinear regression models to describe the relationship between daily PM₁₀ concentrations and an extended set of explanatory variables, resulting in equations with potential for operational application in particulate matter pollution forecasting.

To distinguish meteorological influences from those associated with emissions, some studies have applied the Kolmogorov-Zurbenko (KZ) filter to PM₁₀ time series to extract trend components and assess meteorological effects (Ferenczi et al., 2021). In the present thesis, regression models are used to quantify the relationships between PM concentrations and meteorological parameters without applying the KZ filter, following a similar conceptual framework for distinguishing meteorological and emission-related influences.

European and national guidelines and regulations have promoted the integration of monitoring networks, dispersion models, and objective estimation methods, supported the development of integrated particulate matter monitoring and forecast systems.

Another approach combines ground-based measurements with satellite data and spatial information on land use and traffic, enabling PM_{2.5} and PM₁₀ concentration estimates at high spatio-temporal resolution, including in poorly monitored areas (Buchard et al., 2016; Diao et al., 2020; Li et al., 2015; Sannigrahi et al., 2021).

The 2020 -2026 period is characterized by the widespread use of machine learning methods and hybrid models for PM concentration estimation, including machine learning, deep learning, random forest, and ensemble models. These approaches integrate data from monitoring stations, satellites, numerical models, and geographical variables to predict PM concentrations, distinguish meteorological and emission influences, and estimate exposure-related risks (Doreswamy et al., 2020; Grange et al., 2018; Hu et al., 2025; Song et al., 2026).

In accordance with European legislative requirements for reporting the annual number of exceedances of the daily PM₁₀ limit value and delineating affected areas, some studies have developed probabilistic approaches that combine daily *kriging*-based mapping, using ground-based measurements and outputs from chemical transport models such as CHIMERE, with Poisson distributions. These methods enable the estimation, for each grid cell, of the probability that the number of days with exceedances will surpass the threshold of 35 days per year (Beauchamp et al., 2015).

Jiang et al. (2026) investigated public acceptance of interventions aimed at reducing transport-related air pollution, considering exposure to nanoparticles, environmental attitudes, moral norms, and behavioural intentions. Their findings indicate higher acceptance of interventions perceived as incentive-based compared with restrictive measures, while balanced combinations of intervention types and levels may enhance both the effectiveness and acceptance of air pollution reduction measures.

These recent methodological developments support the use, in the present thesis, of complementary indicators (*EC*, *EI*, *TLI*) and time-series analyses, in line with both current research directions and European and national legislative requirements for air quality assessment.

Chapter 2. Legislative Framework for Ambient Air Quality

Ambient air quality in Romania is regulated by *Law No. 104/2011 on ambient air quality*, as subsequently amended and supplemented, which transposes *Directive 2008/50/EC of the European Parliament and of the Council on ambient air quality and cleaner air for Europe* and *Directive 2004/107/EC of the European Parliament and of the Council relating to arsenic, cadmium, mercury, nickel and polycyclic aromatic hydrocarbons in ambient air*.

Under Law No. 104/2011, air quality must be maintained and, where possible, improved in zones and agglomerations where air quality is satisfactory. Where limit values, target values or, where applicable, long-term objectives established by the legislation are exceeded, the competent authorities must adopt the necessary measures to achieve and comply with these objectives within a specified period.

To ensure compliance with the limit values established by the applicable legislation, Law no. 104/2011 provides for the development, where appropriate, of air quality plans or integrated air quality plans. These plans include measures and actions to reduce pollutant concentrations for which exceedances have been recorded, together with implementation deadlines, so that air quality objectives can be achieved within a specified period.

Based on air quality assessment results and recorded pollutant levels, zones and agglomerations in Romania are classified into air quality management regimes according to the criteria established by national legislation. Regime I includes zones and agglomerations where the levels of one or more pollutants exceed the applicable limit or target values, requiring measures to reduce concentrations. Regime II includes zones and agglomerations where pollutant levels comply with the applicable legislative requirements, requiring measures to maintain and, where appropriate, improve air quality.

In Romania, based on air quality assessments and in accordance with Order No. 2,202/2020 approving the lists of administrative-territorial units classified under the air quality management regimes established for the zones and agglomerations covered by Law No. 104/2011, the municipality of Suceava is classified under Regime I for PM₁₀. The municipality of Iași is classified under Regime I for PM₁₀, PM_{2.5}, and nitrogen dioxide/nitrogen oxides (NO₂/NO_x), while the municipality of Botoșani is classified under Regime II for PM₁₀ and PM_{2.5}.

This classification provides the basis for establishing local air quality management measures and planning documents. Under Regime I, measures aim to reduce concentrations of pollutants for which exceedances have been recorded and achieve the levels established by legislation, whereas under Regime II, measures focus on maintaining air quality levels and preventing their deterioration.

In the context of evolving European requirements for public health protection and the reduction of air pollution, the EU legislative framework has recently been revised through the adoption of *Directive (EU) 2024/2881 of the European Parliament and of the Council of 23 October 2024 on ambient air quality and cleaner air for Europe (recast)*. The new Directive aims to strengthen the protection of public health, ecosystems, and biodiversity by introducing stricter requirements for the concentrations of major air pollutants. It establishes new limit and

target values that are closer to the levels recommended by the World Health Organization and are to be achieved by 2030. Under Article 30 of Directive (EU) 2024/2881, Member States are required to transpose the Directive into national legislation by 11 December 2026.

Therefore, although Law No. 104/2011, as subsequently amended and supplemented, represents the national legislative framework at the time of preparation of this thesis, the provisions of Directive (EU) 2024/2881 constitute the new European framework for the future revision of national ambient air quality legislation.

One of the main changes introduced by the new Directive is the tightening of requirements for permissible pollutant concentrations. Compared with the limit values established under Directive 2008/50/EC, Directive (EU) 2024/2881 sets stricter values for several air pollutants, to be achieved by 2030 (Table 2. 1).

Table 2. 1. Limit Values Established by Directives 2008/50/EC and (EU) 2024/2881 and recommended by WHO

Pollutant	Averaging period	Limit value		Level Recommended by the WHO 2021 Guidelines
		Directive 2008/50/EC (Law 104/2011)*	Directive (EU) 2024/2881**	
PM _{2.5}	Calendar year	25 µg/m ³	10 µg/m ³	5 µg/m ³
	1 day	-	45 µg/m ³ not to be exceeded more than 18 times per calendar year	45 µg/m ³ not to be exceeded more than 3-4 times per calendar year
PM ₁₀	Calendar year	40 µg/m ³	20 µg/m ³	15 µg/m ³
	1 day	50 µg/m ³ not to be exceeded more than 35 times per calendar year	45 µg/m ³ not to be exceeded more than 18 times per calendar year	45 µg/m ³ not to be exceeded more than 3-4 times per calendar year

*** to be attained by 11 December 2026 ; * * to be attained by 1 January 2030**

Directive (EU) 2024/2881 introduces information and alert thresholds for managing episodes of high PM₁₀ and PM_{2.5} pollution. Information thresholds indicate an increased risk for vulnerable and sensitive groups and require the rapid dissemination of relevant information. For PM₁₀ and PM_{2.5}, measurements are performed over one day at representative locations, covering an area of at least 100 km² or the entire zone, whichever is smaller.

Alert thresholds indicate a risk to the general population from short-term exposure and require immediate protective measures. They are assessed based on daily mean concentrations over up to three consecutive days, and their exceedance triggers a short-term action plan for public health protection.

Table 2. 2. Alert thresholds and information thresholds under Directive (EU) 2024/2881

Pollutant	Averaging period	Information threshold	Alert threshold
PM _{2.5}	1 zi	50 µg/m ³	50 µg/m ³
PM ₁₀	1 zi	90 µg/m ³	90 µg/m ³

Chapter 3. Materials and Methods

3.1. Study Area and Climatic Conditions

The study was conducted in three municipalities in north-eastern Romania: Iași, Botoșani, and Suceava, located in a region characterized by a temperate continental climate, with cold winters and warm summers.

The municipalities were selected to represent urban areas with different topographic and population-density characteristics. Their selection considered geographical location and municipality size, which may influence the distribution and intensity of air pollution.

3.2. Data Used in the Study

3.2.1. Data Measured at Air Quality Monitoring Stations

Air quality monitoring in Romania is carried out through the automated stations of the National Air Quality Monitoring Network (RNMCA), using continuous measurements at fixed locations and indicative measurements, in accordance with the provisions of Law No. 104/2011. The present study used PM₁₀ and PM_{2.5} data from RNMCA monitoring stations administered by the National Agency for Environment and Protected Areas (ANMAP), located in the municipalities of Suceava (SV-1, SV-2), Botoșani (BT-1), and Iași (IS-2). For comparative analysis, the rural background station IS-4, located in Aroneanu commune, Iași County, was also included.

In a subsequent stage of the study, presented in Section 4.6, the application of the proposed complementary indicators was extended to RNMCA stations that simultaneously monitor PM₁₀ and PM_{2.5}. Information on station locations, monitored pollutants, and recorded concentrations is available on the national air quality monitoring portal. For PM₁₀ and PM_{2.5}, measurements are performed according to standardized reference methods, with the gravimetric method specified by SR EN 12341. The analysis included annual data series with a minimum data capture of 75%.

3.2.2. Satellite Data

The CHIMERE model and the CAMS multimodel Ensemble product were evaluated by comparing PM₁₀ and PM_{2.5} concentrations with gravimetric measurements performed according to EN 12341 at the urban IS-2 monitoring station during January-December 2024. Hourly data were provided by the Copernicus Atmosphere Monitoring Service (CAMS) and aggregated to daily resolution.

CHIMERE is a regional atmospheric chemistry and transport model with an approximately 10 km spatial resolution and hourly temporal resolution. The Ensemble product represents the median of the outputs from several CAMS regional models. The use of the multimodel median helps reduce uncertainty and provides more robust estimates of particulate matter concentrations (Marécal et al., 2015).

3.3. Statistical and Analytical Methods Used

The analyses employed a combination of descriptive, comparative, correlation, regression, and non-parametric methods.

Descriptive analyses focused on the annual, monthly, and weekly variability of PM₁₀ and PM_{2.5} concentrations, as well as the diurnal and seasonal variation of PM₁₀. The PM_{2.5}/PM₁₀ ratio was also analysed in relation to season, particulate matter concentrations, and air temperature, together with its spatial variability across the monitoring stations. Compliance with legislative requirements was assessed based on the indicators established under Law No. 104/2011 and Directive (EU) 2024/2881. Comparative analyses examined the relationships between PM₁₀, NO₂, and CO, as well as comparisons between in situ observations, satellite data, and the outputs of CAMS models (CHIMERE and Ensemble). Model performance was assessed using statistical metrics including Pearson's correlation coefficient (R), root mean square error (RMSE), mean absolute error (MAE), mean bias, normalized root mean square error (NRMSE), centered root mean square difference (cRMSD), and Taylor diagrams.

Correlation analysis was used to assess the relationships between PM₁₀ and PM_{2.5} concentrations and the main meteorological parameters, with Pearson's correlation coefficient used to identify linear relationships.

Medium-term trends in PM₁₀ and PM_{2.5} concentrations were assessed using non-parametric statistical methods, namely the Mann-Kendall test to detect monotonic trends and Sen's slope estimator to quantify the rate of change. These methods were selected based on the characteristics of air pollution time series, which often exhibit asymmetric distributions, extreme values, and periods of missing data. The Mann-Kendall test is relatively robust to outliers, while Sen's slope provides a median-based estimate of the trend that is less sensitive to isolated episodes of very high concentrations, making these methods suitable for assessing recent changes in PM concentrations in an urban context.

Multiple linear regression analysis was used to assess the relationship between daily PM₁₀ concentrations and the main explanatory variables over the 2017-2024 period.

Initially, the relationships between PM₁₀ and the gaseous pollutants NO₂, CO, SO₂, and benzene were analysed. Subsequently, meteorological parameters, including temperature, relative humidity, precipitation, and atmospheric pressure, were incorporated into the models. The models enabled the simultaneous assessment of the influence of emission sources and meteorological conditions on the variability of PM₁₀ concentrations. The analysis was performed separately for the cold season (November-March) and the warm season (April-October).

3. 4. Proposed Complementary Indicators for the Assessment of PM₁₀ and PM_{2.5} Pollution

To assess compliance with the future PM₁₀ and PM_{2.5} limit values established under Directive (EU) 2024/2881 and to provide a more detailed characterization of particulate matter pollution, three complementary exposure indicators were proposed in this study: Excess Concentration (*EC*), Episode Index (*EI*), and Toxic Load Index (*TLI*).

3.4.1. Excess concentration (*EC*)

EC is defined as the cumulative annual exceedance of the applicable daily limit value for PM₁₀ or PM_{2.5}. It quantifies the magnitude of exceedances by summing, over a calendar year, the positive differences between the observed daily mean concentrations and the corresponding daily

limit value, considering only days on which the limit value is exceeded. The indicator is calculated as follows::

$$EC = \sum_{i=1}^n (C_i - VL) \quad (1)$$

unde

- C_i = daily PM concentration ($\mu\text{g}/\text{m}^3$)
- VL = daily limit value ($45 \mu\text{g}/\text{m}^3$ for PM_{10} and $25 \mu\text{g}/\text{m}^3$ for $\text{PM}_{2.5}$)
- i = number of exceedance days

The classification thresholds for the EC indicator were defined to assess population exposure to particulate matter.

Initially, the EC thresholds were used to classify the counties of north-eastern Romania into four exposure categories: low, moderate, high, and very high. These categories corresponded to EC values of <250 , $251-750$, $751-1000$, and $>1000 \mu\text{g}/\text{m}^3$ for PM_{10} , and <150 , $151-500$, $501-800$, and $>800 \mu\text{g}/\text{m}^3$ for $\text{PM}_{2.5}$, respectively.

Subsequently, the classification was refined by incorporating the number of daily exceedances (DE), using a threshold of 18 days per year. This approach enabled further differentiation of exposure categories based on exceedance frequency and provided a more detailed characterization of the spatial distribution of particulate matter exposure (**Eroare! Fără sursă de referință.**).

Table 3. 1. Exposure categories for annual excess concentrations (EC) updated.

Category / Color code	Annual EC ($\mu\text{g}/\text{m}^3$) PM_{10}	Annual EC ($\mu\text{g}/\text{m}^3$) $\text{PM}_{2.5}$	Interpretation criteria
Low/ Light green	<250 and $\text{DE} \leq 18$	<150 and $\text{DE} \leq 18$	Fewer than 18 daily exceedances and/or low-amplitude exceedances
Moderate/ Dark green	$251-750$ and $\text{DE} \leq 18$	$151-500$ and $\text{DE} \leq 18$	Moderate daily exceedances, but with the number of daily exceedances below the threshold of 18 days per year.
Moderate/ Yellow	$EC < 250$ and $\text{DE} > 18$	$EC < 150$ and $\text{DE} > 18$	Low annual excess, but with the number of daily exceedances exceeding the threshold of 18 days per year.
Moderate/ Orange	$251-750$ and $\text{DE} > 18$	$151-500$ and $\text{DE} > 18$	Moderate, frequent daily exceedances, with the number of daily exceedances exceeding the threshold of 18 days per year.
High/red	$751-1000$	$501-800$	Frequent daily exceedances and/or high daily concentration levels.
Very high/ Purple	>1000	>800	Severe pollution with numerous daily exceedances or extreme episodes of pollution.

3.4.2. Episode Index (EI)

EI takes into account the duration and intensity of pollution episodes, providing an indication of short-term exposure to PM pollution. An episode is defined as a sequence of at least two consecutive days on which the PM concentration exceeds the threshold value. The formula for calculating the *EI* indicator for a given pollution episode is as follows:

$$EI = \sum_{i=1}^n (C_i - P_{alert}) \quad (2)$$

where

C_i = concentration on day i ($\mu\text{g}/\text{m}^3$)

P_{alert} = alert threshold (Directive (UE) 2024/2881: $\geq 50 \mu\text{g}/\text{m}^3$ for $\text{PM}_{2.5}$ and $\geq 90 \mu\text{g}/\text{m}^3$ for PM_{10}).

n = number of consecutive days during the pollution episode where $C_i > P_{alert}$

The proposed threshold intervals for classifying the magnitude of pollution episodes (**Eroare! Fără sursă de referință.**) were established based on data from the scientific literature on the health effects of PM exposure. s-au bazat pe date din literatura de specialitate privind efectele expunerii la PM asupra sănătății (Lu et al., 2015; Pini et al., 2021).

Table 3. 2. Classification of pollution episode index (*EI*) .

Category	<i>EI</i> ($\mu\text{g}/\text{m}^3$) PM_{10}	<i>EI</i> ($\mu\text{g}/\text{m}^3$) $\text{PM}_{2.5}$	Interpretation criteria
High	1-25	1-25	2-3 consecutive days with concentrations close to $90 \mu\text{g}/\text{m}^3$ for PM_{10} , $50 \mu\text{g}/\text{m}^3$ for $\text{PM}_{2.5}$
Very high	26-100	26-100	3-4 consecutive days with concentrations around $100-115 \mu\text{g}/\text{m}^3$ for PM_{10} , $60-70 \mu\text{g}/\text{m}^3$ for $\text{PM}_{2.5}$
Extremely high	>101	>101	Long episodes and/or high peaks ($>115 \mu\text{g}/\text{m}^3$ for PM_{10} , $>75-90 \mu\text{g}/\text{m}^3$ for $\text{PM}_{2.5}$).

3.4.3. Toxic Load Index (*TLI*)

TLI intended to complement standard quantitative indicators, such as daily PM concentration, the number of daily exceedances, and annual mean concentration, as well as the other two indicators proposed in this study, *EC* and *EI*, by incorporating a qualitative dimension related to the potential toxicity of particulate matter.

The *TLI* accounts for both exceedances of the daily $\text{PM}_{2.5}$ limit value and the proportion of $\text{PM}_{2.5}$ within total PM_{10} , expressed as the $\text{PM}_{2.5}/\text{PM}_{10}$ ratio. On days when the $\text{PM}_{2.5}$ concentration does not exceed the applicable daily limit value, the *TLI* is assigned a value of zero, indicating that the predefined conditions for an increased toxic potential are not met.

Daily calculation of the *TLI* enables a rapid assessment of variations in the potential health risk associated with particulate matter pollution by accounting for daily changes in $\text{PM}_{2.5}$ and PM_{10} concentrations. These variations are influenced by both emission levels and meteorological conditions. The *TLI* is calculated using the following formula:

$$TLI_i = \left(\frac{PM_{2.5,i}}{PM_{10,i}} \right) \times (C_i - 25) \quad (3)$$

where

TLI_i = daily *TLI*

C_i = the $\text{PM}_{2.5}$ concentration on day i , when the LV is exceeded ($\mu\text{g}/\text{m}^3$)

$$\frac{PM_{2.5,i}}{PM_{10,i}} = \text{the ratio of concentrations on day } i$$

25 ($\mu\text{g}/\text{m}^3$) = daily limit value for $PM_{2.5}$

In the absence of recent local data on particulate matter chemical composition, the *TLI* uses the $PM_{2.5}/PM_{10}$ ratio as a proxy for the potential toxicity of particulate matter. Lower ratios (< 0.4) are indicative of a greater contribution from coarse mineral dust, whereas higher ratios (> 0.8) may indicate a greater contribution from fine particles associated with biomass combustion and diesel emissions and, consequently, a potentially higher toxic potential (Bodor and Bodor, 2022; Drăgoi et al., 2025a; Fan et al., 2021).

Although no directly equivalent indicator to *TLI* has been identified in the scientific literature, its methodological novelty lies in integrating, within a single quantitative indicator, two complementary dimensions of particulate matter pollution: the magnitude of $PM_{2.5}$ exceedances relative to the applicable daily limit value and the contribution of fine particles to total PM_{10} , expressed by the $PM_{2.5}/PM_{10}$ ratio. Thus, *TLI* extends conventional mass-based indicators by incorporating a proxy for the potential toxicological relevance of the particulate fraction.

The conceptual basis of *TLI* draws on established principles used in environmental and health-risk assessments, including the quantification of exceedances of concentration thresholds associated with adverse health effects (Abbey et al., 1989) and the use of the $PM_{2.5}/PM_{10}$ ratio to characterize the size distribution and potential source contributions of particulate matter (Drăgoi et al., 2025a; Li and Yang, 2020). The indicator therefore does not represent a direct measure of toxicity, rather, it provides a screening-level metric for characterizing variations in the potential health relevance of particulate pollution in the absence of detailed, time-resolved chemical composition data.

This approach is particularly relevant for monitoring networks where measurements of particulate chemical composition are limited. By combining concentration exceedance and particle-size fraction information, *TLI* provides an additional dimension for interpreting particulate matter pollution beyond conventional concentration-based indicators.

The proposed classification thresholds for daily *TLI* values (Table 3. 3) were established based on the magnitude of $PM_{2.5}$ concentration exceedances, weighted by the $PM_{2.5}/PM_{10}$ ratio.

Table 3. 3. Classification of daily toxic load index (*TLI*) for $PM_{2.5}$.

Category	<i>TLI</i> ($\mu\text{g}/\text{m}^3$)	Interpretation criteria
No risk	0	$PM_{2.5}$ below the daily limit value ($25 \mu\text{g}/\text{m}^3$), insignificant toxic load
Low	0.1-5	Slight exceedance or a lower $PM_{2.5}/PM_{10}$ ratio
Moderate	5.1-12	Significant exceedance and a high $PM_{2.5}/PM_{10}$ ratio
High	12.1-20	Severe pollution episodes with toxic composition
Very high	>20	Critical episode, major toxicological risk

3.5. Software and Analytical Tools

Data processing and statistical analyses were performed primarily using Python (version 3.12) and Microsoft Excel.

Chapter 4. Results and Discussion

4.1. Temporal Trend Analysis of Particulate Matter PM_{10} și $PM_{2.5}$

This subsection presents an analysis of the annual, monthly, weekly, diurnal, and seasonal variations in PM_{10} and $PM_{2.5}$ concentrations in the municipalities of Suceava, Iași, and Botoșani.

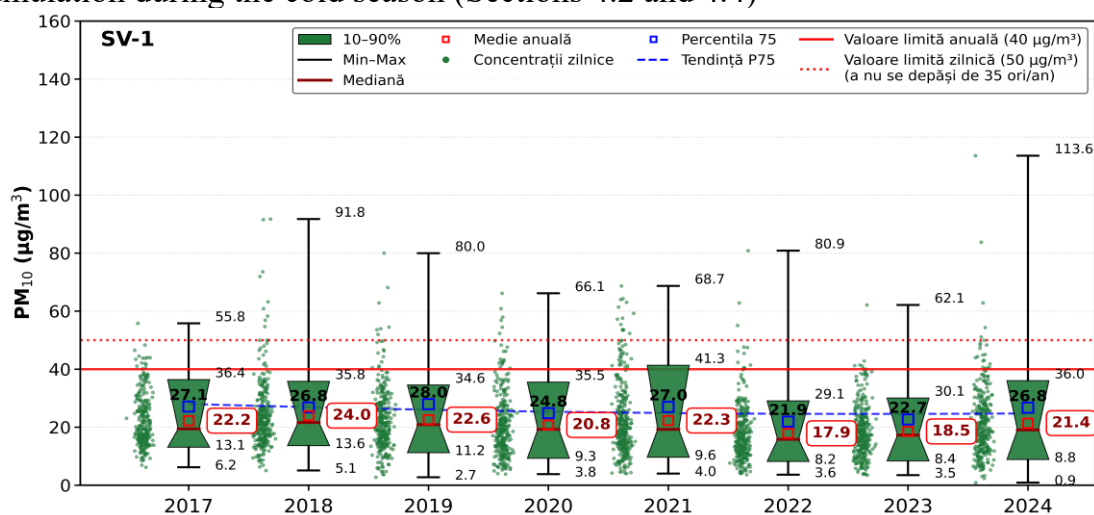
4.1.1. Annual Variability and Long-Term Trends in PM_{10} and $PM_{2.5}$ Concentrations (2017-2024).

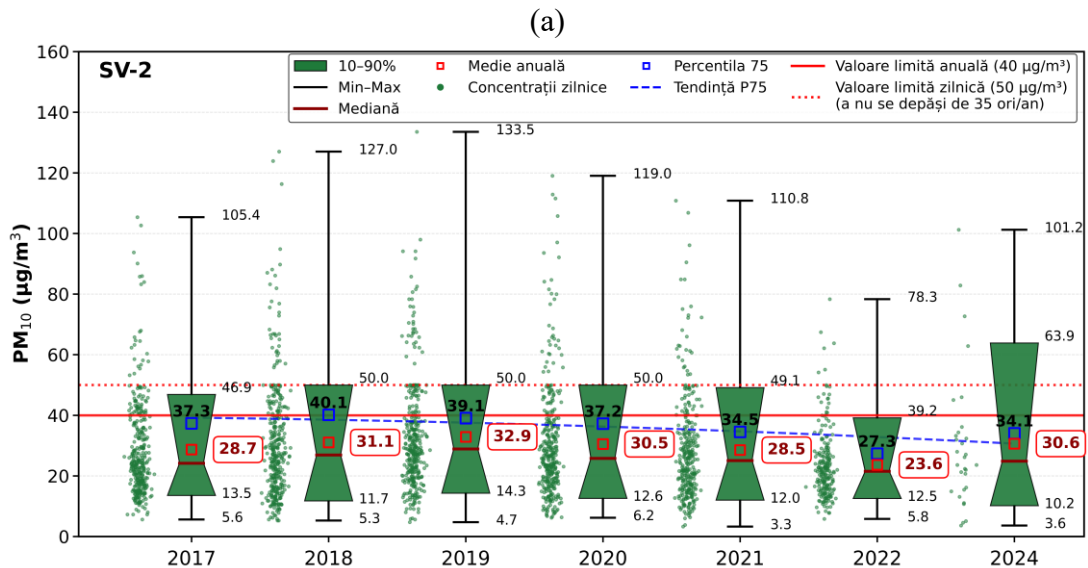
During the 2017-2024 period, the annual mean PM_{10} concentrations recorded at all analysed monitoring stations remained below the annual limit value of $40 \mu\text{g}/\text{m}^3$ established by Law No. 104/2011. The recorded values ranged from $17.88 \mu\text{g}/\text{m}^3$ at station SV-1 in 2022 to $37.29 \mu\text{g}/\text{m}^3$ at station IS-2 in 2018 (Fig. 4. 1).

An overall decreasing trend was observed at all monitoring stations, with the most pronounced reductions recorded at IS-2 and BT-1. At IS-2, the annual mean concentration decreased by approximately 19.4%, from $34.15 \mu\text{g}/\text{m}^3$ in 2017 to $27.53 \mu\text{g}/\text{m}^3$ in 2024. At BT-1, a marked decrease was observed between 2018 and 2023, followed by an increase in 2024. Despite the decrease in annual mean concentrations, episodes of elevated pollution were still recorded at IS-2, with the maximum daily concentration reaching $149.45 \mu\text{g}/\text{m}^3$ in December 2019.

The Mann-Kendall test identified statistically significant decreasing trends at stations BT-1 and IS-2, with Sen's slopes of approximately -1.8 and $-1.1 \mu\text{g}/\text{m}^3/\text{year}$, respectively. At stations SV-1 and SV-2, the observed decreases were not statistically significant (Drăgoi et al., 2026a).

The daily limit value of $50 \mu\text{g}/\text{m}^3$, which must not be exceeded on more than 35 days per calendar year under Law No. 104/2011, was reached at station SV-2 in 2018 and 2019 and exceeded at station IS-2 during 2017-2019, with the threshold reached again in 2020. At stations BT-1 and SV-1, the maximum permitted number of days with concentrations above the daily limit value was never exceeded. The uneven distribution of exceedances reflects the combined influence of local emission sources, particularly road traffic and residential heating, and meteorological conditions that favour pollutant accumulation during the cold season (Sections 4.2 and 4.4)





SV-2: In 2022 and 2024, the annual mean concentrations are considered invalid because the available data were insufficient to meet the data quality objectives and aggregation criteria specified under Law No. 104/2011. The monitoring station was temporarily out of operation from 11.08.2022 to 3.10.2024.

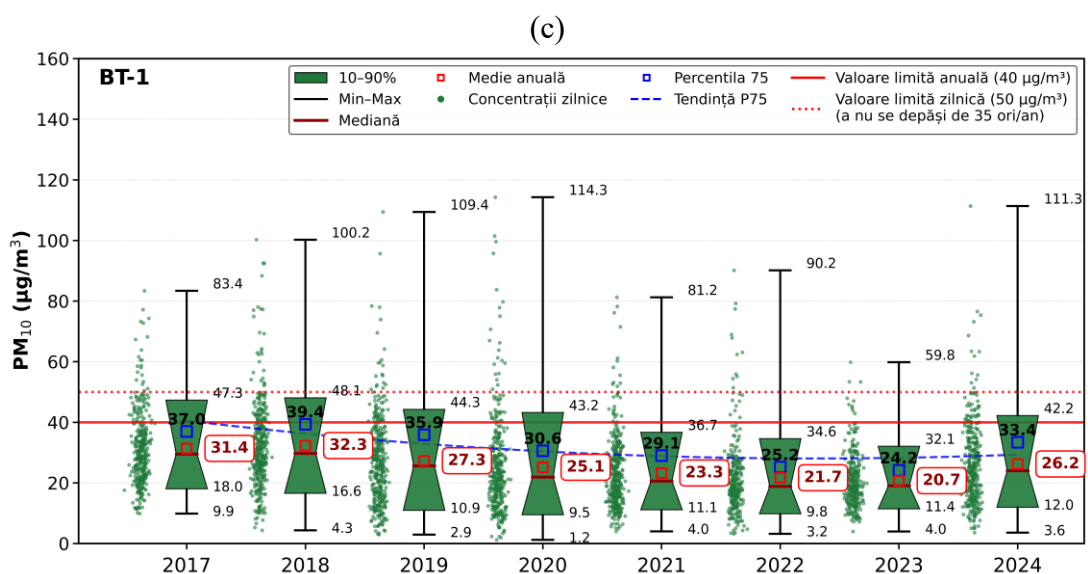
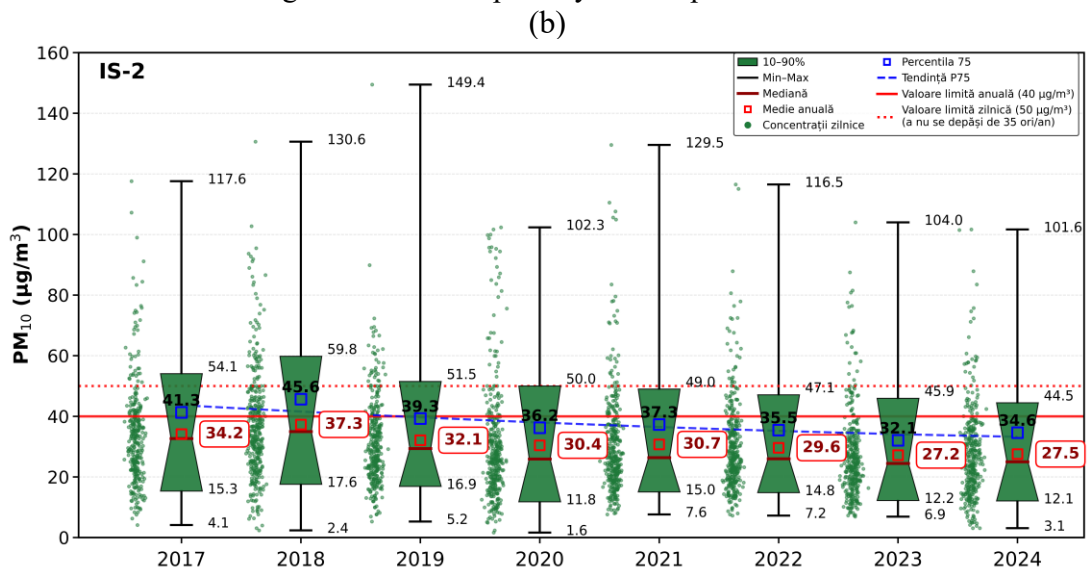


Fig. 4. 1. Evolution of statistical parameters of PM₁₀ concentrations at stations SV-1 (a), SV-2 (b), IS-2 (c), and BT-1 (d) during 2017-2024

The annual mean PM_{2.5} concentrations recorded at the urban background stations SV-1, IS-2, and BT-1 remained below the annual limit value of 25 µg/m³ during 2017-2024, with the exception of 2017 and 2018 at IS-2. PM_{2.5} is not monitored at station SV-2. The annual mean concentrations ranged from 11.23 µg/m³ at SV-1 in 2023 to 28.7 µg/m³ at IS-2 in 2017.

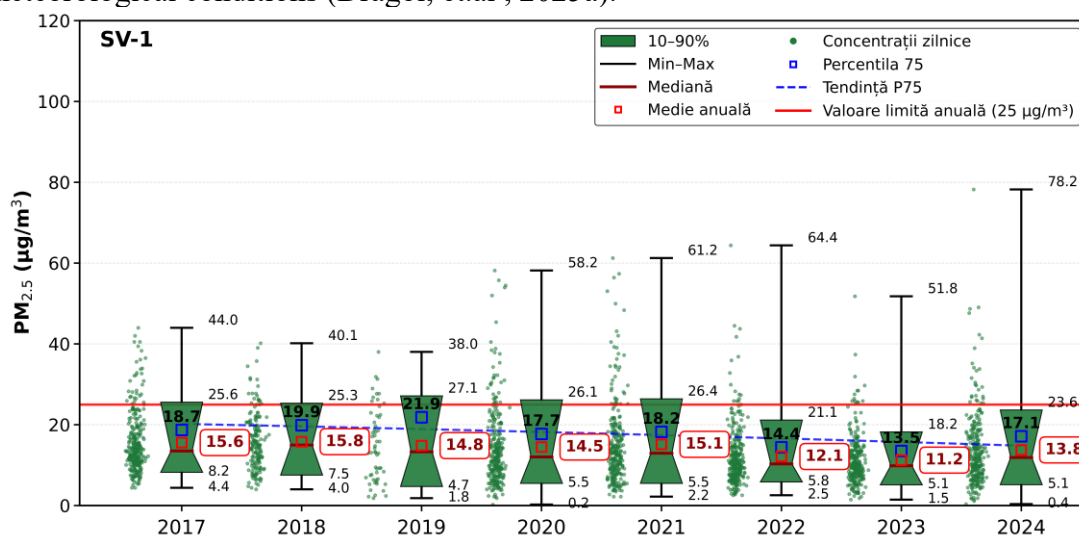
The median and 75th-percentile concentrations showed a gradual decrease up to 2023, followed by a slight increase in 2024. Nevertheless, these values remained below those recorded in 2017, suggesting an overall improvement in air quality. The highest daily concentration, 111.6 µg/m³, was recorded at IS-2 in 2018.

Throughout the study period, daily concentrations between 40 and 60 µg/m³ were frequently recorded, while concentrations exceeding 70 µg/m³ were observed on several occasions, indicating the persistence of episodes of elevated particulate matter pollution (Fig. 4. 2).

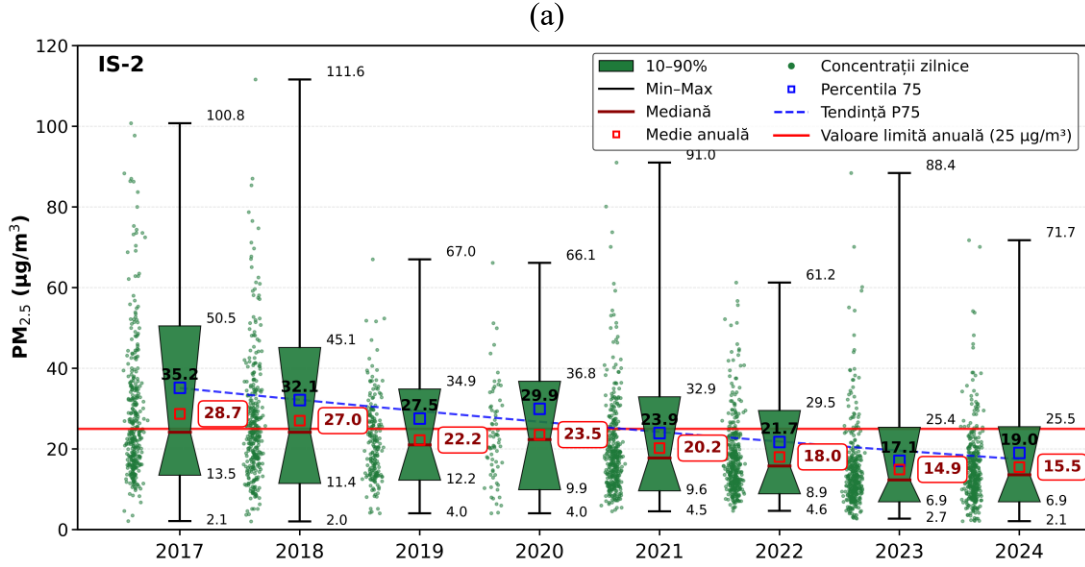
For PM_{2.5}, the Mann-Kendall test identified a statistically significant decreasing trend only at station IS-2 during 2017-2024, with Sen's slope indicating a reduction of approximately 2.9 µg/m³ per year over the study period. Although this decrease may appear relatively modest, recent studies indicate that even reductions of approximately 1 µg/m³ in PM_{2.5} concentrations can be associated with measurable reductions in population mortality and morbidity (Oudin et al., 2024).

At SV-1, the short-term analysis for 2017-2024 did not reveal a statistically significant trend; however, extending the analysis to 2009-2024 identified a statistically significant decreasing trend. This finding suggests a gradual improvement in air quality that may be difficult to detect over shorter periods because of interannual variability. At BT-1, the absence of a statistically significant trend persisted in the long-term analysis (2009-2024), indicating relatively stable PM_{2.5} concentrations over the longer observation period.

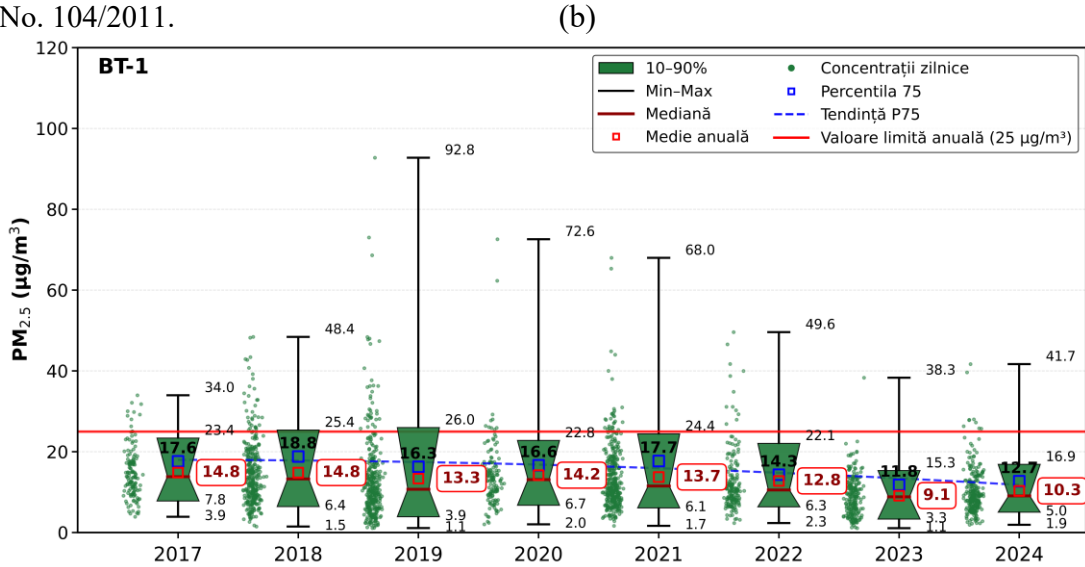
At IS-2, the decreasing trend identified for 2017-2024 was not observed over the 2009-2024 period, suggesting that the recent reduction in PM_{2.5} concentrations represents a relatively recent change rather than a persistent long-term linear trend. This change may be associated with the implementation of measures included in air quality plans, as well as variations in meteorological conditions (Drăgoi, et.al , 2025a).



SV-1: In 2018 and 2019, the annual mean concentrations are considered invalid because the available data were insufficient to meet the data quality objectives and data aggregation criteria specified under Law No. 104/2011.



IS-2: In 2019 and 2020, the annual mean concentrations are considered invalid because the available data were insufficient to meet the data quality objectives and data aggregation criteria specified under Law No. 104/2011.



BT-1: For 2017, 2020, and 2022-2024, the annual mean concentrations are considered invalid because the available data were insufficient to meet the data quality objectives and data aggregation criteria specified under Law No. 104/2011.

Fig. 4. 2. Evolution of statistical parameters of $PM_{2.5}$ concentrations at stations SV-1 (a), IS-2 (b), and BT-1 (c) during 2017-2024

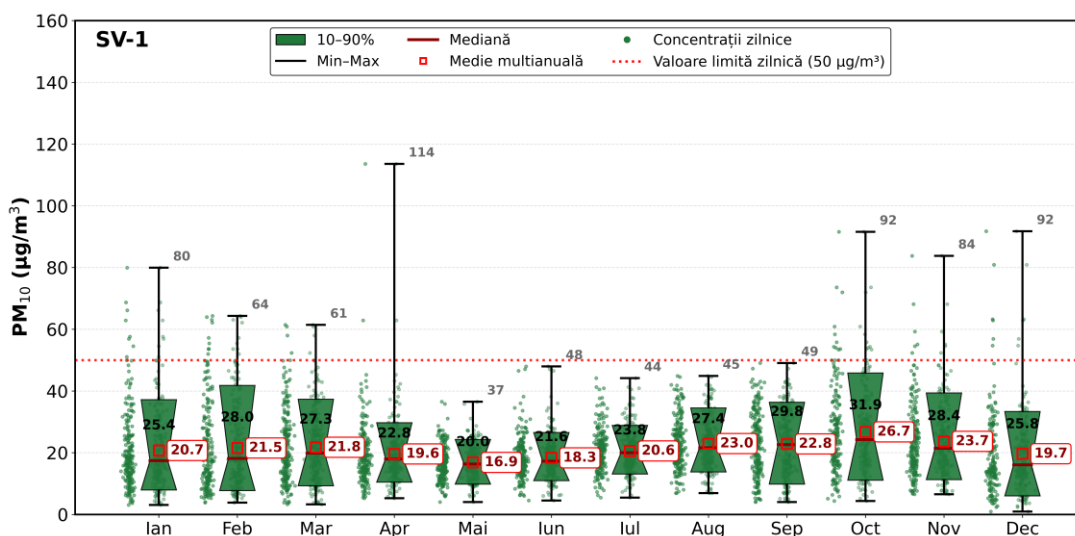
These trends can be attributed primarily to the implementation of emission-control legislation in the energy, industrial, and transport sectors, including the Industrial Emissions Directive, which has contributed to substantial reductions in emissions of PM_{10} , $PM_{2.5}$, and their precursor pollutants (SO_2 , NO_x , and NMVOC). Several European assessments indicate that these policies, together with the modernization of combustion installations and the transition from solid fuels to less-polluting technologies, have contributed to significant reductions in PM_{10} and $PM_{2.5}$ concentrations, with estimated annual declines of approximately 1.5-2.5% over the past two decades (European Environment Agency, 2023, 2025).

4.1.2. Monthly Variability and Long-Term Trends in PM₁₀ and PM_{2.5} Concentrations (2017-2024).

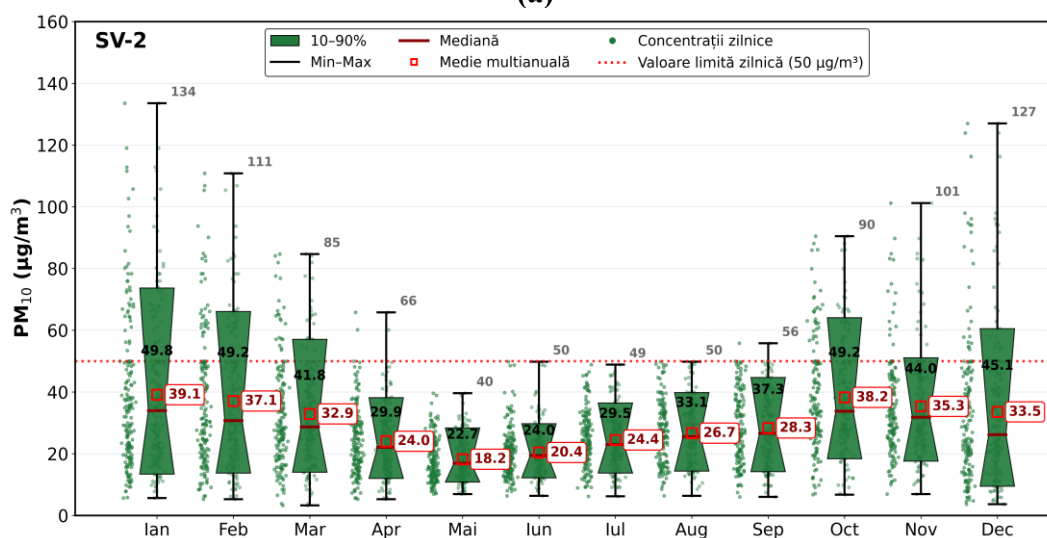
The analysis of daily PM₁₀ concentrations aggregated on a monthly basis revealed a pronounced seasonal pattern, with higher mean and median concentrations during the colder months (October-March) and lower levels during the warm season. Differences between monitoring stations were also evident: IS-2 and SV-2 generally recorded higher mean and median concentrations throughout the year, whereas SV-1 consistently exhibited some of the lowest monthly concentrations.

During the winter months, both daily variability and maximum concentrations frequently exceeded the daily limit value of 50 µg/m³, particularly at IS-2 and SV-2, where the highest peaks reached approximately 130-150 µg/m³. In contrast, SV-1 was characterized by consistently lower concentrations.

The marked contrast between the cold and warm seasons suggests a substantial contribution from residential heating using solid fuels, combined with meteorological conditions that favour pollutant accumulation, such as thermal inversions, weak winds, and limited atmospheric dispersion. These factors are likely to contribute to the occurrence and persistence of PM₁₀ pollution episodes during the cold season. (Fig. 4. 3).



(a)



SV-2: The monitoring station was temporarily out of operation from 11 August 2022 to 3 October 2024. Data from 2022-2024 were excluded from the analysis because the available data were either missing or insufficient to meet the data quality objectives and data aggregation criteria specified under Law No. 104/2011 on ambient air quality.

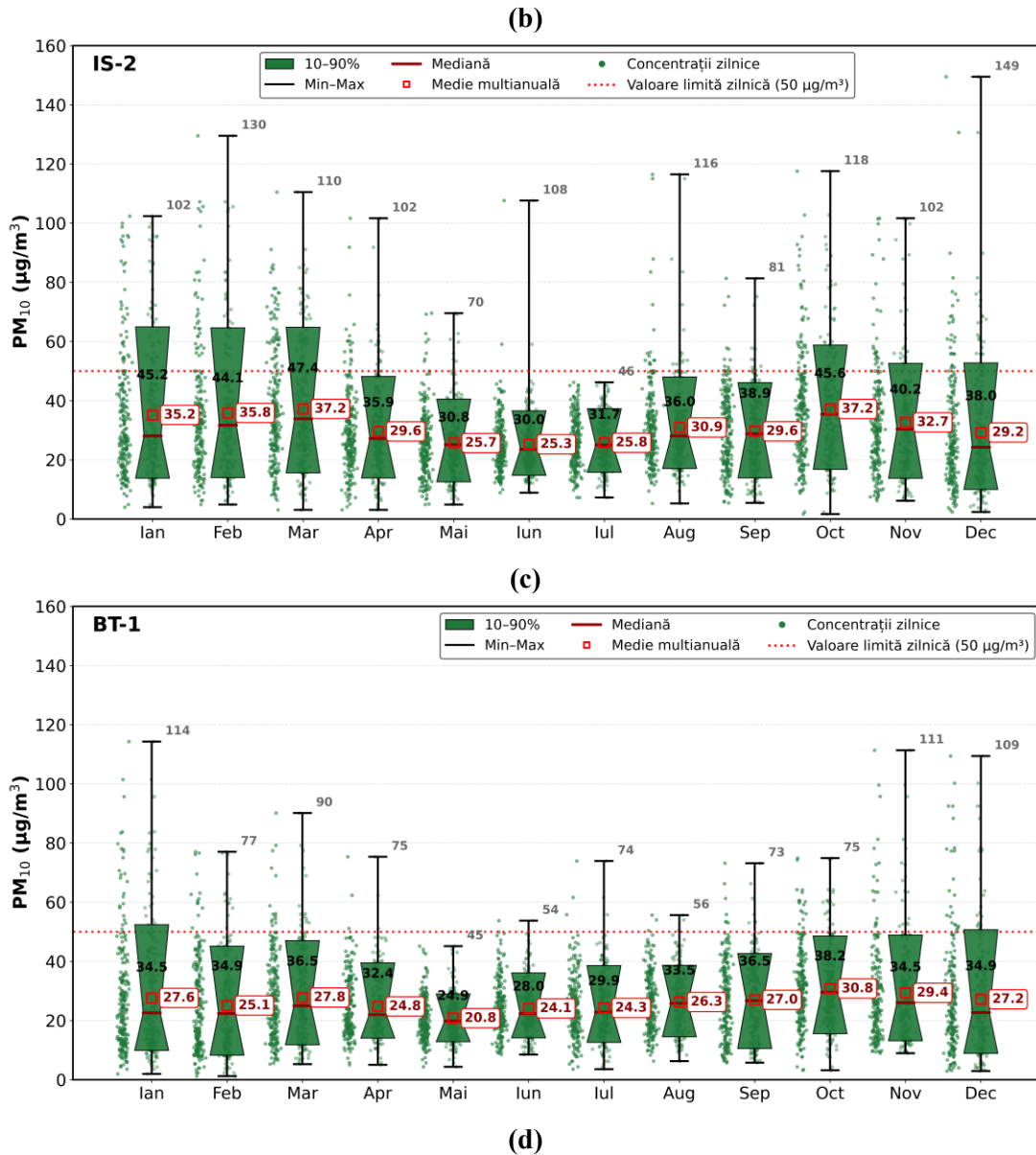


Fig. 4. 3. Monthly variations in daily PM_{10} concentrations at stations SV-1 (a), SV-2 (b), IS-2 (c), and BT-1 (d) during 2017-2024

During 2017-2024, statistically significant decreasing trends in monthly PM_{10} concentrations at station BT-1 were identified only in April and August, with Sen's slopes of -2.3 and -3.1 $\mu\text{g}/\text{m}^3/\text{year}$, respectively, indicating substantial reductions in PM_{10} concentrations during these months over the study period.

At station IS-2, a statistically significant decreasing trend was identified only in June between 2017 and 2024, with a Sen's slope of -1.3 $\mu\text{g}/\text{m}^3/\text{year}$. The remaining months exhibited moderate negative slopes, but these trends were not statistically significant.

At stations SV-1 and SV-2, none of the months showed statistically significant trends, suggesting that monthly PM_{10} variability during 2017-2024 was driven primarily by interannual fluctuations rather than by systematic long-term changes.

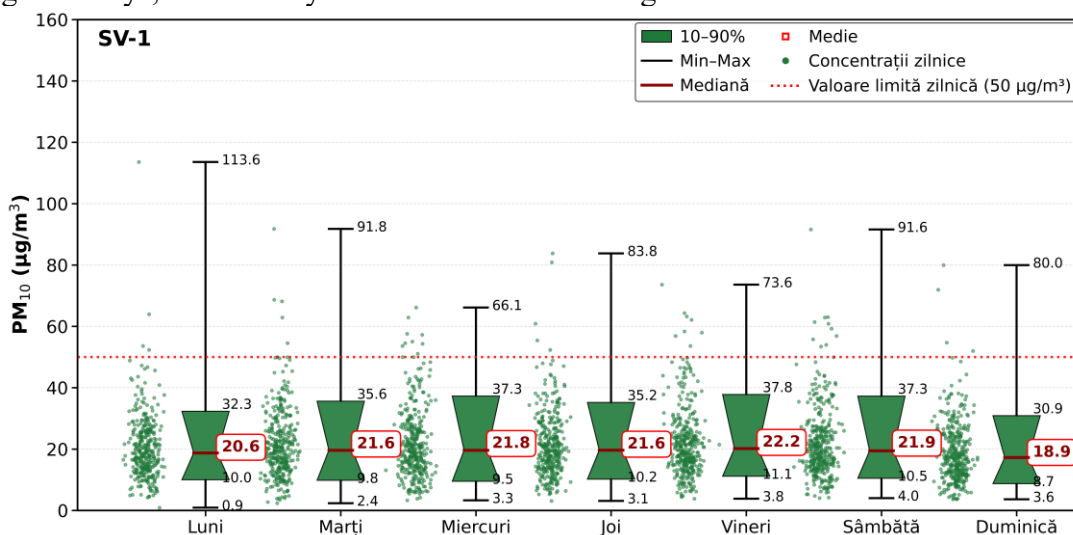
The analysis of monthly PM_{2.5} concentrations revealed a statistically significant decreasing trend at station IS-2 during 2017-2024 in February, March, April, and June, with Sen's slopes ranging from -2.25 to -5.09 µg/m³/year. No statistically significant trends were identified in the remaining months. Over the extended 2009-2024 period, monthly PM_{2.5} concentrations were generally stable, with a statistically significant decrease identified only in April, potentially reflecting seasonal effects or changes in emission sources.

At station SV-1, statistically significant decreasing trends in PM_{2.5} concentrations during 2017-2024 were identified only in March and April, with Sen's slopes of approximately -2.5 and -1.2 µg/m³/year, respectively. These results indicate a reduction in PM_{2.5} concentrations during the spring months over the study period. When the analysis was extended to 2009-2024, the number of months showing statistically significant decreases increased. Significant downward trends ($p < 0.05$) were identified in January, March, April, and December, with Sen's slopes ranging from -1.4 to -2.2 µg/m³/year, indicating reductions during the cold season and spring. No statistically significant trends were identified during the summer and autumn months.

At station BT-1, monthly PM_{2.5} concentrations over the 2009-2024 period showed no statistically significant trends in most months. The only exception was June, when a slight but statistically significant increasing trend was observed, with a Sen's slope of 0.52 µg/m³/year. In the remaining months, Sen's slopes were small, ranging from -1.0 to 0.3 µg/m³/year, indicating that the observed variability was dominated by interannual fluctuations rather than systematic long-term changes.

4.1.3. Weekly Variability in Daily PM₁₀ and PM_{2.5} Concentrations (2017-2024)

During 2017-2024, the highest mean PM₁₀ concentrations were recorded on Fridays at all four monitoring stations analysed, reaching 27.8 µg/m³ at BT-1, 34.2 µg/m³ at IS-2, 32.8 µg/m³ at SV-2, and 22.2 µg/m³ at SV-1. The lowest concentrations were generally recorded on Sundays, with a minimum mean concentration of 26.1 µg/m³ at IS-2. Daily maximum concentrations frequently exceeded 100 µg/m³, particularly on Thursdays and Fridays, indicating that episodes of elevated PM₁₀ pollution tended to occur more frequently during the second half of the week (Fig. 4. 4). This pattern may be associated with increased traffic intensity and economic activity during weekdays, followed by reduced emissions during the weekend.



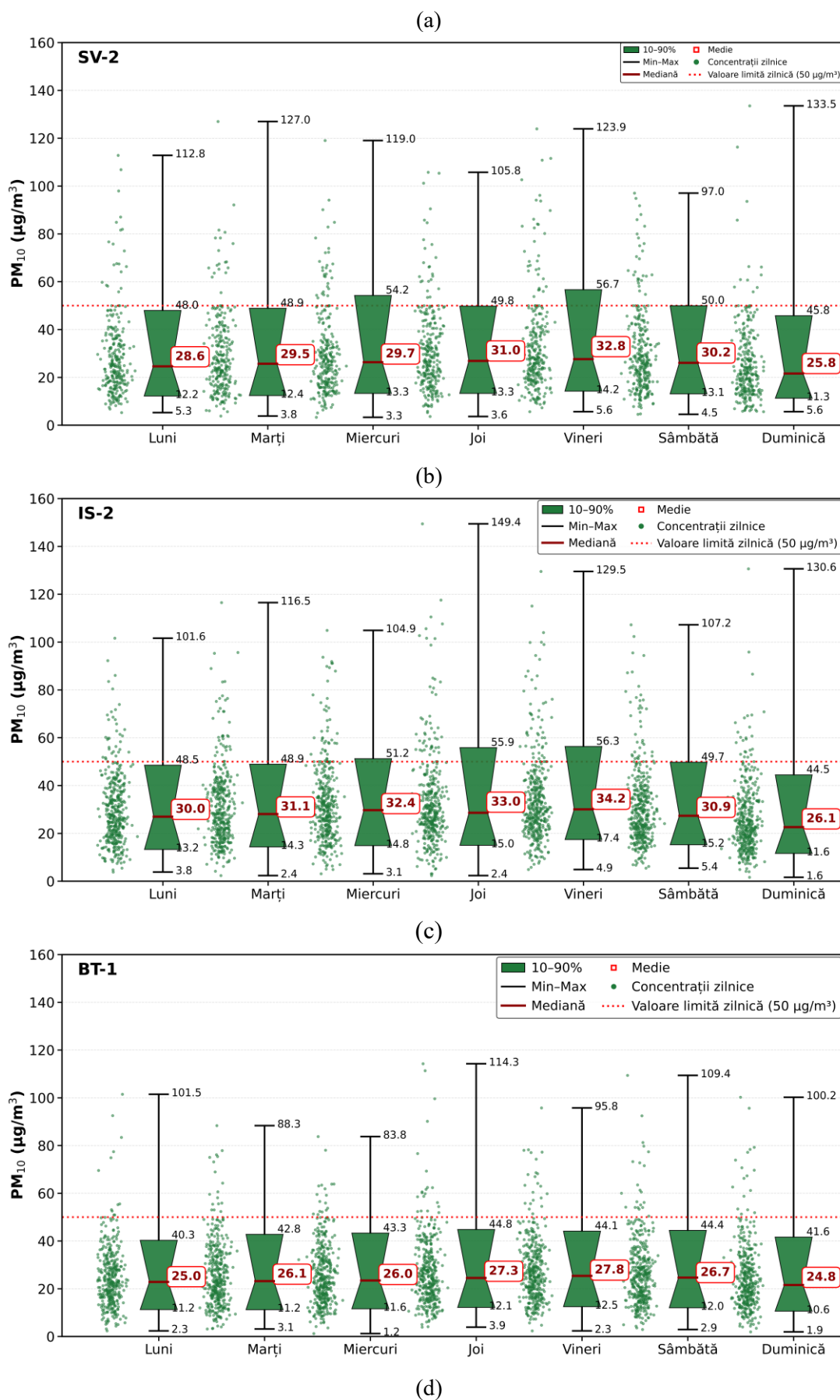


Fig. 4. 4. Multiannual distribution of daily PM₁₀ concentrations by day of the week at stations SV-1 (a), SV-2 (b), IS-2 (c), and BT-1 (d) during 2017-2024

PM_{2.5} concentrations exhibited a weekly variation pattern similar to that observed for PM₁₀, with higher values on weekdays, particularly during the second half of the week, and lower concentrations during the weekend.

4.1.4. Diurnal and Seasonal Variations in Hourly PM₁₀ Concentrations

The analysis of hourly PM₁₀ concentrations during 2017-2024 revealed a bimodal diurnal pattern, with morning peaks around 08:00-09:00 and evening peaks between 20:00 and 23:00. The peaks were more pronounced at stations IS-2 and SV-2, suggesting a stronger influence of local emission sources, including road traffic and residential heating and, at SV-2, industrial emissions.

The highest concentrations were observed during the cold season, particularly in the evening, under stable atmospheric conditions and a reduced planetary boundary layer height, which favour the accumulation of particulate matter. The urban background stations SV-1 and BT-1 recorded lower concentrations and less pronounced seasonal variability. At all stations, the lowest concentrations were generally observed during summer, particularly between 11:00 and 16:00, when the development of the planetary boundary layer enhances the atmospheric dispersion of pollutants (Fig. 4. 5).

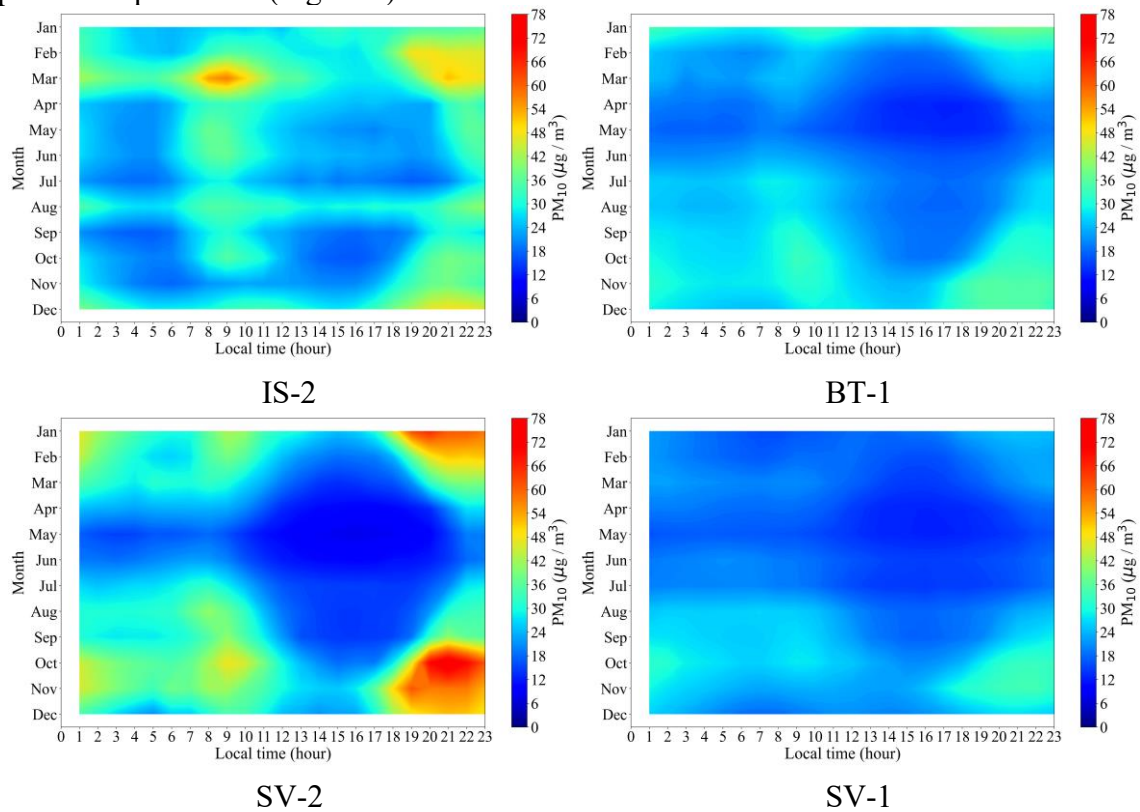


Fig. 4. 5. Monthly mean hourly PM₁₀ concentrations in the study area, 2017-2024

Note: For station IS-2, hourly data were available starting in February 2022.

The analysis of hourly PM₁₀ concentrations during 2017-2024 revealed a pronounced diurnal pattern and differences between weekdays and weekends. Between 01:00 and 06:00, concentrations were slightly higher during the weekend, whereas after 06:00 they generally remained below the levels recorded on weekdays. At all monitoring stations, the lowest daytime concentrations were observed between 11:00 and 16:00, while a pronounced evening peak

occurred between 21:00 and 23:00, particularly at SV-2, where mean weekday concentrations exceeded $40 \mu\text{g}/\text{m}^3$ (Fig. 4. 6).

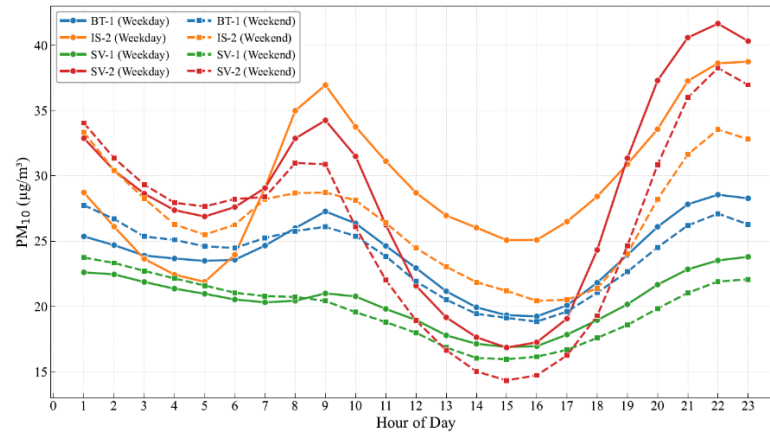


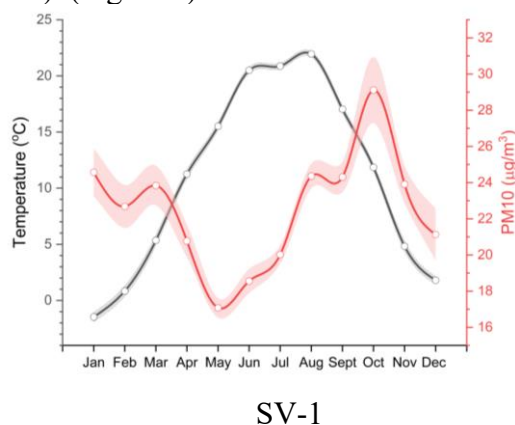
Fig. 4. 6. Diurnal variability of hourly PM_{10} concentrations on weekdays and weekends, 2017-2024

4.1.5. Influence of Air Temperature on PM_{10} Concentrations

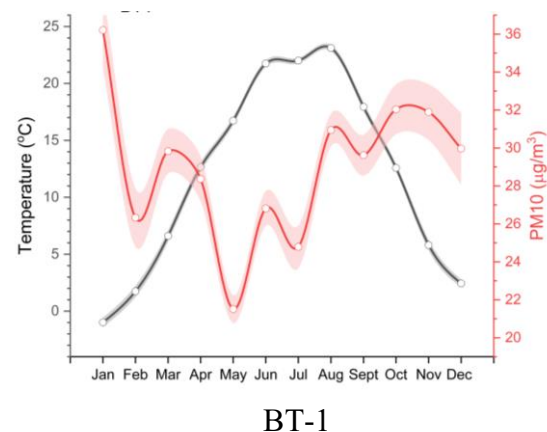
The finding that, across the three cities analysed, the relationship between air temperature and PM_{10} follows seasonal patterns, with negative correlations during winter and positive correlations during summer, suggests that temperature acts primarily as an indirect indicator of a combination of physical processes and human activities rather than as a direct determinant of particulate matter concentrations (Pražnikar, 2024).

During the cold season, low temperatures are associated with increased residential heating and more frequent thermal inversions, which favour the accumulation of PM_{10} near the ground and may explain the negative temperature- PM_{10} relationship observed in continental European cities (González Ortiz et al., 2020). At higher temperatures, improved atmospheric mixing and reduced heating-related emissions generally contribute to lower PM_{10} concentrations, although resuspension and natural sources may modify this seasonal pattern (Cholakian et al., 2019).

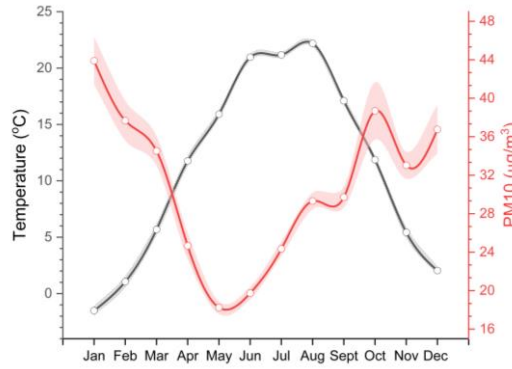
During 2017-2024, the relationship between air temperature and PM_{10} concentrations showed a nonlinear inverse pattern, with higher concentrations occurring at lower temperatures and decreasing concentrations as temperatures increased towards the warm season (Dragoi et al., 2023) (Fig. 4. 7).



SV-1



BT-1



SV-2

Fig. 4. 7. Variability of PM₁₀ concentrations in relation to air temperature at stations SV-1, BT-1, and SV-2 during 2017-2020

Analysis of the 2017-2020 data showed that the highest daily PM₁₀ concentrations occurred during cold-season days characterized by thermal inversions, highlighting the role of this meteorological phenomenon in the accumulation of atmospheric pollutants (Girotti et al., 2025).

4.1.6. Relationships Between PM₁₀, PM_{2.5}, and Gaseous Pollutants (NO₂, SO₂, CO, and Benzene)

4.1.6.1. Statistical Correlations Between Pollutants

During the cold season (November-March), PM₁₀ showed moderate correlations with NO₂ ($R = 0.55$ - 0.66) and CO ($R = 0.58$ - 0.69), with the highest coefficients recorded at station SV-2 ($R = 0.66$ for NO₂ and $R = 0.69$ for CO). Benzene also showed moderate to strong correlations with PM₁₀ ($R = 0.41$ - 0.66), whereas correlations with SO₂ remained weak in both seasons ($R \leq 0.33$). PM_{2.5} showed moderate correlations with NO₂ ($R = 0.54$ - 0.63) and benzene ($R = 0.54$ - 0.65), with the highest correlation with NO₂ observed at SV-1 ($R = 0.63$). These results indicate that elevated particulate matter concentrations frequently coincided with increases in combustion-related gaseous pollutants. This pattern is consistent with findings from European urban studies linking particulate matter concentrations to residential heating and road traffic, particularly under stable atmospheric conditions and thermal inversions.

During the warm season (April-October), correlations between PM₁₀ and NO₂, as well as between PM₁₀ and CO, were generally weak ($R = 0.10$ - 0.39), only occasionally reaching moderate strength. This weaker association suggests that gaseous pollutants and PM₁₀ exhibited less closely coupled temporal variability during the warm season. The observed pattern may be related to the reduced contribution of residential heating and more favourable atmospheric dispersion conditions, which can weaken the association between local combustion-related emissions and particulate matter concentrations.

The strong correlations between PM₁₀ and PM_{2.5} ($R = 0.71$ - 0.93) indicate that the two particulate fractions are substantially influenced by common emission sources and shared atmospheric processes. The strength of these relationships may also reflect the influence of stable

meteorological conditions, which limit atmospheric dispersion and favour the accumulation of particulate matter in the lower atmosphere (Zhou et al., 2016).

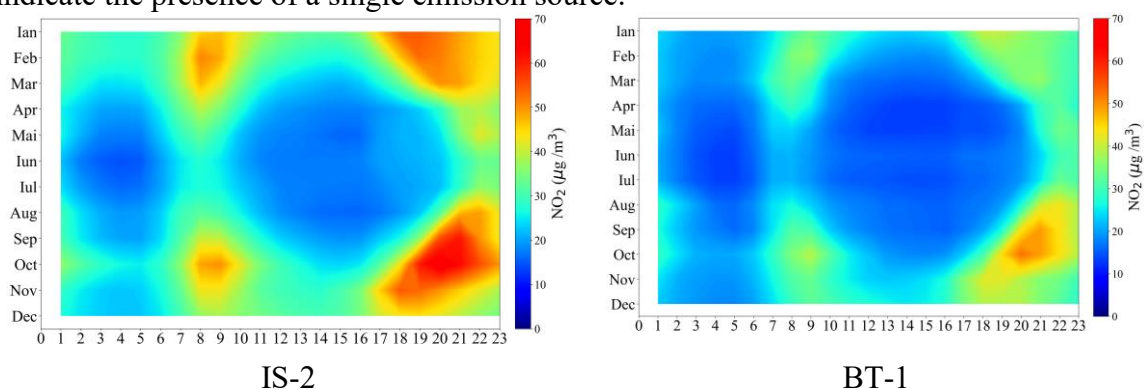
The very weak correlations between particulate matter and SO₂ observed in both seasons may be associated with the relatively low contribution of sulfur-containing fuels to local emissions, including the use of natural gas and biomass, although the correlation analysis alone does not allow source attribution. The low SO₂ concentrations recorded in the study area further support this interpretation. These findings are consistent with results reported in regional studies from Romania and other European urban and residential environments, where similar relationships between particulate matter and combustion-related pollutants have been observed (Bodor, 2020; Kendrick et al., 2015).

4.1.6.2. Diurnal Variability of PM₁₀ and NO₂

The correlation analysis between PM₁₀ and NO₂ revealed similar seasonal and diurnal patterns, particularly during the cold season, when simultaneous increases in the two pollutants were more pronounced in the morning and evening. At IS-2, NO₂ concentrations were the highest, with morning peaks around 08:00-09:00 and evening peaks between 20:00 and 22:00, patterns consistent with increased traffic emissions. PM₁₀ concentrations partially coincided with these NO₂ peaks. However, the elevated PM₁₀ levels observed during nighttime and winter also suggest contributions from residential heating and other particulate matter sources.

At BT-1, both NO₂ and PM₁₀ concentrations were generally lower and more uniform, with slight increases during the cold season, particularly in the evening. This pattern may reflect the combined influence of traffic and residential heating. SV-2 recorded the highest PM₁₀ concentrations, reaching or exceeding 60-70 µg/m³ during winter evenings and nights, particularly between 20:00 and 23:00. At the same station, CO concentrations frequently exceeded 0.70-0.80 mg/m³, further supporting the potential contribution of combustion-related sources during the cold season. At SV-1, concentrations were the lowest and diurnal variability was limited, consistent with the characteristics of an urban background station (Fig. 4. 5 și Fig. 4. 8).

The interpretation of the correlations should be approached with caution, as the observed associations between PM₁₀ and NO₂ may reflect common emission influences or shared meteorological conditions, but do not, by themselves, demonstrate a direct causal relationship or indicate the presence of a single emission source.



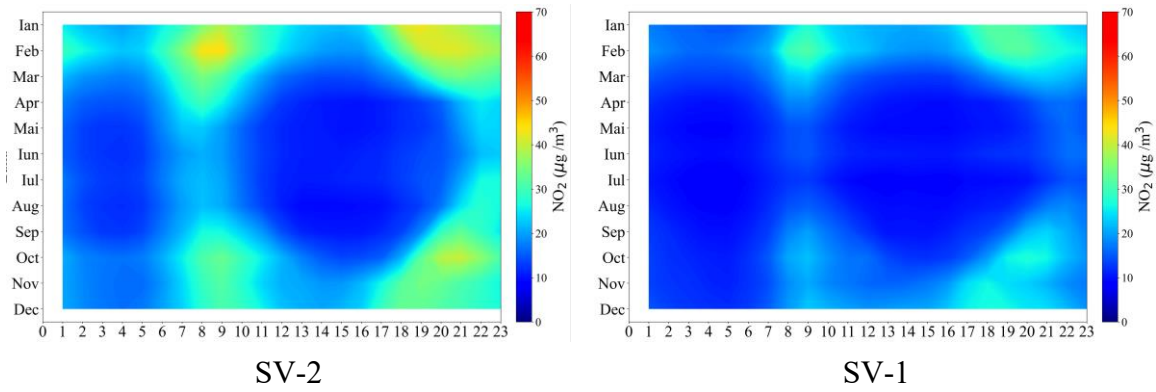


Fig. 4. 8. Mean hourly NO_2 concentrations in the study area, 2017-2024

Across all monitoring stations, NO_2 concentrations were consistently lower during weekends than on weekdays, particularly between 06:00 and 10:00, when the morning traffic-related peak was substantially reduced.

4.1.6.3. Diurnal Variability of PM_{10} and CO

The analysis revealed more pronounced seasonal and diurnal variability during the cold season for both CO and PM_{10} . At SV-1, CO concentrations were the lowest, generally remaining below 0.20 mg/m^3 , with maxima of $0.30\text{-}0.40 \text{ mg/m}^3$ during November-February, particularly in the evening and at night, while PM_{10} concentrations were moderate and exhibited limited diurnal variability. At SV-2, the highest concentrations were recorded, with CO frequently exceeding $0.70\text{-}0.80 \text{ mg/m}^3$ between 18:00 and 23:00 during the cold season, while PM_{10} reached or exceeded $60\text{-}70 \text{ }\mu\text{g/m}^3$, with maxima around 20:00-23:00. At BT-1, CO concentrations were intermediate, with maximum values of $0.60\text{-}0.90 \text{ mg/m}^3$ during the cold season, particularly in the evening and at night, whereas PM_{10} showed a more uniform hourly pattern. Across all stations, the diurnal CO profile exhibited two main peaks, in the morning around 08:00-09:00 and in the evening between approximately 20:00 and 22:00, with the latter being more pronounced. PM_{10} generally followed a similar pattern, with higher concentrations in the evening and at night and lower values around midday.

Overall, SV-2 recorded the highest concentrations, BT-1 intermediate levels, and SV-1 the lowest concentrations. The observed differences are consistent with varying contributions from traffic and residential heating, as well as differences in atmospheric dispersion conditions and station elevation.

Differences between weekdays and weekends were evident for both pollutants but appeared more pronounced for PM_{10} . This pattern suggests an important contribution from residential heating and other sources not directly associated with road traffic. It may also reflect differences in the atmospheric persistence of the two pollutants, as particulate matter can remain suspended for longer periods than CO under certain atmospheric conditions, favouring its accumulation during periods of atmospheric stability (Gualtieri et al., 2015).

4.1.6.4. Daily Regression Analysis of PM₁₀ and Gaseous Pollutants (NO₂, CO, SO₂, and Benzene) and Meteorological Parameters

Regression models including both gaseous pollutants and meteorological variables explained between 41% and 65% of the daily variability in PM₁₀ concentrations ($R^2 = 0.41-0.65$), depending on the monitoring station and season. At SV-1, the models explained 44% of the variability during winter and 41% during summer; at BT-1, 48% during winter and 54% during summer; and at SV-2, 44% during winter and 63% during summer. The highest explanatory power was observed at IS-2, where the models accounted for 57% of the variability in winter and approximately 65% in summer.

Among the gaseous pollutants considered, NO₂ and CO showed statistically significant associations with PM₁₀ at several monitoring stations, while benzene exhibited the strongest associations, particularly at BT-1 and IS-2. Meteorological variables, particularly air temperature, atmospheric pressure, and relative humidity, also contributed to the variability in PM₁₀ concentrations, although their effects differed by station and season.

Overall, the results indicate that PM₁₀ variability is influenced by the combined effects of emission-related and meteorological factors. However, the regression coefficients represent statistical associations and should not be interpreted as direct evidence of causality or as definitive attribution of PM₁₀ concentrations to specific emission sources.

4.2 Spatial and Temporal Analysis of the PM_{2.5}/PM₁₀ Ratio

This study analysed the characteristics of PM_{2.5} and PM₁₀ pollution in the municipalities of Suceava, Iași, and Botoșani during 2019-2023, with particular emphasis on the PM_{2.5}/PM₁₀ ratio. The analysis focused on the temporal and seasonal variability of particulate matter concentrations, as well as on the relationship between the PM_{2.5}/PM₁₀ ratio, air temperature, and potential emission sources.

4.2.1. Annual Variability of the PM_{2.5}/PM₁₀ Ratio

The analysis of the annual variability of the PM_{2.5}/PM₁₀ ratio during 2019-2023 at the three urban background monitoring stations (SV-1, IS-2, and BT-1) revealed both interannual variability and spatial differences. Throughout the study period, PM_{2.5}/PM₁₀ values ranged from 0.60 to 0.71 at SV-1, from 0.54 to 0.72 at IS-2, and from 0.48 to 0.61 at BT-1.

These values indicate that the characteristics of particulate matter pollution in north-eastern Romania are broadly comparable with those reported at regional and international levels. The annual mean PM_{2.5}/PM₁₀ ratios recorded at SV-1 (0.66) and IS-2 (0.62) were close to the reference value of 0.65 proposed for developed regions (Ostro, 2004). The results suggest a substantial contribution of fine particles associated with combustion-related processes in urban environments, particularly in Suceava and Iași. The lower values recorded at BT-1 suggest a relatively greater contribution from the coarse fraction and potentially associated local sources, including particle resuspension. In addition, the interannual variability of the ratio was lower at SV-1 and IS-2 and more pronounced at BT-1, indicating greater variability in local emission and atmospheric conditions at the latter station.

4.2.2. Monthly Variability of the PM_{2.5}/PM₁₀ Ratio

The analysis of the monthly distribution of the PM_{2.5}/PM₁₀ ratio revealed higher values during the colder months and lower values during the warm season, suggesting a shift from a regime more strongly influenced by combustion-related sources, such as residential heating and traffic, towards conditions characterized by a greater contribution from coarse particles and resuspension processes. The seasonal amplitude was most pronounced at BT-1, where the ratio ranged from approximately 0.75 in December-January to 0.40 in April-June, whereas the variations observed at SV-1 and IS-2 were more moderate.

4.2.3. Comparison of the Monthly Variability of the PM_{2.5}/PM₁₀ Ratio with PM₁₀, PM_{2.5}, CO, NO₂, and SO₂ Concentrations

The comparative analysis revealed a consistent seasonal pattern between the PM_{2.5}/PM₁₀ ratio and pollutant concentrations, with higher ratio values during the cold season coinciding with increased NO₂ and CO concentrations, whereas the lower ratios observed during the warm season corresponded to a relatively greater contribution of coarse particles.

4.2.4. Monthly Variability of PM_{2.5} and PM_{2.5-10} Concentrations

To distinguish between the contributions of the fine and coarse particulate fractions, PM_{2.5-10} was calculated as the difference between PM₁₀ and PM_{2.5} concentrations (Fig. 4. 9). The analysis of monthly variability revealed higher PM_{2.5} concentrations during the cold season, particularly at IS-2, whereas the PM_{2.5-10} fraction increased during the warm season, with higher values recorded at BT-1. This pattern suggests a greater contribution from coarse-particle sources, such as resuspended dust and agricultural activities, during the warm season.

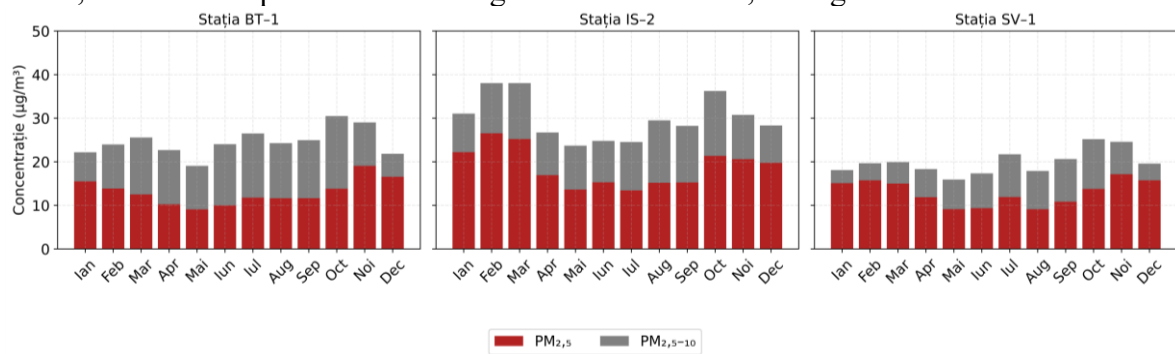


Fig. 4. 9. Monthly mean concentrations of PM_{2.5} and PM_{2.5-10} at stations BT-1, IS-2, and SV-1 during 2019-2023

4.2.5. Seasonal Variability of the PM_{2.5}/PM₁₀ Ratio

At station SV-1, the PM_{2.5}/PM₁₀ ratio reached relatively high values during winter (0.8-0.9), coinciding with increased PM_{2.5} concentrations and a potentially greater influence of combustion-related sources. During spring and autumn, the ratio generally ranged from 0.5 to 0.7, while in summer it decreased to 0.5-0.6, coinciding with a lower relative contribution of PM_{2.5} and a greater contribution of the coarse fraction (Fig. 4. 10).

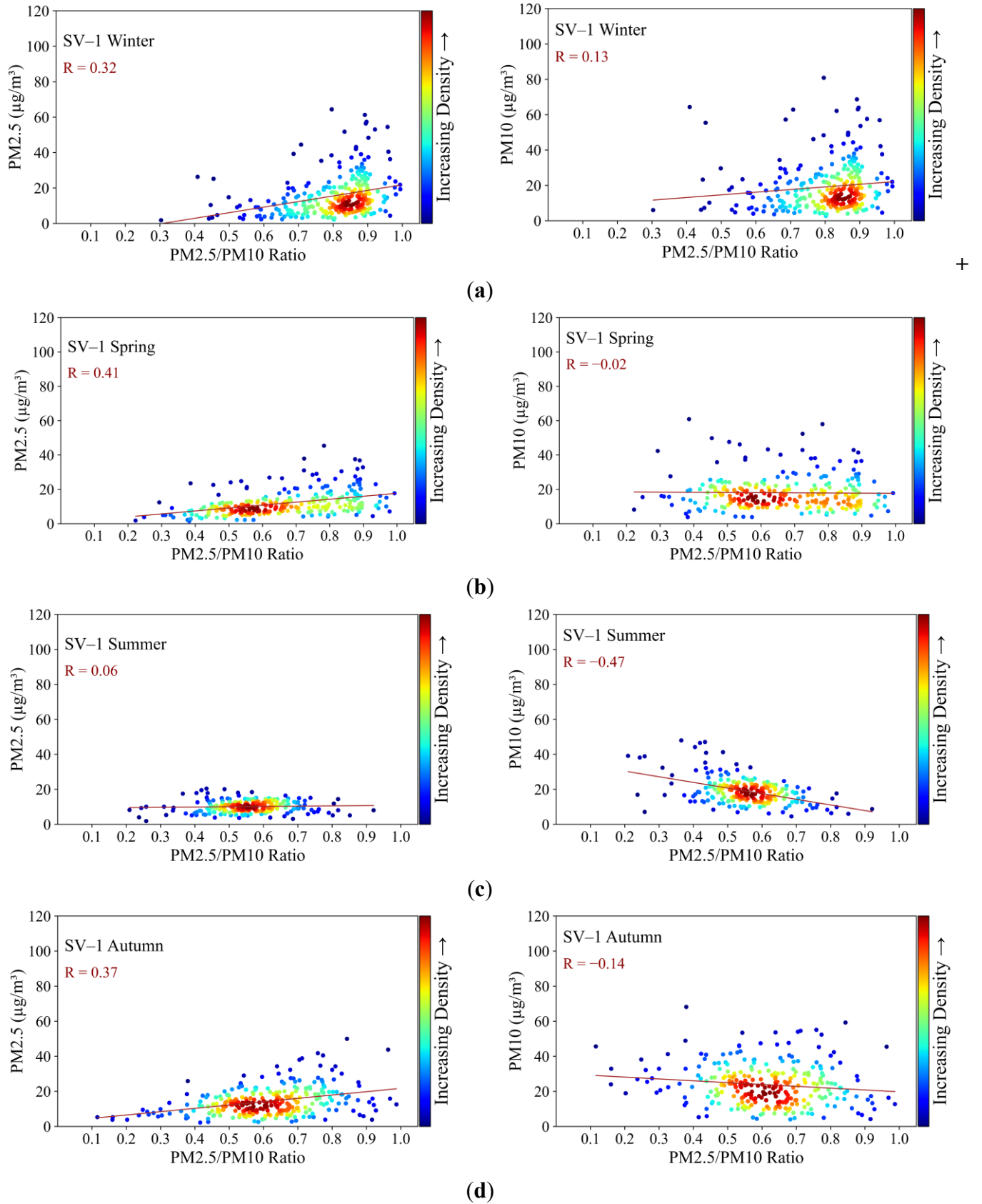


Fig. 4. 10. Relationships between $PM_{2.5}$ and PM_{10} concentrations and the $PM_{2.5}/PM_{10}$ Ratio at station SV-1 for Winter (a), Spring (b), Summer (c), and Autumn (d), 2019-2023

The analysis of the seasonal distribution of the $PM_{2.5}/PM_{10}$ ratio at SV-1 revealed a higher frequency of low values (≤ 0.5) during the warm season, reaching 34.5% in summer 2023. In contrast, high values (> 0.7) predominated during winter, with frequencies ranging from 75.9%

to 80% during 2020-2022. In 2023, the frequency of values >0.7 decreased to 56.3%, while the concurrent reduction in high-ratio values observed during spring and summer suggests a relatively lower contribution of the fine particulate fraction. This evolution coincided with the implementation of measures under the Air Quality Plan. However, the observed changes may also reflect interannual variability in meteorological conditions (Fig. 4. 11).

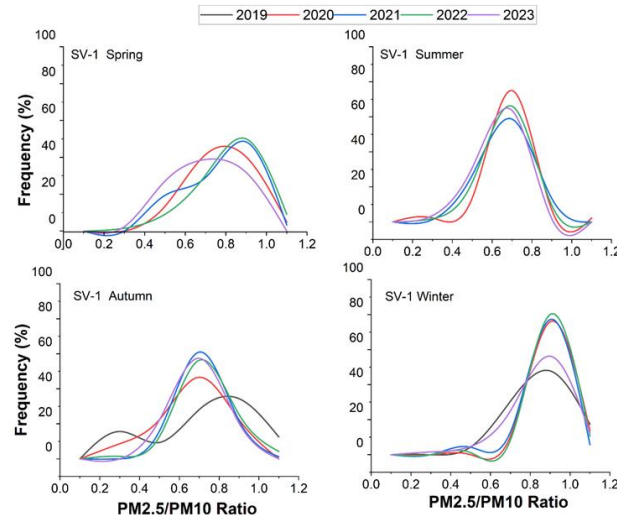
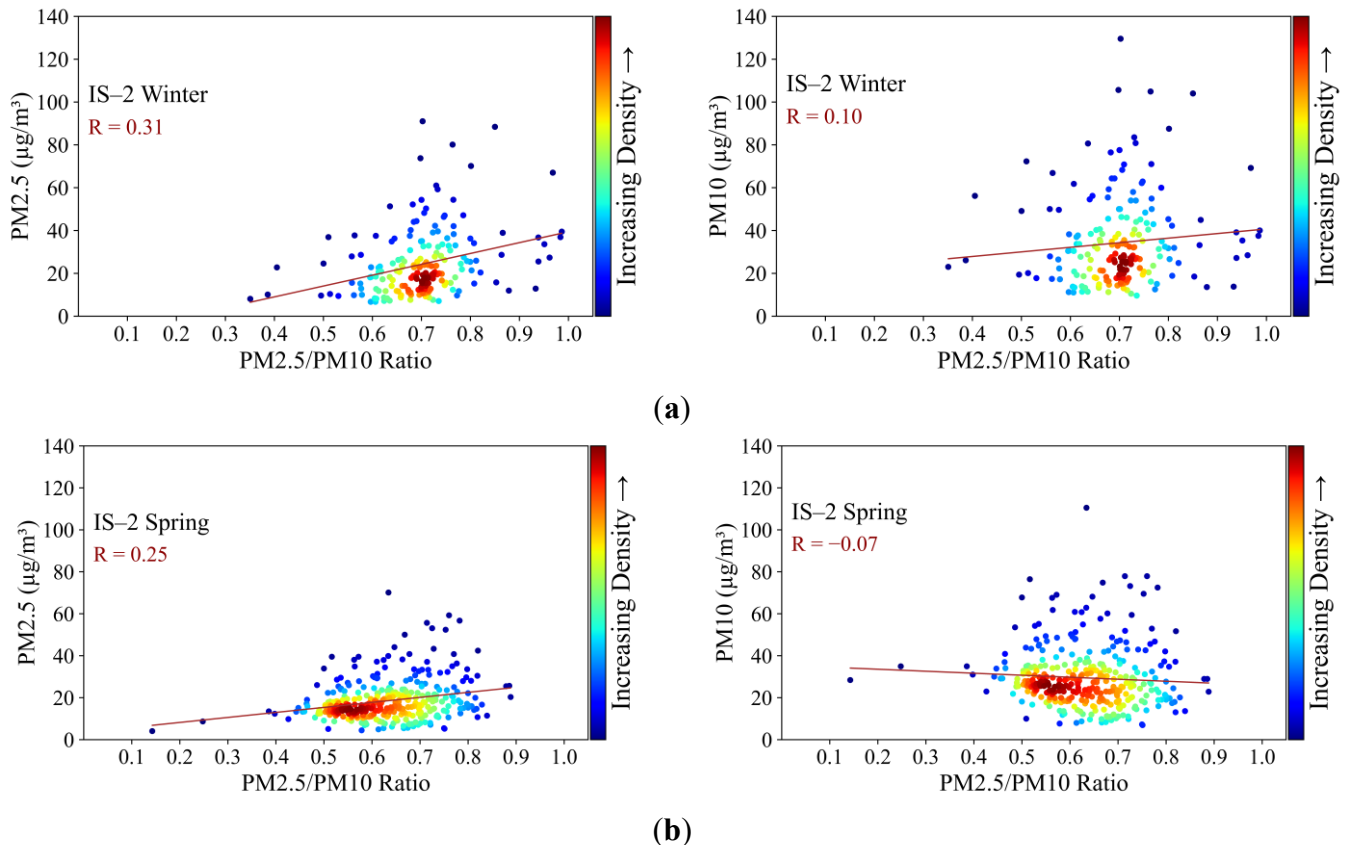


Fig. 4. 11. Seasonal frequency distribution of $PM_{2.5}/PM_{10}$ Ratio values at SV-1 during 2019-2023

At station IS-2, during the cold season, the $PM_{2.5}/PM_{10}$ ratio was predominantly between 0.65 and 0.75 and was associated with higher $PM_{2.5}$ concentrations. During spring and autumn, values between 0.5 and 0.65 predominated, whereas in summer the ratio was generally between 0.5 and 0.6, coinciding with a lower relative contribution of the fine fraction and a greater contribution of the coarse fraction. (Fig. 4. 12).



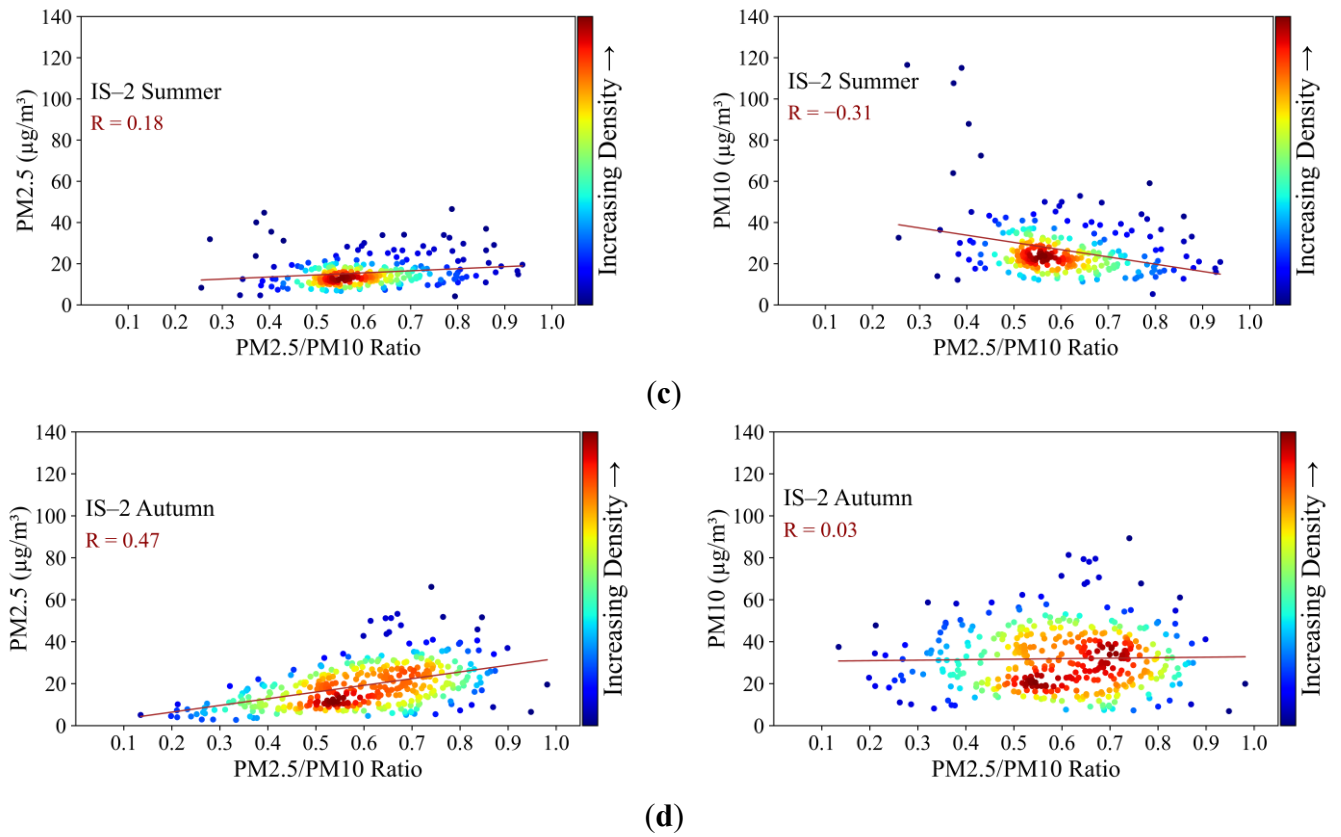


Fig. 4. 12. Relationships between $PM_{2.5}$ and PM_{10} concentrations and the $PM_{2.5}/PM_{10}$ ratio at station IS-2 for winter (a), spring (b), summer (c), and autumn (d), 2019-2023

At station IS-2, the seasonal distribution of the $PM_{2.5}/PM_{10}$ ratio showed a predominance of values >0.5 during the cold season, with frequencies of values >0.7 reaching up to 95% during 2020-2022. In contrast, the warm season was characterized by an increased frequency of lower ratio values, reaching 66.7% in summer 2023. In 2023, a decrease in the frequency of high ratio values was observed across all seasons, suggesting a lower relative contribution of the fine particulate fraction. This change may be associated with both variations in anthropogenic emissions and interannual variability in meteorological conditions (Fig. 4. 13).

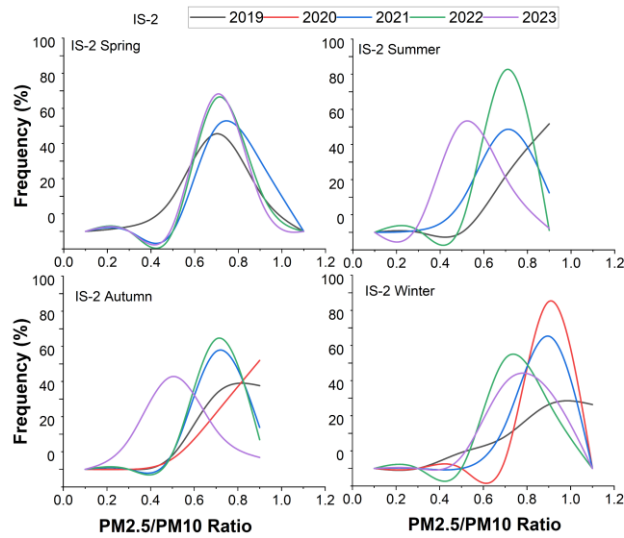


Fig. 4. 13. Seasonal frequency distribution of $PM_{2.5}/PM_{10}$ Ratio values at IS-2 during 2019-2023

At BT-1, the $PM_{2.5}/PM_{10}$ ratio was highest during winter (0.65-0.85), with weaker positive correlations with $PM_{2.5}$ ($R = 0.33$) than in spring, autumn, and summer ($R = 0.54, 0.45$, and 0.56 ,

respectively). During the warm season, lower ratio values predominated (0.3-0.6), indicating a greater relative contribution of the coarse fraction. The relationship between PM_{10} and the ratio was not significant in winter and spring and was negative in summer and autumn. (

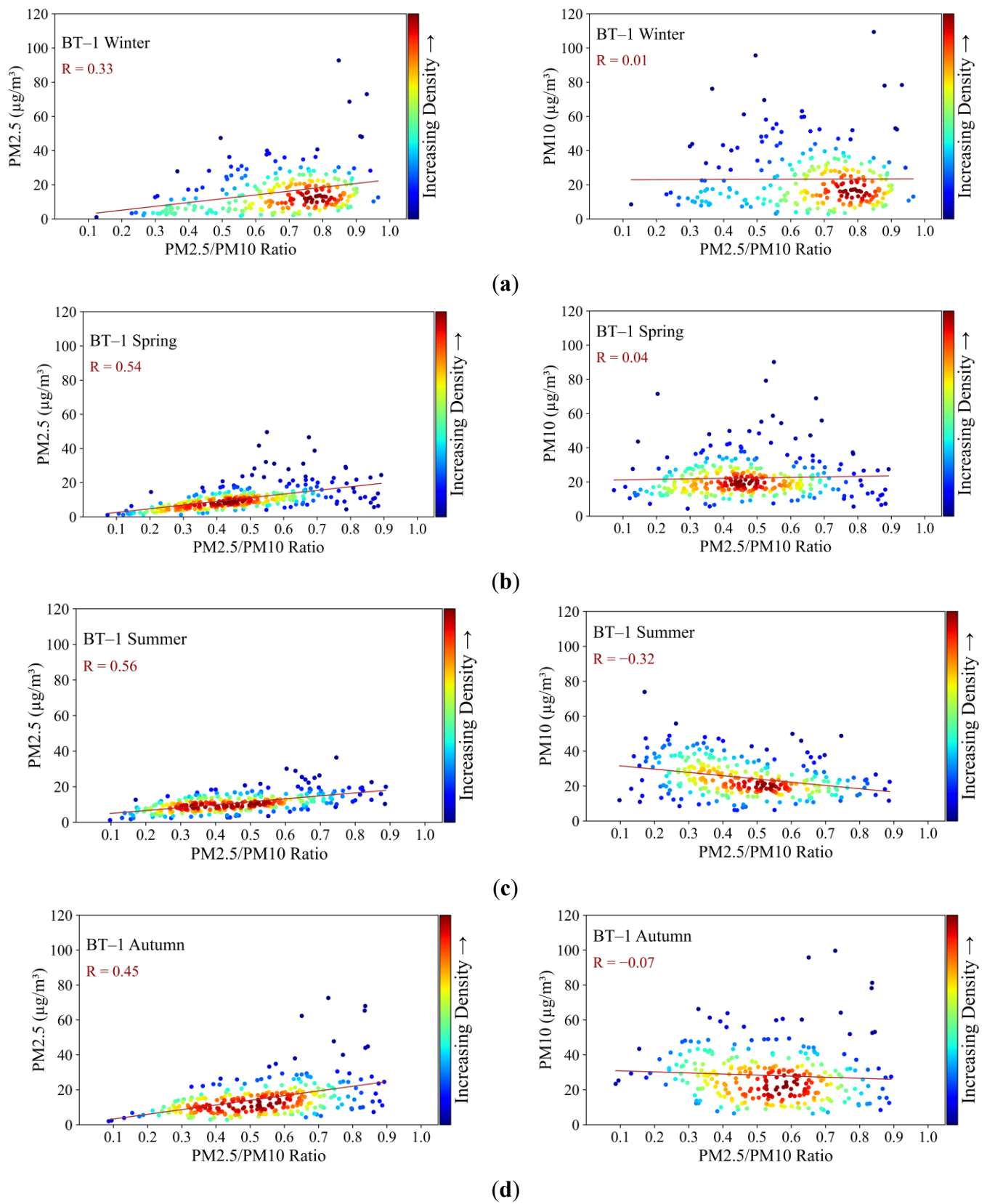


Fig. 4. 14).

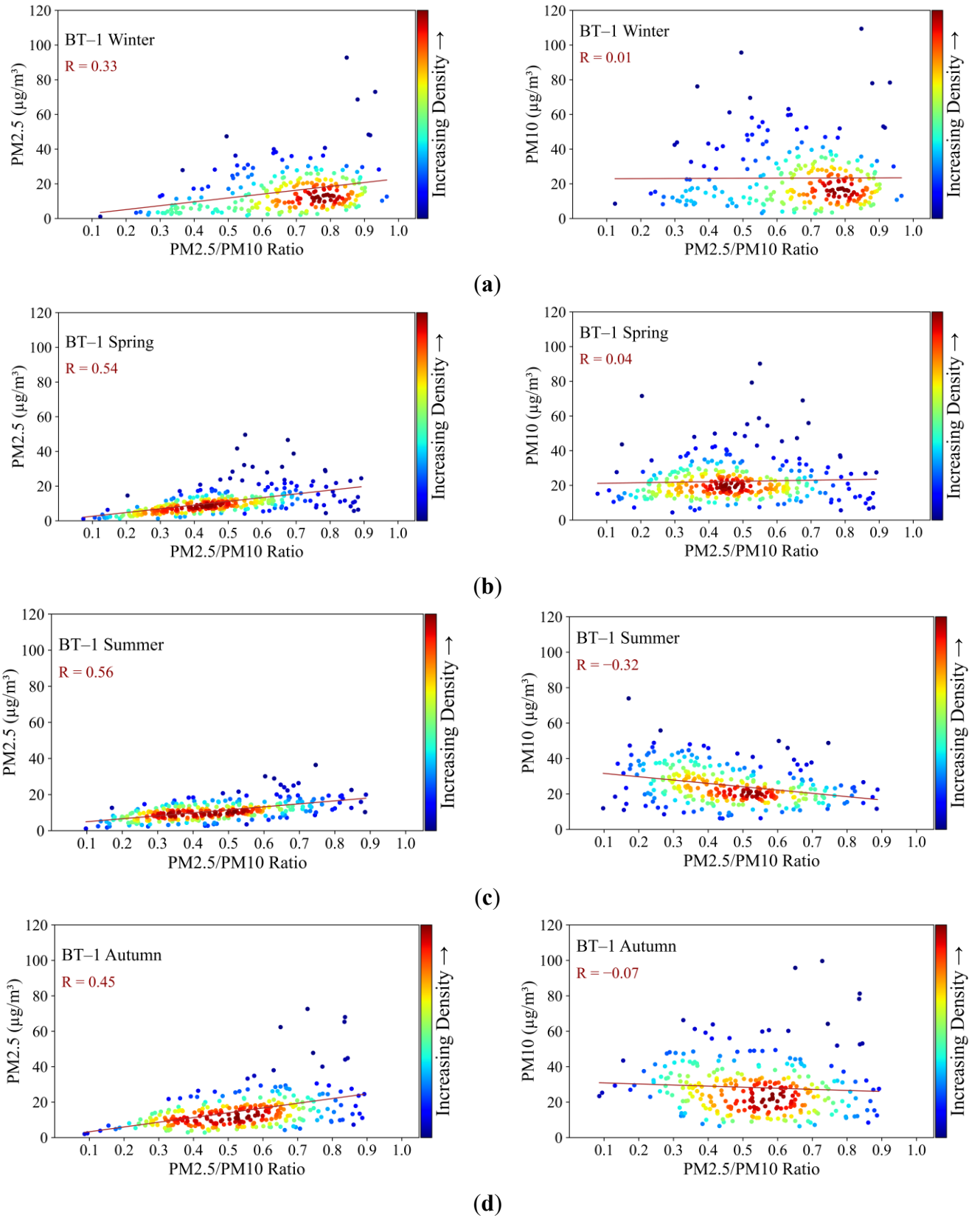


Fig. 4. 14. Relationships between $PM_{2.5}$ and PM_{10} concentrations and the $PM_{2.5}/PM_{10}$ Ratio at station BT-1 for Winter (a), Spring (b), Summer (c), and Autumn (d), 2019-2023

At BT-1, 2023 was characterized by a marked increase in the frequency of $PM_{2.5}/PM_{10} \leq 0.5$ during winter (88.5%) and spring (84.4%), indicating a lower relative contribution of the fine fraction. In contrast, values >0.5 predominated during 2019-2022, particularly in winter, when their frequency decreased from 81.3% in 2019 to 11.5% in 2023. In summer, the

frequency of values ≤ 0.5 decreased from 96.2% in 2022 to 34.2% in 2023, suggesting an increased contribution of the fine fraction. (Fig. 4. 15).

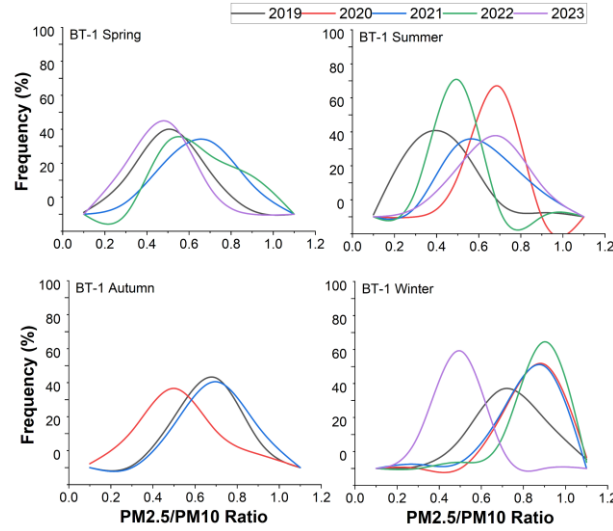


Fig. 4. 15. Seasonal frequency distribution of $PM_{2.5}/PM_{10}$ Ratio values at BT-1 during 2019-2023

4.2.6. Influence of Air Temperature on the $PM_{2.5}/PM_{10}$ Ratio

In Suceava, high $PM_{2.5}/PM_{10}$ ratios (0.8-0.9) were mainly observed at temperatures of 0-5°C, consistent with conditions associated with residential biomass combustion and thermal inversions, whereas at temperatures above 20°C the ratio generally decreased below 0.5, indicating a greater relative contribution of coarse particles. In Botoșani, higher ratios (0.6-0.8) were more dispersed at low temperatures, while the highest data density occurred at 20-25°C, with ratios of 0.3-0.4. In Iași, the highest data density was observed at 0-10°C, with ratios of 0.6-0.75, whereas at 20-30°C the ratio decreased to 0.35-0.5. Overall, temperature appears to be an indicator of shifts in the relative contribution of particulate fractions rather than a direct determinant of $PM_{2.5}/PM_{10}$ ratio variability (Fig. 4. 16).

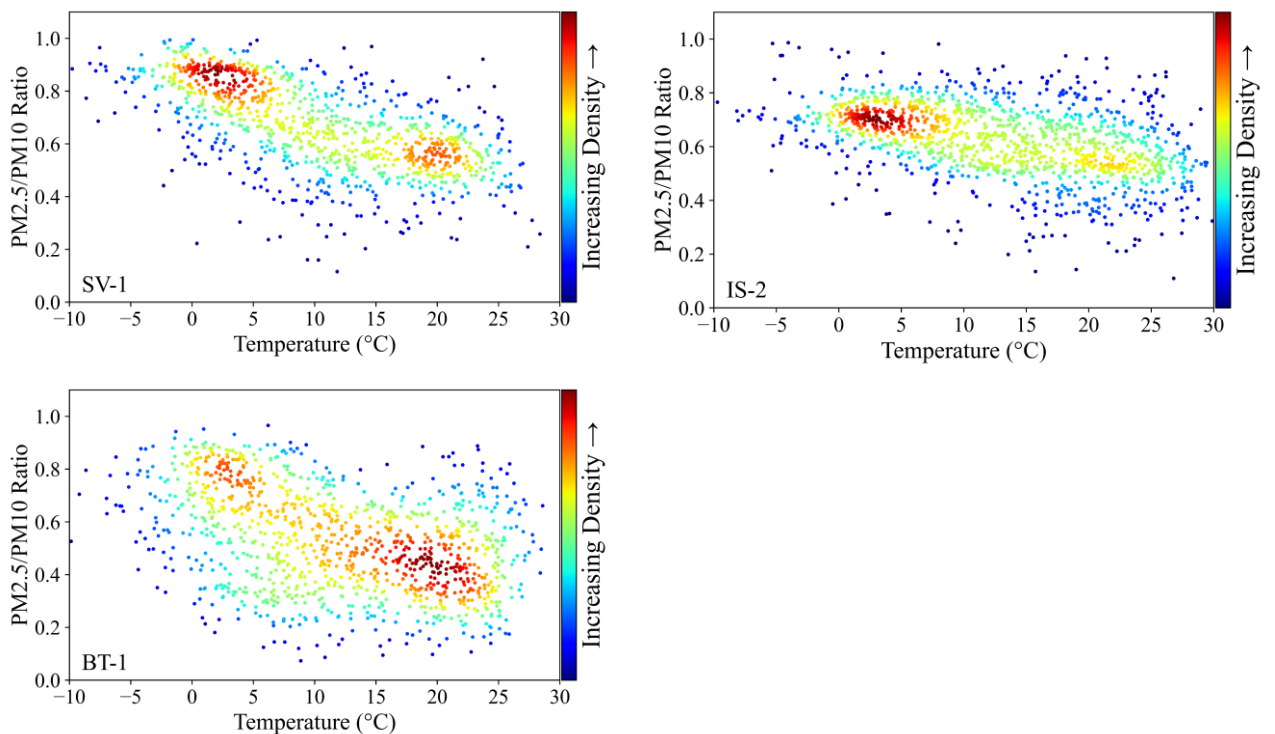


Fig. 4. 16. Distribution of $PM_{2.5}/PM_{10}$ Ratio values as a function of air temperature at SV-1 (a), IS-2 (b), and BT-1 (c), 2019-2023

4.2.7. Spatial Variability of the $PM_{2.5}/PM_{10}$ Ratio

The analysis for 2019-2023 revealed spatial differences among the three monitoring stations. The mean $PM_{2.5}/PM_{10}$ ratio was 0.66 at SV-1, 0.62 at IS-2, and 0.54 at BT-1, indicating a relatively greater contribution of the fine fraction at SV-1 and IS-2.

The coefficients of variation were 0.251 at SV-1 and 0.213 at IS-2, indicating moderate variability of the $PM_{2.5}/PM_{10}$ ratio. In contrast, BT-1 recorded the lowest mean ratio (0.54) and the highest coefficient of variation ($CV = 0.360$), suggesting a relatively greater contribution of the coarse fraction and higher variability in particulate fraction characteristics (Table 4.1).

The coefficient of divergence (COD) indicated the highest similarity between SV-1 and IS-2 ($COD = 0.135$) and the greatest divergence between BT-1 and IS-2 ($COD = 0.236$). These results indicate spatial differences in particulate fraction characteristics, emission influences, and local or meteorological conditions (Table 4. 1).

Table 4. 1. Mean, coefficient of variation, and coefficient of divergence of the $PM_{2.5}/PM_{10}$ ratio, 2019-2023

	Mean	CV	COD IS-2	BT-1
SV-1	0.66	0.251	0.135	0.208
IS-2	0.62	0.213		0.236
BT-1	0.54	0.360		

The strongest Pearson correlation was observed between SV-1 and IS-2 ($R = 0.441$), despite their distance of approximately 115 km, while the correlation between SV-1 and BT-1, located 32 km apart, was $R = 0.414$. The weakest correlation was observed between BT-1 and IS-2 ($R = 0.090$), despite their distance of approximately 96 km.

These results indicate that spatial proximity may contribute to similarities in $PM_{2.5}/PM_{10}$ ratio variability but is not the only influencing factor. Differences between stations may also reflect local emission sources and site-specific meteorological conditions. Thus, the relationship between inter-station distance and correlation strength is not strictly linear.

4.3. Comparative evaluation of CHIMERE and ENSEMBLE model estimates versus in-situ measurements of particulate matter in the Iași urban area

The study evaluated the performance of the CHIMERE and Ensemble models in estimating PM_{10} and $PM_{2.5}$ concentrations in Iași, using measurements from the IS-2 monitoring station in 2024 as the reference dataset. The evaluation assessed the ability of the models to reproduce temporal variability, mean concentrations, and pollution episodes using a set of statistical performance metrics (Drăgoi et al., 2026b).

4.3.1. Observed and Modeled PM_{10} Concentrations

In 2024, the PM₁₀ daily limit value (50 µg/m³, with a maximum of 35 exceedances per year) was exceeded on 25 days, with the highest number of exceedances recorded in October and November. CHIMERE reproduced 7 of these exceedances, whereas the Ensemble model reproduced none (Fig. 4. 17). Both models captured the overall temporal pattern of PM concentrations but underestimated the observed peak values. Model performance was generally better during the cold season and transitional periods, while larger errors were observed during the warmer months.

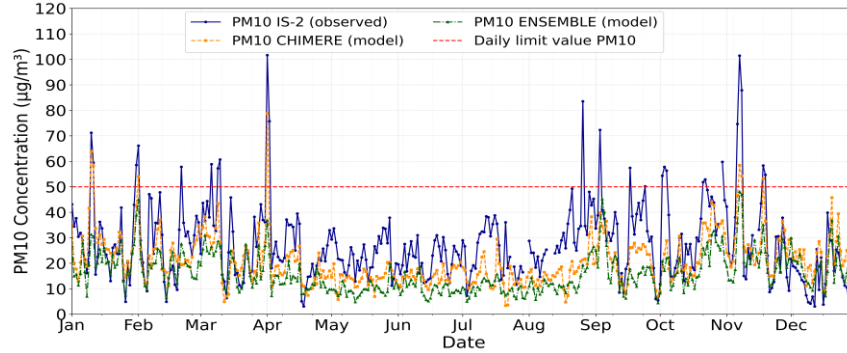


Fig. 4. 17. Daily PM₁₀ concentrations, Observed vs Models (2024)

A representative pollution episode was identified on 1 April 2024, when the PM₁₀ concentration measured at IS-2 reached 101.63 µg/m³. For the same day, CHIMERE estimated a concentration of 78.8 µg/m³ and indicated PM₁₀ concentrations of 50-80 µg/m³ across north-eastern Romania (Fig. 4. 18). The simulated spatial distribution was consistent with the long-range transport of Saharan dust, highlighting the usefulness of regional modelling for interpreting pollution episodes that cannot be adequately explained by local measurements alone.

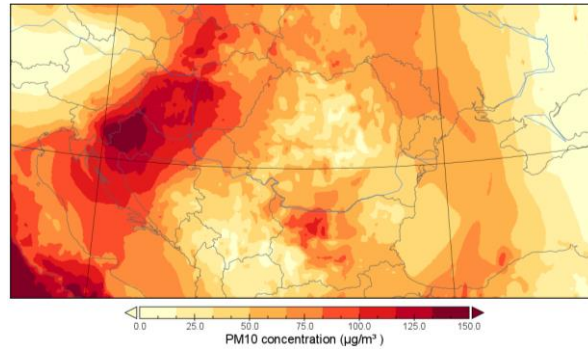


Fig. 4. 18. Spatial distribution of PM₁₀ concentrations (µg/m³) over Romania and surrounding regions on April 1, 2024, as simulated by the CHIMERE model

4.3.2. Observed and Modeled Daily PM_{2.5} Concentrations

For PM_{2.5}, both models reproduced the seasonal variability, with higher concentrations during the cold season and lower concentrations during the warm season, although they underestimated the observed peak values. The performance of the two models was more similar than for PM₁₀ (Fig. 4. 19). The Ensemble and CHIMERE models simulated similar annual mean PM_{2.5} concentrations, of 13.15 and 13.56 µg/m³, respectively. Both values were below the annual mean concentration recorded at IS-2 (15.52 µg/m³), although the relative underestimation was smaller than that observed for PM₁₀.

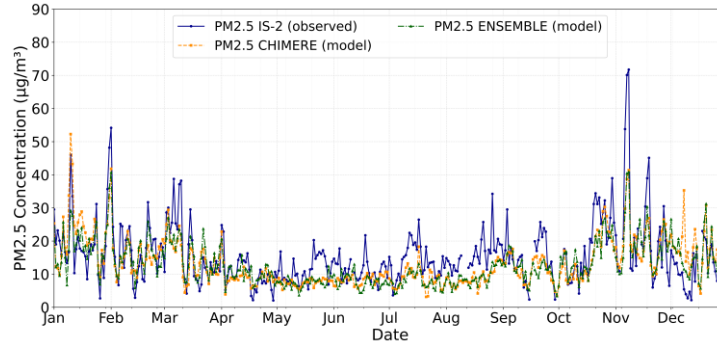


Fig. 4. 19. Daily $PM_{2.5}$ concentrations, Observed vs Models (2024)

The Taylor diagram for PM_{10} (Fig. 4. 20 a) confirms these findings. CHIMERE exhibited a normalized standard deviation of approximately 0.66, which was closer to the reference value of 1 than that of the Ensemble model (0.54), as well as a slightly higher correlation coefficient ($R = 0.64$ versus 0.60). The cRMSD values also indicated a better agreement between CHIMERE and the observations. Overall, the Taylor diagram shows that CHIMERE reproduced both the temporal variability and the overall structure of the PM_{10} concentrations observed at IS-2 more closely.

For $PM_{2.5}$, the Taylor diagram (Fig. 4. 20 b) indicated slightly better agreement for the Ensemble model, which achieved $R = 0.68$, a normalized standard deviation of 0.73, and cRMSD = 0.73, compared with $R = 0.65$, $\sigma = 0.80$, and cRMSD = 0.78 for CHIMERE. The RMSE was also slightly lower for Ensemble ($7.22 \mu\text{g}/\text{m}^3$) than for CHIMERE ($7.52 \mu\text{g}/\text{m}^3$). Thus, the Taylor diagram indicates a slightly better ability of the Ensemble model to reproduce the daily variability of $PM_{2.5}$, although both models showed amplitude errors and underestimated the observed variability.

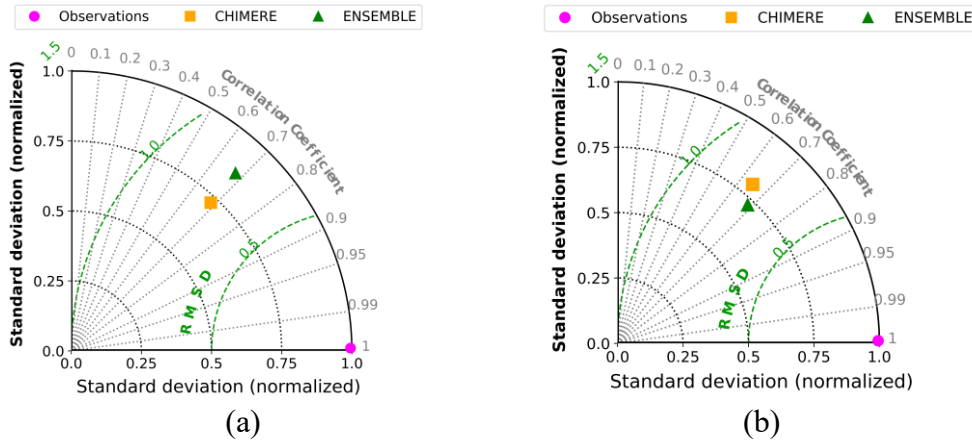


Fig. 4. 20. Taylor diagrams of daily concentrations (normalized) predicted by Chimere and Ensemble versus reference station IS-2 (01.01. - 31.12.2024): (a) PM_{10} ; (b) $PM_{2.5}$.

Overall, the results indicate that CHIMERE showed better agreement with the observations for PM_{10} , whereas the Ensemble model performed slightly better for $PM_{2.5}$. Both models reproduced the overall temporal patterns and seasonal variability of particulate matter concentrations, but underestimated the observed pollution peaks, particularly during the warm season. The combined use of measurements from air quality monitoring stations and atmospheric modelling provides a more comprehensive representation of the temporal and spatial variability of particulate matter concentrations in the urban environment.

4.4. Assessment of PM₁₀ and PM_{2.5} Concentrations against Current and Future Limit Values under Law No. 104/2011, Directive 2008/50/EC, and Directive (EU) 2024/2881

Assessing the evolution of PM₁₀ and PM_{2.5} concentrations in relation to current limit values and the more stringent values introduced by the new Directive is important for evaluating not only the current air quality status but also the extent to which observed particulate matter concentrations comply with forthcoming regulatory requirements.

4.4.1. Assessment of Annual Mean PM₁₀ and PM_{2.5} Concentrations

The annual mean PM₁₀ concentrations recorded at the monitoring stations during 2017-2024 were below the annual limit value of 40 µg/m³ established under Law No. 104/2011.

With respect to the future annual PM₁₀ limit value of 20 µg/m³ established by Directive (EU) 2024/2881, only station SV-1 complied with this value during 2022-2023. The compliance assessment did not include data from station SV-2 for the 2022-2024 period or data from IS-2 for 2024, as these datasets were insufficient to meet the data quality objectives and aggregation criteria established under Law No. 104/2011 (Section 4.4.1 of the thesis).

The annual mean PM_{2.5} concentrations recorded at the urban background stations SV-1, IS-2, and BT-1 during 2017-2024 were below the annual limit value of 25 µg/m³ established under Law No. 104/2011, except at IS-2 in 2017 and 2018.

If the annual limit value of 10 µg/m³ established by Directive (EU) 2024/2881 had been applied, the annual mean PM_{2.5} concentrations recorded at all three monitoring stations in the study area where PM_{2.5} is monitored would have exceeded this future limit value.

4.4.2. Assessment of Daily PM₁₀ and PM_{2.5} Concentrations

During the analysed period (2017-2024), the frequency of exceedances of the daily PM₁₀ limit value of 50 µg/m³ did not exceed the maximum of 35 exceedances permitted under Law No. 104/2011, except at station IS-2 in 2019, when 36 exceedances were recorded. The maximum permitted number of 35 exceedances was also reached at station SV-2 in 2019 and at station IS-2 in 2020.

Applying the future daily limit value of 45 µg/m³, which may not be exceeded more than 18 times in a calendar year under Directive (EU) 2024/2881, results in a substantial increase in the number of exceedances at all stations analysed. Most stations would exceed the threshold of 18 exceedances per year, except SV-1 in certain periods and BT-1 during 2021-2023. The highest numbers of exceedances were recorded at IS-2 and SV-2, indicating potentially higher population exposure within the areas represented by these stations. For example, in 2019, the number of exceedances at SV-2 would increase from 35 under the current limit value of 50 µg/m³ to 73 under the future limit value of 45 µg/m³. At IS-2, the number of exceedances would increase from 36 to 52. In contrast, SV-1 exhibited a lower level of exceedances.

Overall, a general decreasing trend in the number of exceedances was observed between 2018 and 2023, followed by a partial increase in 2024 at BT-1 and SV-1, suggesting an overall

improvement in air quality. This pattern may reflect both the effects of emission reduction measures and interannual variability in meteorological conditions.

For PM_{2.5}, Law No. 104/2011 does not establish a daily limit value.

The analysis of exceedances relative to the future daily PM_{2.5} limit value of 25 µg/m³, which may not be exceeded more than 18 times in a calendar year under Directive (EU) 2024/2881, revealed substantial differences among the monitoring stations during 2017-2024. At IS-2, the highest number of values above the future limit value was recorded, exceeding 150 days per year in 2017-2018, followed by a decrease after 2020. Nevertheless, the number of exceedances remained above the threshold of 18 per year, indicating that the station would continue to record exceedances under this future requirement. At SV-1, the number of exceedances was lower and fell below the threshold of 18 exceedances per year in 2022-2023. For BT-1, the available data were insufficient to assess compliance with this future limit value.

Achieving the 2030 targets will require substantial additional reductions in emissions, particularly from road traffic, residential heating, and certain industrial sectors (EEA, 2025).

4.5. Proposed Additional Indicators for Assessing PM₁₀ and PM_{2.5} Concentrations

The compliance indicators defined in the EU Ambient Air Quality Directives (annual/daily limit values, maximum number of exceedances, information and alert thresholds) are necessary for assessing compliance with legislation. However, they only indicate whether a standard has been exceeded, without quantifying the extent to which it has been exceeded.

The study proposes three complementary and original indicators designed to quantify the magnitude of exceedances, the duration of pollution episodes, and the differences in toxicity between the fine PM_{2.5} fraction and the coarse PM fraction: Excess Concentration (*EC*), which aggregates the concentration excess above the daily limit value over the course of a year, Episode Index (*EI*), which integrates both the intensity and duration of pollution episodes, and Toxic Load Index (*TLI*), which combines the PM_{2.5}/PM₁₀ ratio, relevant to the characterization of emission sources, with the exceedance of the PM_{2.5} limit value (Drăgoi et al., 2025b).

The calculation methodology for these three indicators, together with the proposed exposure categories for each indicator, is presented in Section 3.4.- Chapter 3. Materials and Methods.

4.5.1. Excess Concentration at air quality monitoring stations in the Municipalities of Suceava, Iaşi, and Botoşani

Based on the *EC* classification of PM according to the thresholds initially established (Section 3.4.1), the results show significant variations in PM levels across years and locations. Relating the number of daily exceedances of the future limit value to the annual excess makes it possible to distinguish between situations characterized by frequent, low-magnitude exceedances and those involving fewer but substantially higher exceedances.

In the municipality of Iași, the *EC* values for PM₁₀ show a decreasing trend, from the *Very High* level in 2020 to *Moderate* in 2022-2024, while the *EC* values for PM_{2.5} range from *Very High* in 2021 to *Moderate* in 2023-2024.

In the municipality of Suceava, the *EC* values for PM₁₀ show significant variation between monitoring stations: the industrial-type SV-2 station records *Very High* values in 2019-2020, whereas the urban background SV-1 station falls within the *Low* class. For PM_{2.5}, at the SV-1 station, the *EC* values range from *Moderate* in 2020, 2021, and 2024 to *Low* in 2022-2023.

In the municipality of Botoșani, the *EC* values for both PM₁₀ and PM_{2.5} range from *Moderate* to *Low* (Fig. 4. 21).

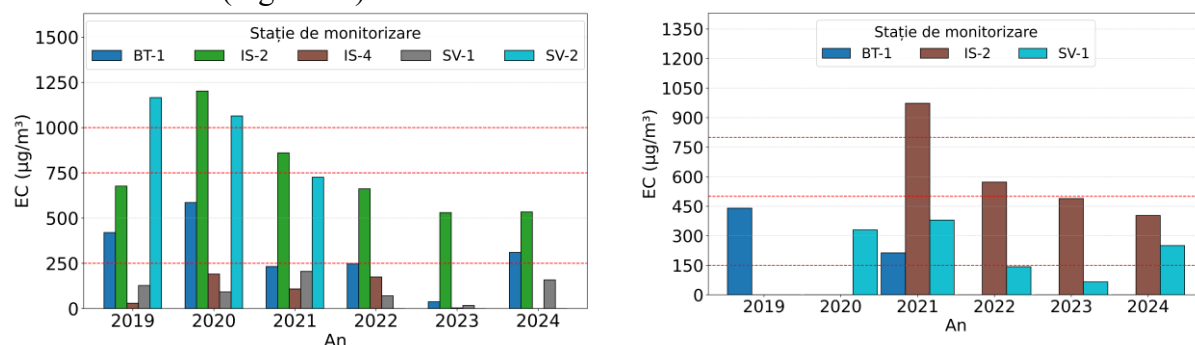


Fig. 4. 21. Annual Excess Concentration PM₁₀ and PM_{2.5}

(— *EC* thresholds according to Section 3.4)

4.5.2. Episode Index (*EI*) at air quality monitoring stations in the Municipalities of Suceava, Iași, and Botoșani

Applying the *EI* indicator (Section 3.4.2), significant pollution episodes were identified in Iași, including an Extremely High PM_{2.5} episode ($EI = 106 \mu\text{g}/\text{m}^3$), indicating a high level of air quality-related risk. In Suceava (SV-2), the *EI* indicator identified *Very High* PM₁₀ pollution episodes, whereas in Botoșani, the *EI* indicator identified occasional but intense PM_{2.5} pollution episodes.

Table 4.2 summarizes the main types of exceedances and the *EC* and *EI* classifications across the municipalities included in the study for the 2019-2024 period.

Table 4.2. Summary of key findings on exceedances and *EC*/*EI* classifications by city, 2019-2024

City (Station Code)	PM ₁₀ Exceedances*	PM _{2.5} exceedances**	<i>EC</i> categories	<i>EI</i> highlights
Iași (IS-2)	Frequent (≥ 50 days in 2019-2021)	Up to 83 days (2021)	PM ₁₀ - ranges from <i>Very High</i> (2020) to <i>Moderate</i> (2022 -2024) PM _{2.5} ranges from <i>Very High</i> (2021) to <i>Moderate</i> (2023, 2024)	Severe episodes, including one extreme PM _{2.5} event ($EI= 106 \mu\text{g}/\text{m}^3$)
Suceava (SV-2, SV-1)	<i>Very High</i> at SV-2 (73 days in 2019); <i>Low</i> at SV-1 (≤ 20 days in 2022-2024)	Up to 37 days SV-1 (2021)	PM ₁₀ - ranges from <i>Very High</i> SV-2 (2019, 2020) to <i>Low</i> (SV-1) PM _{2.5} (SV-1) - ranges from <i>Moderate</i> (2020, 2021, 2024) to <i>Low</i> (2022, 2023)	Episodic PM ₁₀ peaks at SV-2; minor episodes at SV-1

City (Station Code)	PM ₁₀ Exceedances*	PM _{2.5} exceedances**	EC categories	EI highlights
Botoșani (BT-1)	Moderate (≤30 days in 2021-2024)	Up to 36 days (2019)	Moderate to Low	Occasional moderate PM _{2.5} episodes

*Exceedances - Days above 45 µg/m³ (PM₁₀, Directive 2024/2881).

**Exceedances - Days above 25 µg/m³ (PM_{2.5}, Directive 2024/2881).

4.5.3. Toxic load Index (TLI) at air quality monitoring stations in the Municipalities of Suceava, Iași, and Botoșani

The analysis of the *TLI* indicator for the years that met the data quality objectives for PM_{2.5} revealed differences among the monitoring stations analyzed. The most frequent and highest *TLI* values were recorded at the IS-2 station, where the largest number of episodes in the *High* and *Very High* risk categories was also identified. The SV-1 station showed an intermediate pattern, whereas at BT-1, *TLI* > 0 values were less frequent and were generally associated with Low to Moderate levels.

The results confirm the usefulness of the *TLI* indicator as a complementary indicator to PM_{2.5} concentrations and the number of exceedances, by integrating the concentration level of fine particulate matter and its contribution to the total PM₁₀ mass.

4.5.4. Added Value of EC, EI, and TLI

The use of the proposed indicators in conjunction with established regulatory indicators enables authorities to identify risks at an earlier stage, differentiate between distinct categories of pollution episodes, develop locally tailored intervention strategies, and prioritize measures aimed at reducing air pollution (Fig. 4. 22).

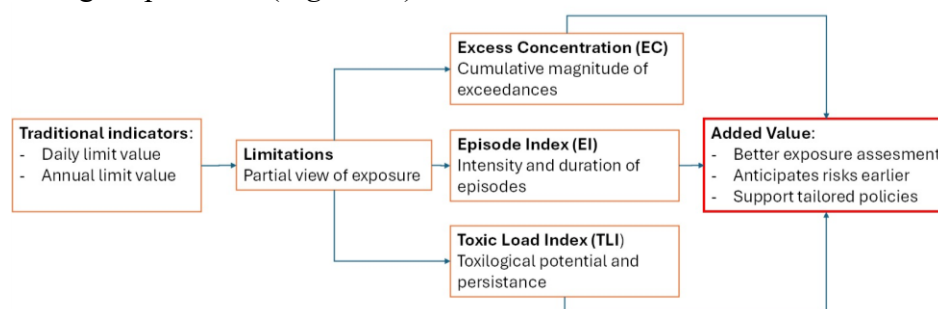


Fig. 4. 22. Complementarity of Excess Concentration (EC), Episode Index (EI), and Toxic Load Index (TLI) with regulatory indicators

4.5.5. Policy Implications

The study proposes the use of the *EC*, *EI*, and *TLI* indicators to complement conventional air quality assessment by providing information on cumulative exposure, pollution episode intensity and duration, and the toxicological potential of particulate matter. Their application can support policies tailored to local and seasonal conditions.

In Suceava, *EC* values highlight the need for long-term measures targeting emissions from residential heating and road traffic. In Iași, *EI* values support the implementation of rapid

response measures during short and intense pollution episodes. In Botoșani, *TLI* values emphasize the importance of continued monitoring and the protection of vulnerable groups.

Overall, the findings support a locally tailored approach to air pollution management. The proposed indicators can assist authorities in identifying priorities, selecting targeted measures, and improving the effectiveness of air quality management.

4.6. Extension of the Analysis of Proposed Additional Indicators for Assessing PM_{10} and $PM_{2.5}$ Concentrations in Romania

In this study, the analysis of the three indicators was extended to the entire national network of stations that simultaneously monitor PM_{10} and $PM_{2.5}$. Data obtained using the gravimetric reference method were used, with the exception of certain stations where automated measurements were employed: for PM_{10} , stations DJ-2 (2023-2025), TR-3, TR-5, and stations B-12-B-30, and for $PM_{2.5}$, stations B-12-B-30, TR-3, TR-5, IS-1 (2024-2025), IS-4, IS-5, CJ-5, and DJ-2 (2023-2025). It should be noted that data obtained from automatic analyzers must be treated with caution, given that they were not obtained through gravimetric reference measurements. Furthermore, at the time of writing this thesis, the data for 2025 had not yet been certified.

A total of 80 stations from the RNMCA were included, classified as follows:

- 57 urban background stations;
- 4 suburban stations: B-7; CJ-3; CL-2; IS-5.
- 12 traffic-type stations: B-12; B-13; B-14; B-15; B-16; B-17; B-18; B-19; B-20; B-30; B-6; IS-1
- 2 industrial stations located in urban areas: B-5; MH-1.
- 5 rural stations: B-8; CL-3; DJ-6; IS-4; IS-6.

The objective was, on the one hand, to test the stability and relevance of the three indicators under different climatic conditions, station types, and geographic areas, and, on the other hand, to provide a comprehensive picture of exposure patterns at the national level, relevant for planning measures to reduce PM pollution by 2030.

4.6.1. Excess Concentration (*EC*) at Air Quality Monitoring Stations in Romania

The *EC* was calculated for the period 2015–2025, based on daily concentrations and the number of exceedances of the limit values reported under Directive (EU) 2024/2881 (Section 3.4.1).

A combined analysis of the number of exceedances and the *EC* value highlights two distinct components of PM_{10} exposure: the frequency and intensity of pollution episodes. Some monitoring stations record few exceedances but with very high *EC* values, indicating rare and intense episodes, while others have many exceedances but moderate *EC* values, suggesting more frequent and prolonged exposure.

At the national level, exposure patterns vary widely, ranging from stations with low *EC* values and few exceedances to situations characterized by extreme values and numerous

exceedances. Stations that consistently remain within intermediate *EC* ranges over time indicate chronic exposure to moderate or high levels of PM₁₀, while a shift from higher to lower *EC* categories may indicate an improvement in air quality.

Therefore, risk assessment requires an integrated analysis of the frequency and intensity of exceedances, as neither of these two indicators, when analyzed separately, provides a complete picture of exposure.

For PM₁₀, during the period analyzed, exceedances of the *EC* value exceeding 1000 µg/m³ were recorded both at traffic monitoring stations in major cities (Bucharest, Iaşi) and at urban background stations in central Romania (Braşov, Harghita).

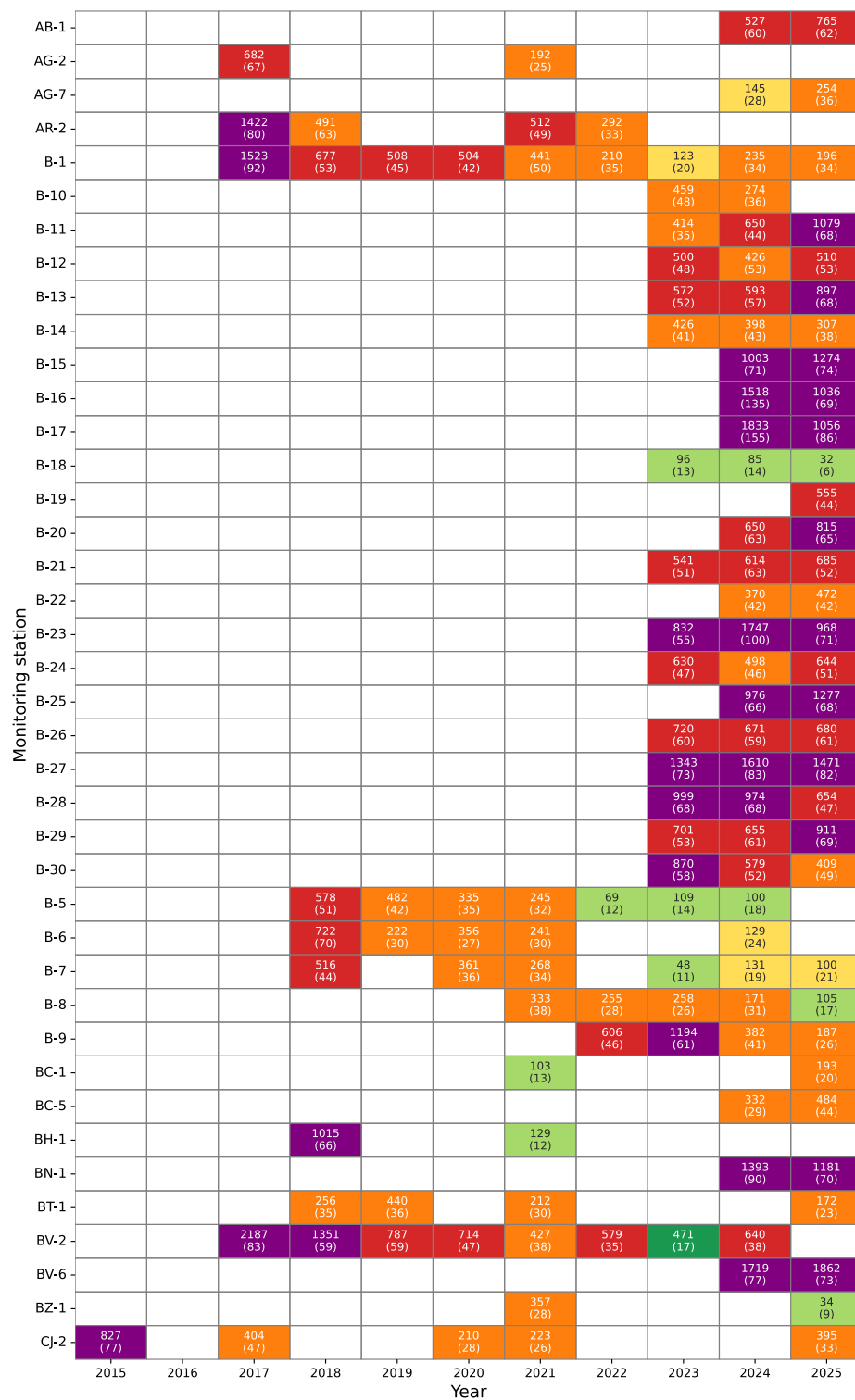
At station CV-2 (Covasna), although the number of exceedances in 2025 remained similar to that in 2024, the decrease in the *EC* indicates a slight improvement in air quality.

In contrast, stations such as AG-7, BH-1, GL-2, CJ-3, and VS-1 predominantly had a Low or Moderate risk level. Overall, the risk of non-compliance with the new PM₁₀ limit values is concentrated in certain urban and industrial areas and is not evenly distributed nationwide.

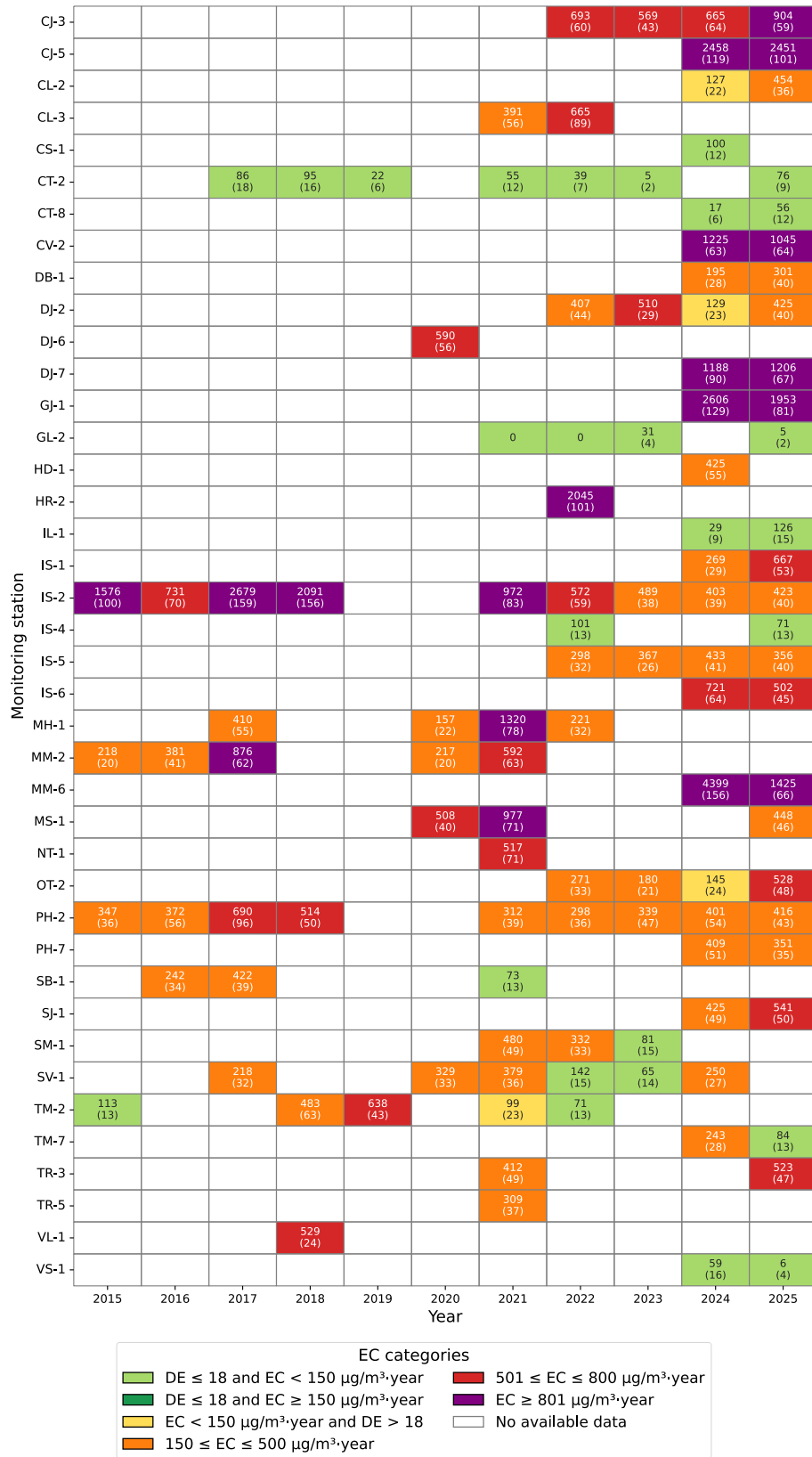
Another finding from the application of the *EC* indicator shows that very low annual *EC* values can coexist with exceedances of the annual limit value. For example, at a station where the *EC* is only 0.4 µg/m³ (BZ-1 in 2023), the *EC* indicator would suggest relatively low exposure to intense episodes, even though the annual average still exceeds the limit value. This situation demonstrates that *EC* is sensitive to the magnitude of daily exceedances, and less so to a moderate but nearly constant background level that is slightly above the annual limit value. In such cases, combining the proposed indicator with standard indicators (daily limit value and annual limit value) becomes useful for interpretation: exceeding the annual limit value signals the need for policy measures, while the very low *EC* value indicates that, from the perspective of short-term pollution episodes, the situation is less severe than in areas where *EC* is in the *High* category.

The *EC* analysis for PM_{2.5} and the number of daily concentrations exceeding the future daily limit value shows that PM_{2.5} is more pronounced in certain large cities, such as Iaşi (IS-2), Bucharest (traffic-based stations and urban background stations), and Cluj (CJ-3 suburban station and CJ-5 urban background station), where some stations have *High* or *Very high* *EC* values and numerous exceedances of the daily limit. In contrast, stations such as CT-2 (Constanţa) show low levels and few exceedances.

The BV-2 station is one of the most problematic, particularly in 2017-2018, when PM_{2.5} exceeded 800 µg/m³, and the number of exceedances relative to the limit value in Directive 2024/2881 was over 50. Subsequently, the situation improved temporarily, but in 2024 the level returned to the *High* category (Fig. 4. 23).



(a)



(b)

Fig. 4. 23. Annual PM_{2.5} EC and daily limit value exceedances at RNMCA monitoring stations (2015-2025). (a) stations AB-1 to CJ-2; (b) stations CJ-3 to VS-1. (EC thresholds are defined in Section 3.4 1).

A comparison of EC values for PM₁₀ and PM_{2.5} shows that the EC for PM₁₀ declined more markedly after 2019-2020, while PM_{2.5} remained high at several monitoring stations. This

suggests that the measures implemented were more effective at reducing dust and coarse particles, but less effective for fine particles originating from residential heating, industry, and traffic.

Over time, many monitoring stations shifted from high to moderate or low levels, though some showed fluctuating trends. These variations indicate that improvements are not always stable and can be influenced by weather conditions and changes in local emissions.

Depending on *EC* trends, areas can be divided into areas with persistent episodes of pollution, which require structural measures, areas with partial success, where PM_{10} has decreased but $PM_{2.5}$ remains high, stabilized areas, with low levels of both pollutants and fluctuating areas, where pollution levels vary from year to year.

In conclusion, the *EC* indicator allows for a clearer identification of high-risk areas and helps prioritize pollution reduction measures, particularly for sources of fine particulate matter.

4.6.2. Episode Index (*EI*) at Air Quality Monitoring Stations in Romania

The *EI* indicator extends the *EC*, based analysis from the annual scale to the episode scale (Section 3.4.2).

An analysis of PM_{10} pollution episodes from 2015 to 2025 reveals the existence of episodes of very high intensity, classified as *Extremely High* and *Very High*. The most intense episodes were recorded between 2015 and 2018, while after 2020 the maximum magnitude of extreme episodes was generally lower, though they continued to occur, confirming the persistence of the risk of episodic pollution.

Very High episodes are the most numerous and occur repeatedly at stations such as BV-2, BV-6, HR-2, CV-2, IS-6, as well as at several stations in Bucharest. Their predominant duration of 2-3 days and their widespread spatial distribution indicate a recurrent nature of short-term pollution episodes. *High* episodes, although less frequent, also indicate situations where the alert threshold is exceeded and contribute to characterizing episodic risk.

In terms of seasonality, the episodes are concentrated primarily during the cold season, with a high frequency in January, February, November, and December. This distribution is consistent with the influence of residential heating, combustion, and meteorological conditions unfavorable to dispersion. However, episodes also occur during the warm season, indicating the involvement of additional sources, such as traffic, dust resuspension, industrial activities, construction work, or regional particle transport.

Spatially, the recurrence of episodes at certain stations, particularly BV-6, HR-2, CV-2, and IS-6, suggests the existence of recurring combinations of local sources, urban characteristics, and atmospheric dispersion conditions. The simultaneous occurrence of episodes at stations in different cities, such as on February 25-26, 2025, also indicates the possibility of common regional atmospheric factors, not just local sources (Fig. 4. 24).

The results indicate a reduction in the magnitude of extreme episodes after 2020, but not their disappearance. The persistence of *Very High* and *Extremely High* episodes, their concentration during the cold season, and their spatial recurrence confirm the relevance of assessing short-duration episodes for characterizing the risk associated with PM_{10} .

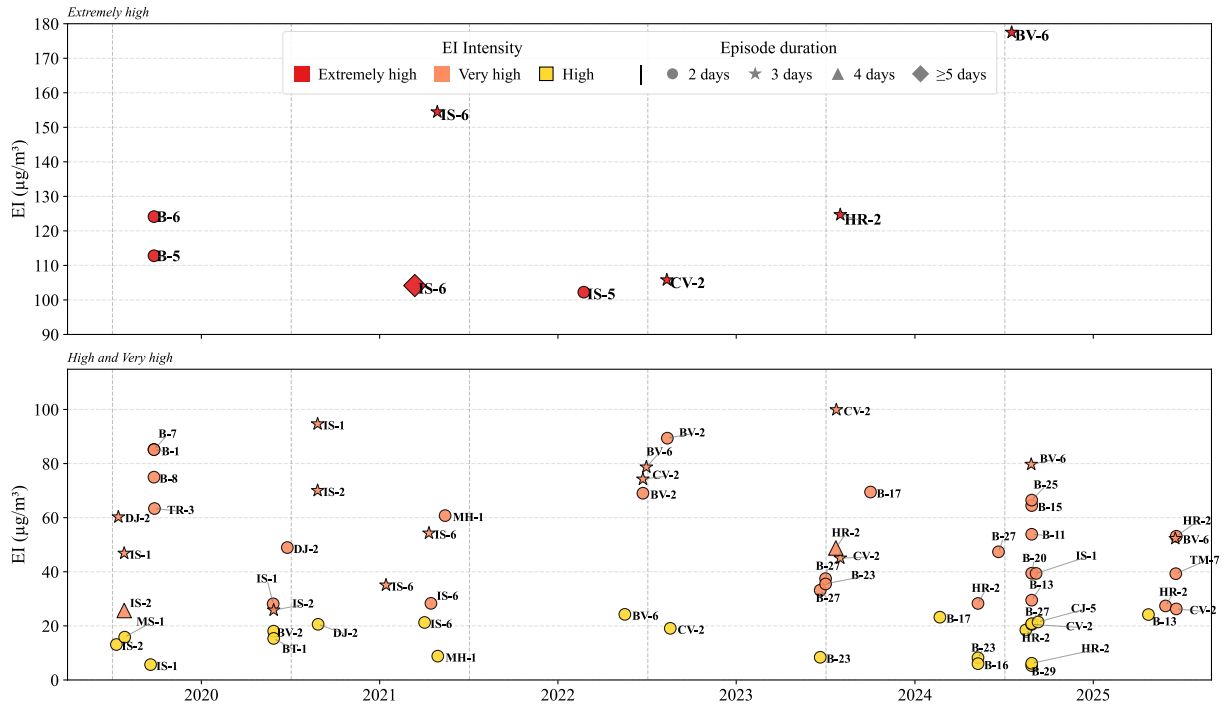


Fig. 4. 24. Temporal distribution of PM_{10} pollution episodes characterized by the EI indicator in national monitoring stations, 2020-2025.

The analysis of $PM_{2.5}$ episodes identified substantially more *Extremely High* events than for PM_{10} , with the most pronounced episodes occurring during the cold season. The most severe situation was recorded at MM-6 (Maramureş), where a 14-day episode in December 2024 reached an EI of $596.43 \mu\text{g}/\text{m}^3$. Other significant episodes were recorded at stations in central Romania (BV-2, BV-6, CV-2, HR-2), as well as at GJ-1, AR-2, IS-2, and several Bucharest stations.

Very High episodes were considerably more frequent and occurred across a broad geographical area, with the highest frequencies recorded at IS-2, GJ-1, MM-6, BV-2, and BV-6. These recurrent episodes, generally lasting several days and occurring predominantly during winter, indicate a regional pattern of $PM_{2.5}$ accumulation rather than isolated local events. In Bucharest, the concentration of extreme episodes during recent winters suggests the combined influence of unfavorable atmospheric dispersion conditions and urban emission sources, particularly residential heating and traffic.

Overall, the results indicate a widespread seasonal pattern of severe $PM_{2.5}$ pollution across several Romanian urban areas. The increasing number of episodes observed in 2023-2025 should, however, be interpreted with caution, as it is partly influenced by improved data coverage and more continuous monitoring. The recurrent *High*, *Very High*, and *Extremely High* episodes highlight the need for both long-term emission reduction measures and short-term episode-management actions, including traffic restrictions, limitations on solid-fuel combustion, and health warnings for vulnerable population groups (Fig. 4. 25).

4.6.3. Toxic Load Index (*TLI*) at Air Quality Monitoring Stations in Romania

TLI is defined as a daily indicator calculated by multiplying the daily $\text{PM}_{2.5}$ concentration by the daily limit value of $25 \mu\text{g}/\text{m}^3$ by the $\text{PM}_{2.5}/\text{PM}_{10}$ ratio. This simultaneously captures the severity of the exceedance and provides an indirect estimate of the toxicological potential associated with fine particles (see Section 3.4.3).

The distribution of days across *TLI* categories in 2025 shows that the majority of *Moderate-Very High* *TLI* episodes are recorded at urban and traffic monitoring stations in major metropolitan areas, particularly Bucharest and Cluj. The CJ-5 urban station recorded 87 days with a *TLI* of at least 5, followed by several Bucharest stations (B-27, BV-6, B-15, B-17, B-11 and B-25) with 50-60 days in the *Moderate*, *High* or *Very High* categories. This indicates a high frequency of repeated episodes involving potentially toxic fine particulate matter. In contrast, rural and suburban stations generally have fewer such episodes. IS-6 (rural) recorded 27 days with a *TLI* of at least 5, B-8 (rural), only seven days; and B-7 and IS-4 (suburban/rural) each recorded five days. This indicates a lower frequency of episodes with a high *TLI*.

Several urban and traffic monitoring stations in Bucharest and Cluj reach *TLI* maximums of over $80\text{--}95 \mu\text{g}/\text{m}^3$, while rural and suburban stations generally remain below $40 \mu\text{g}/\text{m}^3$. Of the urban stations with high *TLI* values, the DJ-7 urban background station stands out, with 47 days falling into the moderate to very high categories, a figure comparable to some urban and traffic stations in Bucharest. The high frequency of episodes with *TLI* values of at least 5 suggests that, in addition to large metropolitan areas, there are also smaller urban centres where fine particulate matter significantly contributes to the toxicological load of ambient air.

Furthermore, data analysis indicates that high *TLI* values generally coincide with episodes identified by *EI*. This suggests that periods of greater duration and intensity tend to be associated with increased toxicological potential. A notable example is the episode that occurred in Bucharest on 25-26 February 2025.

Episodes classified as *Moderate* to *Very High* occur during the colder months, which indicates a significant contribution from residential heating, traffic and meteorological conditions that hinder the dispersion of air pollutants.

The results suggest that air quality management measures should not be based exclusively on urban or regional averages, but rather on identifying stations that repeatedly record high *TLI* values. The recurrence of significant episodes at stations such as CJ-5, B-27, BV-6, B-15 and B-17 indicates the need for further investigation into local emission sources, dispersion conditions and land use characteristics in the vicinity of these sites.

The results of this chapter confirm that *EC*, *EI* and *TLI*, when applied at the level of the national monitoring network, can identify high-risk stations and the spatial patterns of particulate matter pollution. This serves as a complementary tool to support air quality management measures, in line with the requirements of Directive (EU) 2024/2881.

Conclusions

The research highlights the complex nature of air pollution caused by PM₁₀ and PM_{2.5} particulate matter in north-eastern Romania, emphasising the need for an integrated approach to spatial and temporal variations, emission sources, meteorological conditions and current and future air quality requirements.

An analysis of PM₁₀ and PM_{2.5} concentrations from 2017 to 2024 at monitoring stations in the Suceava, Iași and Botoșani municipalities revealed significant variability over time and space. The highest concentrations were predominantly recorded during the cold season under conditions favourable to pollutant accumulation. Diurnal cycles generally exhibited a bimodal pattern with peaks in the morning and evening. The results of the Mann-Kendall and Sen slope tests indicated significant downward trends for certain stations and periods, while in other cases, variations mainly reflected interannual fluctuations.

An analysis of the relationships between PM₁₀, PM_{2.5} and gaseous pollutants revealed stronger correlations between particulate matter and NO₂, CO and benzene during the cold season, indicating a significant contribution from combustion processes, particularly from road traffic and residential heating. During the warm season, weaker correlations between particulate matter and gaseous pollutants associated with combustion processes suggest a relatively greater contribution from atmospheric transformation processes and particle resuspension. The results of the regression models confirm the combined influence of emission sources and meteorological conditions on PM levels.

Analysis of the PM_{2.5}/PM₁₀ ratio revealed spatial and seasonal variations in particle composition. High values during the cold season, particularly in Suceava, indicated the importance of anthropogenic sources, including residential heating and biomass burning. During the warm season, however, the relative contribution of coarse particles from natural processes and resuspension increased. This indicator was useful for further characterising the type of pollution and the variation in the contribution of the fine fraction to the total particulate matter.

An evaluation of satellite estimates and CAMS models against in situ measurements in Iași showed that, while the CHIMERE and Ensemble models reproduce the general trends and seasonal variability in concentrations, they tend to underestimate the observed values. The results demonstrated varying performance depending on the particle fraction analysed, supporting the use of a combination of models and data from the national air quality monitoring network.

An analysis of PM₁₀ and PM_{2.5} concentrations in relation to current limit values for protecting public health indicates compliance with these limits in recent years. However, when these concentrations are compared to the stricter requirements set out in Directive (EU) 2024/2881, which will apply from 1 January 2030, it is evident that additional challenges will need to be overcome to achieve air quality objectives. In this context, achieving compliance with future limit values will require further reductions in particulate matter emissions and their precursors, necessitating integrated measures that target both local sources and regional contributions.

An original contribution of this research is the definition, substantiation, and application of three additional indicators for characterizing particulate matter exposure: Concentration Excess (*EC*), Episode Index (*EI*), and Toxic Load Index (*TLI*). These indicators complement the standard compliance metrics by providing information on the magnitude of exceedances, the intensity of pollution episodes, and the contribution of the fine PM_{2.5} fraction to the total PM₁₀ mass.

Applying these indicators at the local level and subsequently expanding them to the entire national monitoring network has demonstrated their usefulness in identifying areas and stations characterised by prolonged exposure, intense pollution episodes, or an increased risk due to a high proportion of PM_{2.5}. The results confirm that pollution assessment cannot be fully characterised by limit value exceedances alone: the magnitude, duration and composition of particle fractions must also be considered.

This study shows that combining the analysis of measured concentrations and trends with data on emission sources, weather conditions, the composition of particle fractions and the *EC*, *EI* and *TLI* indicators offers a thorough approach to evaluating exposure to particulate matter. The proposed indicators can serve as complementary tools for air quality management and for informing measures tailored to local conditions, in light of the requirements established by Directive (EU) 2024/2881.

The results emphasise the importance of reducing emissions, particularly from residential heating and road traffic, and of implementing specific measures to manage periods of high particulate matter concentrations.

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