Evaluation of air pollution by particulate matter PM10 in the NE region of Romania

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ABSTRACT: PM10 particulate matter in the ambient air is a complex combination of very small particles and liquid drops. Exposure to pollution caused by such particulate matter represents a threat to public health. The analysis hereafter shows the monthly and yearly evolution and dynamics of PM10 suspended particulate matter concentration over the period of time between years 2015 and 2019 and provides quantitative, real and precise data relating to the space-time shifting in the amount of air pollution caused by PM10 suspended particulate matter as well as the quality of ambient air. With a few exceptions, the multiannual concentrations of particulate matter measured at the EPA stations in the NE region of Romania fell within legally accepted limits (under 40 μg/m^3). The highest multiannual averages were calculated at traffic-aimed and industrial stations, as follows: Iași City 1 (40.90 μg/m^3), Iași City 6 (34.51 μg/m^3), Iași City 2 (34.51 μg/m^3), Iași City 5 (31.28 μg/m^3) and Suceava City 2 (30.87 μg/m^3). The lowest multiannual average was recorded at Poiana Stampei station, which is a background type station (16.70 μg/m^3). The highest monthly averages of air pollution caused by PM10 occurred over the cold season, whereas the lowest averages were calculated during the summer season. The monthly averages indicating maximum values of PM10 concentration (over 40 μg/m^3) were collected at traffic-aimed or urban stations. The highest daily levels of suspended particulate matter PM10 concentration were recorded at IS1 (193.83 μg/m^3 – November 5th, 2015), IS5 (182.76 μg/m^3 – November 5th, 2015) and IS6 (170.38 μg/m^3 – October 19th, 2018). The administration and the population must adopt and follow practical measures to limit air pollution caused by PM10 suspended particulate matter.

KEY WORDS: PM10, quality of ambient air, the NE region of Romania.

1. Introduction

The suspended particulate matter in the ambient air consists of a complex combination of very small particles and liquid drops. PM10 are fine particles measuring 10 micrometres in diametre or even less. The majority of particles are formed in the atmosphere as a result of complex reactions between chemical substances (COx, NOx, SOx), which are pollutants released by power plants, industries and vehicles. Others are emitted by sources such as construction sites, unpaved roads, fields, smoke or fires. The concentrations of particulate matter less than 10 microns in diametre in the ambient air are worked out by use of the annual limit value (40 µg/m^3) or the daily limit value (50 µg /m^3) set up for the protection of human health (Law 104/2011).

To begin with, the impact of low-height land sources of emission associated with residential heating activities is noticeable on the variation of PM10 suspended particulate matter concentration. The daily concentration averages of PM10 suspended particulate matter are also direcly influenced by weather conditions, such as wind direction and speed, rainfall, air temperature, atmospheric pressure as well as by area-specific geographical factors applying to the location of the monitoring station in question.

There is a large number of studies regarding this issue. The research carried out in the case of the European continent has come to prove the relationship between weather factors and changes in PM10 concentration, especially in the cities (Houthuijs et al., 2001; Chaloulakou et al., 2003; Grivas et al., 2004; Ferrario et al., 2008; Rost et al., 2009; Galindo et al., 2011; Unal et al., 2011; Bielawska et al., 2014; Fortelli et al., 2016; Largeron et al., 2016).

Similarly, in the case of Romania there are studies which have analysed the role of the meteorological parameters in correlation with the PM10 numerical values in our country (Maco et al., 2016). The variation in PM10 particulate matter concentration at regional level has been pinpointed in numerous articles. In the west of Romania, maximum levels of concentrations were reported in Arad County, as a result of the high amount of traffic (ascribed to a large number of border checkpoints and major drive routes) (Bodescu and Copolovici, 2018). The quality of air was better in spring and summer in the SE of Romania, due to an increase in air temperature levels in the summer and relative moisture variability and rainfall (Bălănică et al., 2019). Quite an interesting study takes into account the influence of the potential evapotranspiration (PET – Potential Evapotranspiration) and the climatic water balance (CWB - Climatic Water Balance) on the shifts in PM10 concentration in the Ciuc water catchment area (Reka Boga et al., 2019). There is a negative correlation between the particulate matter concentration, the potential evapotranspiration and rainfall and it is possible to apply these findings to other regions in Romania (river valley corridors, natural or man-made lakes, etc.). Furthermore, a large number of studies have shown data regarding air quality and PM10 concentration levels in Romania’s urban areas – Bucharest, Cluj Napoca, Târgu Jiu, Craiova, Iași (Mateescu et al., 2010; Proorocu et al., 2014; Căpățână et al., 2016; Carmen et al., 2016; Bodor et al., 2020). These highlighted not only the impact of weather parameters (wind direction and speed, relative moisture, air temperature), but also the differentiations in PM10 distribution in urban areas as opposed to the surrounding rural ones. As a result, extreme values of PM10 concentrations were reported in the sheltered urban areas with jammed traffic (both annual and daily allowed value limits were exceeded), whereas in the surrounding suburban areas lower PM10 concentration values were reported.

In the case of the NE Region of Romania, the research carried out so far refers to the influence of meteorological conditions on PM10 concentration changes in crowded urban areas – Iași, Piatra Neamț, Suceava, Botoșani (Lazurcă, 2015; Czernecki et al., 2016; Roșu et al., 2020; Mihăilă et al., 2020). The study on the metropolitan area of Iași was aimed at evaluating the role of meteorological conditions and local factors in correlation with the time-space PM10 particulate matter variability. Moreover, the connection between thermal inversions and the pollution levels applying to the urban area of Iaşi was highlighted (Sfîcă et al., 2018). Concerning Suceava City, the studies have shown the influence of meteorological factors (temperature, rainfall, humidity, atmospheric pressure, wind direction and speed) on the distribution of PM10 concentration and air quality. The highest levels of PM10 were recorded in the cold season, in cloudy weather and in thermal inversion situations, as well as in places directly affected by intense industrial activity

(Lazurcă, 2015; Mihăilă et al., 2020). The National Environmental Protection Agency also had a major contribution to research (by means of the Reports with regard to the ambient air quality in the counties in the NE Region of Romania for years 2015 – 2019 as well as the Plans to maintain air quality in the hereabove counties). These documents contain measures leading to maintaining and/or improving air quality in the areas under study.

The aim of our study is to analyse the space-time distribution of PM10 concentration in the NE Region of Romania and to identify the areas where the quality of air must be maintained or improved. The herein study ***objectives*** aim at:

- analysing PM10 particulate matter concentrations over the time period between 2015 and 2019 in the NE Region of Romania;
- highlighting connections between meteorological factors (temperature, rainfall, humidity, atmospheric pressure, wind) and the space-time distribution of PM10 concentrations;
- identifying the share of PM10 pollution into the general pollution in the area under study.

2. Study area

The study area, namely the NE Region of Romania comprises 6 counties: Suceava, Botoșani, Iași, Neamț, Bacău, and Vaslui. The total area of the region is 36850 km^2.

The relief is varied, and all the landforms and elevation levels known in Romania are found here. In the counties of Suceava, Neamț and Bacău there are mountain subunits belonging to the Oriental Carpathians as well as subunits belonging to Moldova's Sub-Carpathians. All six counties cover Moldova Plateau subunits. In the counties of Botoșani, Iași and Vaslui there are territories belonging to Moldova Plain and the Prut River Floodplain, and the counties of Suceava, Neamț, Iași and Bacău are crossed by the corridors of the Moldova and Siret Rivers. The climate is closely related to the relief (the annual average temperature ranges between 0.5°C in the Călimani Mountains and 9.8°C in Bârlad City, and the annual rainfall ranges between 1000 - 1200 mm in the mountain area and 400-500 mm in the low plain area).

The population in the region is 3959219 people (as of July 1st, 2018) and is mainly located in rural areas. The largest cities in the region are Iași (376180 people), Bacău (197386 people), Suceava (124161 people), Botoșani (120535 people), Vaslui (117465 people) and Piatra Neamț (113164 people). The public road density is 36.3 km / km^2 and higher in Iași, Bacău and Suceava, which are counties crossed by a number of European roads (E85, E576, E583, E574).

3. Methods

This analysis captures the annual and multiannual dynamics of the degree of air pollution caused by PM10 suspended particulate matter in the NE Region of Romania over the period 2015 – 2019. We identified the maximum daily values of PM10 concentration per each place and year within the period under study and analysed the daily air quality values relative to the PM10 concentrations from a time-space perspective. Our study uses daily, monthly and annual data collected at the EPA stations in the investigated area, which were organised in tables and graphically represented by stations from the region's north to south and west to east in order to follow and highlight the space distribution in latitude and longitude of PM10 particulate matter.

3.1. Research Methods

Two methods are used to determine the amount of PM10 which makes the dimensional fraction of toxicological concern in the urban ambient air:

- the gravimetric method GRAV, which is the reference method;

- the nephelometric method LSPM (automated measurement) – is used for information purposes, and exceeding records may be confirmed or discarded by ulterior measurement results obtained by analysis by means of gravimetric method.

The gravimetric method is used in analytical chemistry to determine the quantity of an analyte within a sample considering its mass. The principle of gravimetric analysis consists of determining the mass of a compound derived from the sample analyte (by means of weighing), and the analyte mass can be established based on the chemical reactions which led to the compound. The nephelometric method represents a procedure for analysis consisting of measuring the concentration of an emulsion by comparing its transparency to a standard preparation. The validation of these values is confirmed after processing the data found by means of gravimetric method. This is the reason why the gravimetric method is the reference method in conformity with the national and European law and also why the herein study takes into consideration data acquired in such way. Only in cases in which data collected by such method were not available did we consider data obtained by means of the nephelometric method.

The reference method for sampling and measuring PM10 is stated in the EN 12341 Standard «Ambient air. Standardized method for gravimetric measurement to determine the mass fraction of PM10 or PM2.5 of suspended particulate matter».

Our research methods are as follows:

- collecting various information, including reading scientific articles and papers on the topic and region under study;
- analysing, and comparatively interpreting statistical data based on their causal distributive and evolutionary components;
- representing graphically the PM10 concentration values and interpreting the resulting graphical and cartographic material.

The use of such methods is necessary in the analysis of suspended PM10 concentration, as it provides quantitative, real and exact data regarding the shifting over time and the space distribution of ambient air quality in relationship with their causality.

3.2. Means

To do this, different devices to measure the particulate matter concentration are used. The LSV3 device can be used both in the open air and for indoor measurements. The particulate matter caught on filter can be gravimetrically analysed not only to determine the concentration values but also the composition by means of physico-chemical lab investigation. The Dust Trak Aerosol Monitor analyser is a laser photometer which provides real-time readings of mass concentrations of aerosols and their size fractions. The SkyPost PM FG device is an automated system for sampling particulate matter from the ambient air which ensures automated continuous sampling around the clock.

The County Environment Protection Agencies have labs equipped with a number of devices for airborne pollutants concentration, quantity and level measurement purposes. The use of atomic absorption spectrometry can establish the presence of heavy metals in various environmental samples: vegetation, water, ground, waste, dust (PM10, PM2.5, total matter). Both atomic absorption spectrometres (VarianSpectrAA220 and Perkin – Elmer) are used to determine the presence of heavy metals (Pb, Cd, As, Ni, Cr, Mn, Fe) in the suspended particulate matter. The climate lab serves to create or maintain constant temperature and moisture conditions in order to ensure conditioning of filters used to determine the amount of PM10 by means of gravimetric method. The Berghof system is used for PM10 mineralisation of filters to detect heavy metals.

3.3. Air Quality Monitoring Network – The North- East Region

Law No. 104 of 2011 which transposes the Directive 2008/50/CE provisions regarding the ambient air quality and cleaner air for Europe into the national law refers to the initiation of the national air quality monitoring network. According to Law No. 104/2011 regarding ambient air quality, the sampling points for the purpose of human health protection must be located so as to provide the following data:

- the areas within zones and agglomerations with the highest concentrations to which the population is susceptible to be exposed directly or indirectly over a significant time period in relation to periods of average values / limit values;
- levels in other areas within the zones and agglomerations of interest for determining the exposure level of the population;
- residue that represents the indirect exposure of population via the food chain.

The air quality monitoring system in Moldova (illustrated in Table 1 and Fig. 1**)** comprises 19 stations for the evaluation of various type sources and their influence over air quality, as well as background level evaluation away from any source. More specifically, in Moldova there are 2 traffic-aimed stations (SV3, IS1), 6 industrial stations (SV2, IS3, NT2, NT3, BC2, BC3), 7 urban stations (BT1, SV1, IS2, NT1, VS2, VS1, BC1), 1 suburban-type station (IS5), 1 rural station (IS4), 1 urban/traffic station (IS6) and 1 EMEP station (EM3), located from N to S and W to E.

Table 1 Location of stations in the regional air quality monitoring network – the NE region of Romania (2015 – 2019).

Station Name	Type	Address	Latitude	Longitude	Altitude
SV 3 – Siret	Traffic	Siret, Str. Alexandru cel Bun	47°57’21” N	26°04’06” E	302 m
BT 1 – Botoșani 1	Urban	Botoșani, B-dul M. Eminescu, nr. 44	47°44’27” N	26°39’31” E	168 m
SV 2 – Suceava 2	Industrial	Suceava, Str. Tineretului, FN	47°40’09” N	26°16’53” E	289 m
SV 1 – Suceava 1	Urban	Suceava, Str. Mărășești, nr. 57	47°38’58” N	26°14’59” E	373 m
EM 3 – Poiana Stampei	EMEP	Poiana Stampei	47°19’28” N	25°08’03” E	910 m
IS 6 – Bosia Ungheni	Urban/Traffic	Ungheni, sat Bosia	47°12’56” N	27°46’07” E	34 m
IS 4 – Aroneanu	Rural	Aroneanu, sat Aroneanu	47°12’47” N	27°36’39” E	186 m
IS 3 – Oancea Tătărași	Industrial	Iași, Str. Han Tătar, nr. 14	47°09’28” N	27°36’45” E	64 m
IS 1 – Podu de Piatră	Traffic	Iași, B-dul N. Iorga, FN	47°09’24” N	27°34’29” E	40 m
IS 2 – Decebal Cantemir	Urban	Iași, Aleea Decebal, nr. 10	47°09’03” N	27°34’54” E	42 m
IS 5 – Tomești	Suburban	Tomești, Str. M. Codreanu, FN	47°08’09” N	27°41’35” E	37 m
NT 2 – Roman	Industrial	Roman, Str. Ștefan cel Mare, nr. 274	46°56’53” N	26°55’04” E	207 m
NT 1 – Piatra Neamț	Urban	Piatra Neamț, Str. Valea Albă, FN	46°56’03” N	26°23’20” E	357 m
NT 3 – Tașca	Industrial	Tașca, sat Hamzoaia	46°54’11” N	26°02’15” E	558 m
VS 2 - Huși	Urban	Huși, Str. Recea, nr. 1	46°40’30” N	28°05’47” E	80 m
VS 1 – Vaslui	Urban	Vaslui, Str. Ștefan cel Mare, nr. 56	46°37’56” N	27°43’49” E	107 m
BC 1 – Bacău 1	Urban	Bacău, Str. Războieni, nr. 11	46°33’50” N	26°54’37” E	169 m
BC 2 – Bacău 2	Industrial	Bacău, Str. Izvoare, nr. 1 bis	46°33’19” N	26°55’32” E	157 m
BC 3 – Onești	Industrial	Onești, Str. Cauciucului, nr. 1	46°15’24” N	26°47’33” E	200 m

The data focused on in this study regarding PM10 refer to years 2015 to 2019 and were provided by the Environment Protection Agencies in the counties of Suceava, Botoșani, Iași, Neamț, Bacău, Vaslui. Mainly data collected at the stations using the gravimetric methods were used. In case of lack of such data, values obtained by means of nephelometric method were used instead (stations IS3 – Oancea Tătărași, NT2 – Roman, BC3 – Onești have been collecting their data exclusively via this method).

According to Annex 4 to Law 104/2011, the objective of quality in data monitoring regarding the minimum amount of data records over an average period of 1 year is 90 %. Given that the recording requirement of 90 % does not include data loss caused by calibration, checks and ongoing maintenance, data records of minimum 75 % are still considered to be consistent (Table

2). Therefore, it may be noticed that generally-speaking, in the region under study the percentage of valid data was accomplished in whole (stations Botoșani 1, Iași 1, Suceava 1 for the entire period under study) or in part (for example, stations Bacău 1, Bacău 2, Iași 2, Iași 4, Neamț 1, Suceava 2 in different years). Year 2018 was the only one within the period under study when the majority of the EPA stations provided enough data records to comply with the objectives of quality for the evaluation of ambient air quality correlated with PM10 pollutant concentrations. The majority of stations went through servicing (the industrial-type station SV2 was temporarily stopped for technical reasons from January 2014 to May 2016) or were relocated (for example, rural background station IS4 was relocated from Aleea Sadoveanu – Copou on November 5th, 2015 and reinstalled in Aroneanu Village on March 2nd, 2016).

The air quality monitoring system allows the local environment protection authorities to take quick measures to reduce or eliminate the instances of pollution, as well as to inform, to warn and to protect the population in emergency cases when the allowed PM10 concentrations are exceeded. The results obtained in the air quality monitoring at the automated stations under RNMCA in Moldova – The NE Region of Romania, over the time period between years 2015 - 2019 indicate that the levels of pollutants falling under law provisions are in general lower than the limit or target values stated in Law 104/2011, with a few exceptions, however.

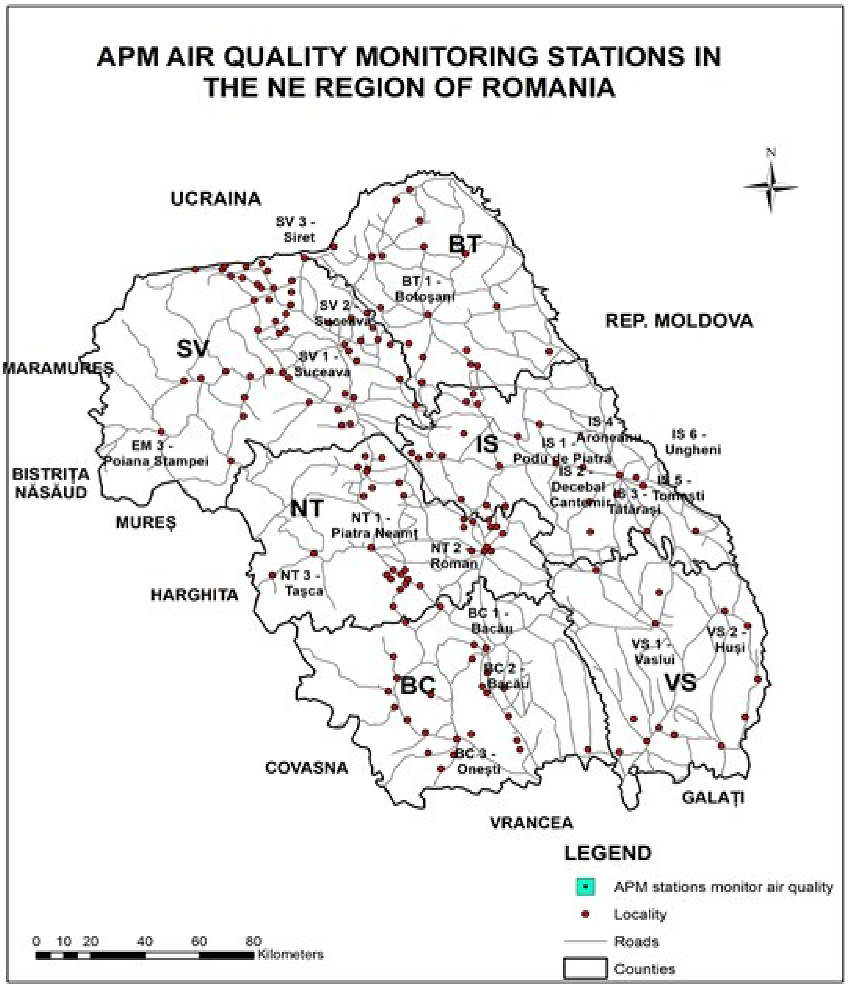

Figure 1 Location of stations in the Regional Air Quality Monitoring Network – The NE Region of Romania (2015 – 2019).

According to Order No. 598/2018 for classification in regimes of management of zones and agglomerations provisioned in Annex No 2 to Law No. 104/2011 regarding ambient air quality, the NE Region of Romania – Moldova was classified as follows:

- *management regime I* (exceeded limit values): Iași City;
- *management regime II* (limit values compliance): Counties of Bacău, Botoșani, Iași (except the City of Iași), Neamț, Suceava, Vaslui.

Table 2 Data records (%) of PM 10 concentration at EPA stations in the NE Region of Romania (2015 – 2019).

No.	Station Name	Type	Method	2015	2016	2017	2018	2019	Avg.
1.	SV 3	Traffic	Gravimetric	86.07	49.40	0	77.97	94.05	**61.50**
2.	BT 1	Urban	Gravimetric	96.39	99.14	95.66	98.60	98.61	**97.68**
3.	SV 2	Industrial	Gravimetric	0	0	94.79	97.76	97.78	**58.07**
4.	SV 1	Urban	Gravimetric	83.18	79.62	77.90	91.72	87.51	**83.99**
5.	EM 3	EMEP	Gravimetric	24.88	12.03	0	75.22	26.07	**27.64**
6.	IS 6	Traffic	Gravimetric	23.60	17.58	66.01	91.78	65.63	**52.92**
7.	IS 4	Rural	Gravimetric	72.28	53.66	80.97	87.31	87.26	**76.30**
8.	IS 3	Industrial	Nephelometric	83.86	25.54	97.83	93.01	64.01	**72.85**
9.	IS 1	Trafic	Gravimetric	90.93	75.74	93.54	98.01	91.39	**89.92**
10.	IS 2	Urban	Gravimetric	0	0	86.34	98.59	93.30	**55.65**
11.	IS 5	Suburban	Gravimetric	75.53	76.85	33.60	88.88	65.99	**68.17**
12.	NT 1	Urban	Gravimetric	48.70	41.09	94.79	96.96	84.92	**73.29**
13.	NT 2	Industrial	Nephelometric	0	42.30	94.81	94.67	98.77	**66.11**
14.	NT 3	Industrial	Gravimetric	0	41.63	49.02	92.93	98.89	**56.49**
15.	VS 2	Urban	Gravimetric	90.52	27.77	0	0	0	**23.66**
16.	VS 1	Urban	Gravimetric	0	42.39	39.13	95.61	92.13	**53.85**
17.	BC 1	Urban	Gravimetric	92.89	82.58	15.81	90.56	50.72	**66.51**
18.	BC 2	Industrial	Gravimetric	83.60	67.90	13.65	91.78	95.62	**70.51**
19.	BC 3	Industrial	Nephelometric	66.02	96.67	97.11	96.01	68.38	**84.84**

In the case of Iași City, as a result of its classification under regime I of air quality management, a Plan for air quality and further reduction of PM10 concentration in the ambient air was issued, whereas in the case of the remaining settlements in the counties of the NE region of Romania Plans for maintaining air quality and control of pollutants concentration in the ambient air were drawn up.

3.4. PM10 Pollution Sources in the NE Region of Romania

The main pollution sources generating PM10 suspended particulate matter in the counties of the region of Moldova are listed in Table 3 and Fig. 2.

Table 3 The total quantity of PM10 emissions in the counties of the NE Region of Romania (t/year).

	SUCEAVA	BOTOȘANI	IAȘI	NEAMȚ	BACĂU	VASLUI
evaluation period	2010 - 2014	2013 - 2015	2010 - 2014	2012 - 2016	2010 - 2015	2010 - 2014
stationary sources	98.39342	9.99976	41.26806	73.88608	340.28311	94.36956
mobile sources	298.83972	131.40715	192.24643	83.85200	106.41472	274.57062
land sources	5400.06559	3425.64294	5970.63048	1406.74117	1467.86261	2582.44552
TOTAL	5797.29874	3567.04984	6204.14498	1564.47925	1914.56046	2951.38570

The main *stationary sources* are: The industrial site of Bacău (SC Sofert SA Bacău, SC Letea SA Bacău), the chemical plant area of Onești (SC Chimcomplex SA Borzești, SC Rafo Onești), The industrial site of Botoșani, SC TERMICA Botoșani, The industrial site of Iași (SC Fortus SA Iași, SC Antibiotice Iași), CET I Iași, CET II Holboca Iași, the chemical plant area of Săvinești, the cement plant Tașca – Bicaz, The industrial site of Suceava (SC Ambro SA Suceava), Termica Suceava, The industrial site of Vaslui. A number of these operators either reduced or stopped their operations, however, other PM10-generating activity has been developing at the sites.

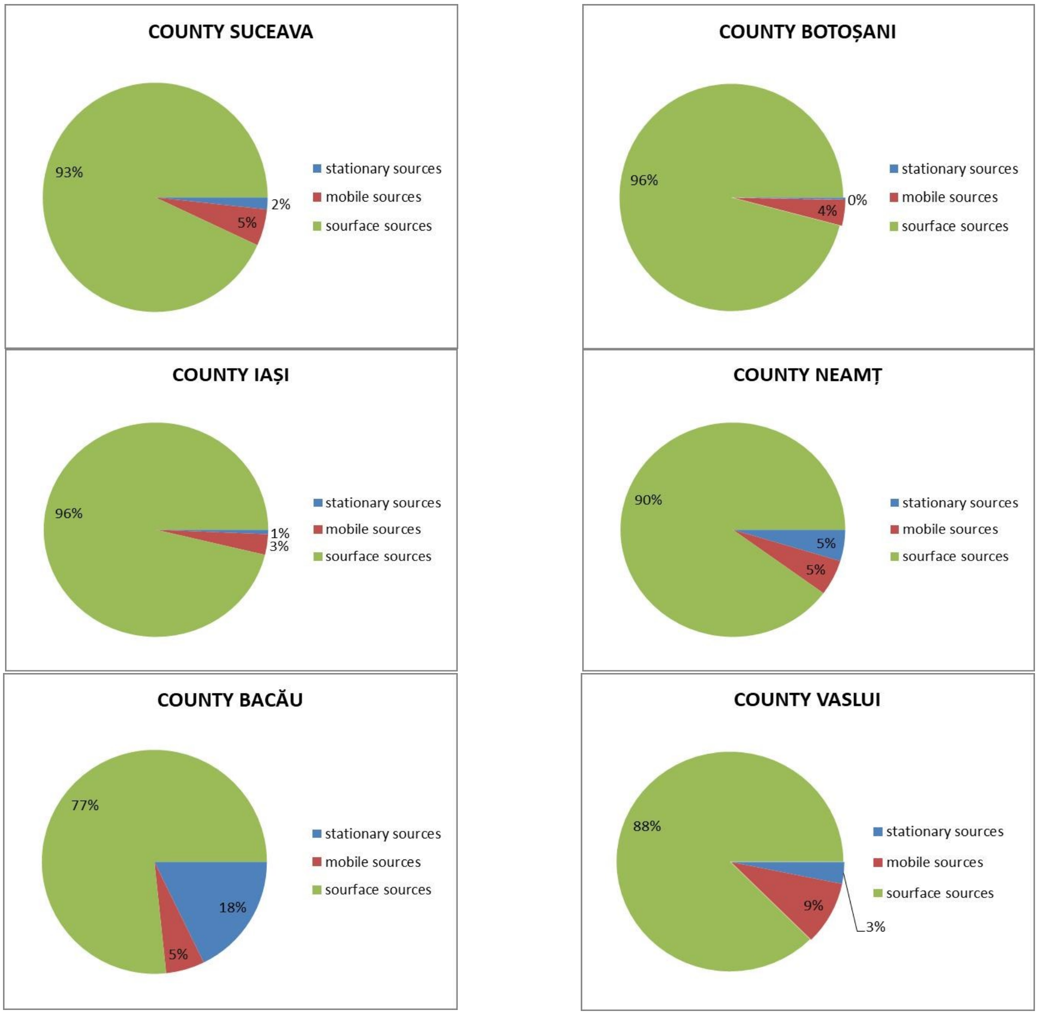

Figure 2 Pollution sources generating PM10 emissions (%) in the counties of the NE Region of Romania.

The mobile sources refer to means of transportation. The region is crossed by a number of European roads: București – Bacău – Suceava – Siret (E85), Suceava – Vatra Dornei – Dej (E58), Bacău – Brașov (E574), București – Bârlad – Albița (E581), Roman – Tg. Frumos – Iași – Sculeni (E583) as well as national and county roads. *The land sources* refer to home-use burning equipment. The shifting in the PM10 suspended particulate matter concentration reported at the beginning and the end of the year over the cold season is explained by the increased share of low height emission sources on land associated with house heating.

The impact of weather factors (still air and fog conditions lead to particulate matter build-up on ground surface, which in turn results in high pollutant concentration recordings) regarding stagnation or dispersion of PM10 particulate matter is extremely important. The inadequate technical conditions of roads (caused by lack of investment in the major road infrastructure) in the NE Region of Romania together with delayed spring cleaning activities lead to high levels of PM10 concentrations whenever the meteorological conditions are unfavourable.

4. Results and Discussion

4.1. The interannual evolution and multiannual distribution of PM10 concentration at the monitoring points in the NE Region of Romania

Concentrations of suspended particulate matter smaller than 10 microns in diametre in the ambient air are evaluated by means of the annual limit value (40 μg/m^3). The situation of annual average concentration values of PM10 recorded at the air quality monitoring stations located in the NE Region of Romania is shown in Table 4 and Figs 2 and 3.
The annual average concentration values of PM10 recorded at the EPA stations in the NE Region of Romania fell, with a few exceptions, within legally permitted boundaries (under 40 μg/m^3). The highest multiannual average (years 2015 – 2019) was recorded at Station Iași 1 (40.90 μg/m^3). This value exceeded the allowed annual limit. During the entire period under study, Station Iași 1 registered values over or very close to the allowed annual limit. Other high multiannual average values were recorded at the traffic-aimed or industrial-type stations Iași 6 (34.51 μg/m^3), Iași 2 (34.51 μg/m^3), Iași 5 (31.28 μg/m^3) and Suceava 2 (30.87 μg/m^3). In contrast, the lowest multiannual average of PM10 concentration (16.70 μg/m^3) was recorded at the cross-border station in Poiana Stampei, located at an altitude of 891 m. Other low values of PM10 concentration were collected at Neamț 1 (20.26 μg/m^3), Bacău 1 (21.88 μg/m^3), Vaslui (23.67 μg/m^3), all urban-type stations.

Table 4 Annual and multiannual average values of PM 10 concentration at the EPA stations in the NE Region of Romania (2015 – 2019).

No.	Station Name	Type	Method	2015	2016	2017	2018	2019	Avg. 2015-2019
1.	SV 3	Traffic	Gravimetric	26.97	~~22.40~~	-	31.20	31.03	**27.90**
2.	BT 1	Urban	Gravimetric	29.93	27.78	31.38	32.31	27.30	**29.74**
3.	SV 2	Industrial	Gravimetric	-	-	28.69	31.05	32.87	**30.87**
4.	SV 1	Urban	Gravimetric	25.59	21.22	22.23	23.98	22.60	**23.12**
5.	EM 3	EMEP	Gravimetric	~~20.49~~	~~14.68~~	-	16.25	~~15.37~~	**16.70**
6.	IS 6	Traffic	Gravimetric	~~24.14~~	~~37.16~~	~~31.55~~	42.42	~~37.28~~	**34.51**
7.	IS 4	Rural	Gravimetric	~~20.94~~	~~23.06~~	22.17	24.19	20.22	**22.12**
8.	IS 3	Industrial	Nephelometric	27.58	~~30.10~~	21.06	25.91	~~22.82~~	**25.49**
9.	IS 1	Traffic	Gravimetric	39.22	39.06	43.87	45.01	37.32	**40.90**
10.	IS 2	Urban	Gravimetric	-	-	34.15	37.29	32.10	**34.51**
11.	IS 5	Suburban	Gravimetric	32.11	29.08	~~37.19~~	31.22	~~26.79~~	**31.28**
12.	NT 2	Industrial	Nephelometric	-	~~16.79~~	29.06	20.89	19.73	**21.62**
13.	NT 1	Urban	Gravimetric	~~13.22~~	~~20.05~~	24.35	23.47	20.19	**20.26**
14.	NT 3	Industrial	Gravimetric	-	~~24.60~~	~~35.86~~	26.44	25.66	**28.14**
15.	VS 2	Urban	Gravimetric	26.31	~~25.39~~	-	-	-	**25.85**
16.	VS 1	Urban	Gravimetric	-	~~19.40~~	~~25.24~~	24.95	25.08	**23.67**
17.	BC 1	Urban	Gravimetric	27.85	19.38	~~21.34~~	23.88	~~16.95~~	**21.88**
18.	BC 2	Industrial	Gravimetric	31.20	~~22.12~~	~~25.73~~	25.04	22.75	**25.37**
19.	BC 3	Industrial	Nephelometric	~~28.74~~	25.69	18.38	23.71	~~14.66~~	**22.24**

* *~~28.36~~ – annual value inconsistent with valid data recordings*
* *-- missing data caused by servicing or malfunctioning of data collecting devices*

It is obvious (Fig. 3 and Table 4) that the highest annual averages (including those exceeding the allowed annual limit) were recorded at the stations in the County and the City of Iași, both largely populated, with developed economy and infrastructure, intense road and railway traffic. The county also makes country border. In consequence, Iași City is subject to the Plan for air quality and further reduction of PM10 concentration in the ambient air.

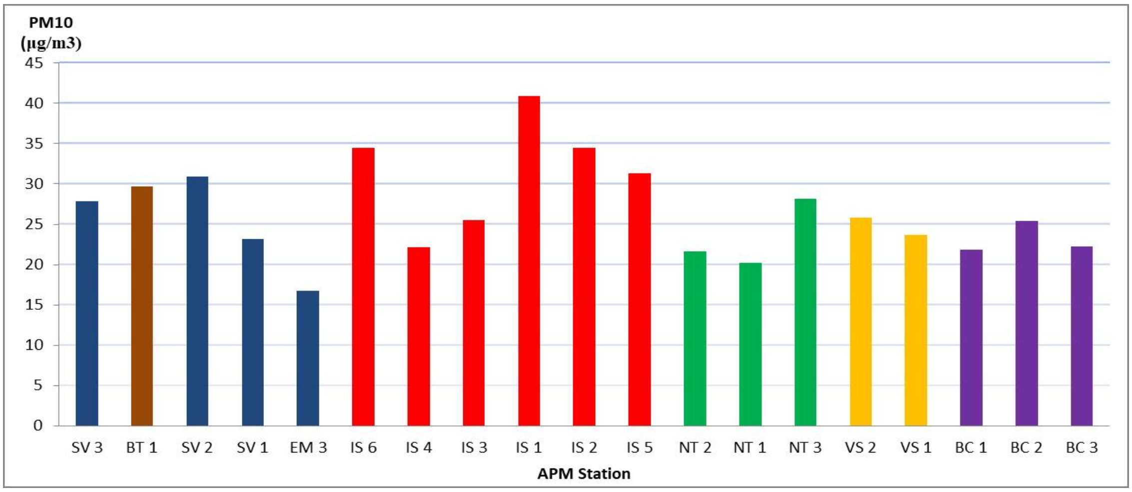

Figure 3 Multiannual average values of PM 10 concentration at the EPA stations in the NE Region of Romania (2015 – 2019).

The annual limit value (40 µg/m^3) was exceeded at stations Iași 1 (years 2017 and 2018) and Iași 6 (year 2018). Both stations are traffic-type.

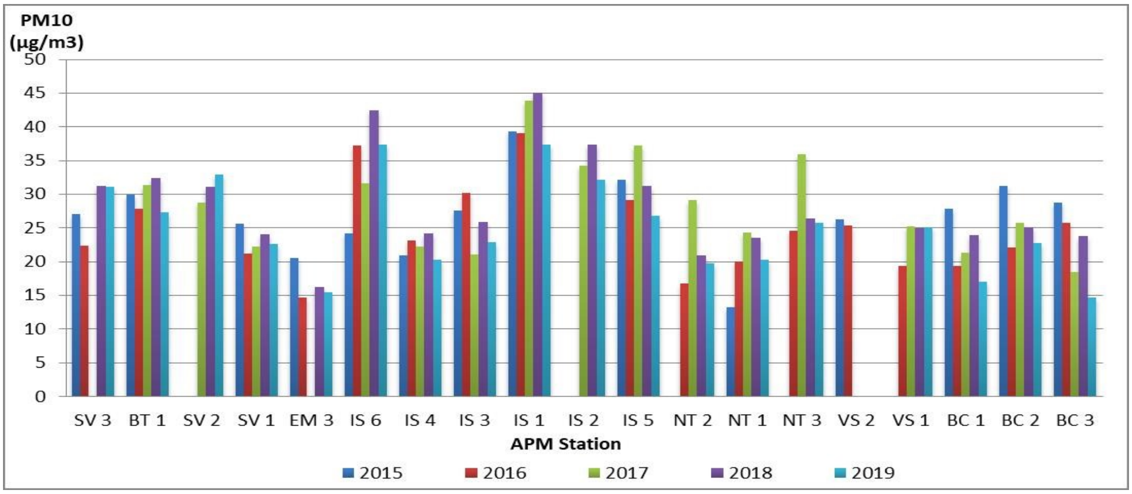

Figure 4 Multiannual values of PM 10 concentrations at the EPA stations in the NE Region of Romania (2015 – 2019).

According to statistics (Table 4) and graphical representations (Fig. 4), the year with the highest averages PM10 concentration ever recorded and calculated over the given period of time was 2018, whereas the year averaging the least was 2015 (notice that many stations were under maintenance, malfunctioning or out of order).

4.2. The annual evolution and distribution (as per monthly average values) of PM10 concentration at the monitoring points in the NE Region of Romania

In the NE Region of Romania, over the period of 2015 – 2019, the highest monthly averages of PM10 concentrations were recorded during the cold season months (October, November, February, March), whereas the lowest averages resulted in the spring and summer months (May to August) (Table 5 and Fig.5).

Table 5 Monthly average values of PM 10 concentration at the EPA stations in the NE Region of Romania (2015 – 2019).

No.	Station Name	I	II	III	IV	V	VI	VII	VIII	IX	X	XI	XII
1.	SV 3	26.4	31.6	30.1	23.5	22.7	23.3	25.0	31.3	28.9	37.4	31.7	32.4
2.	BT 1	31.9	30.4	31.5	27.9	23.9	26.7	25.5	32.2	30.4	29.6	34.0	33.7
3.	SV 2	39.3	40.1	33.3	21.9	18.6	20.3	25.1	30.2	28.9	40.6	33.3	38.9
4.	SV 1	23.7	24.4	23.6	18.7	19.1	20.1	22.0	26.2	25.6	28.8	25.0	21.5
5.	EM 3	25.2	17.8	22.6	13.8	14.0	12.6	17.7	15.6	14.6	14.2	14.6	15.0
6.	IS 6	33.3	30.0	39.9	32.0	34.2	33.5	35.9	45.0	35.8	47.3	33.8	29.2
7.	IS 4	22.5	24.4	21.6	20.1	19.9	19.4	21.6	26.0	23.8	23.1	19.2	20.9
8.	IS 3	27.4	22.8	20.0	13.6	16.1	17.3	19.6	24.5	27.1	37.9	37.5	31.6
9.	IS 1	40.9	44.5	48.6	40.4	37.5	35.8	35.3	42.4	41.7	44.8	41.3	37.7
10.	IS 2	36.4	39.3	41.1	33.6	32.4	29.1	27.5	34.4	32.2	41.2	32.4	36.0
11.	IS 5	35.6	33.1	32.0	25.2	23.5	24.1	26.4	31.9	33.5	32.7	33.7	39.8
12.	NT 2	33.0	32.7	23.4	23.6	9.2	14.5	16.4	17.1	16.2	22.7	28.8	31.0
13.	NT 1	38.1	35.6	26.4	15.8	13.7	16.2	12.9	14.3	17.2	20.7	28.7	23.8
14.	NT 3	44.8	36.4	28.1	20.2	16.6	15.4	13.8	17.4	22.8	28.4	37.5	34.4
15.	VS 2	28.2	22.1	22.8	23.3	27.9	25.0	29.4	27.9	31.2	27.8	26.2	31.8
16.	VS 1	34.1	22.6	24.6	18.1	17.0	18.1	25.3	22.5	19.2	28.6	31.5	33.2
17.	BC 1	26.8	25.7	24.1	23.9	17.8	16.8	17.7	20.5	21.2	22.3	25.0	36.3
18.	BC 2	31.5	26.8	24.5	26.7	19.1	17.6	21.2	23.9	25.5	23.3	28.4	33.9
19.	BC 3	28.2	30.9	22.2	14.7	13.6	13.5	15.7	21.5	21.5	30.2	26.3	35.2

The industrial-type EPA stations exhibited a relatively uniform evolution of PM10 concentration values over the year, recording their maximum in the month of October (approximately 40 $\mu g/m^3$) and their minimum during the months of May through July. The urban and traffic-type EPA stations exhibited a more fluctuant evolution of PM10 values over the year, with maximum values in March and October to November and minimum values during the summer months. At the rural-type EPA station (IS4) the montly averages recorded and calculated varied little throughout the year, falling between 20 – 25 $\mu g/m^3$; a similar situation occurred at the EMEP-type EPA station, only that the monthly average values of PM10 concentration were lower over an average year, resulting in a multiannual average of only 15 $\mu g/m^3$. The suburban-type EPA station (IS5) exhibited high monthly averages throughout the year (over 25 $\mu g/m^3$), with an annual maximum recorded in December (40 $\mu g/m^3$).

Maximum monthly values of PM10 concentration (over 40 $\mu g/m^3$) were recorded at IS1 (all year through), SV3 (February, October), IS6 (August, October), IS2 (March, October), SV2 (January, February, November). The above mentioned stations are either traffic or urban-type.

The main causes for these high PM10 concentrations are: car traffic (namely the emissions generated by traffic of cars transiting the main city sectors on the main roads, outside cities along national roads and in the settlements located alongside; in addition, tyre wear upon start/stop can be mentioned as another source of PM10), residential sources (which generate combustion to produce thermal energy), and last but not least, meteorological conditions (the still air and fog conditions typical to the cold season lead to particulate matter build-up on ground surface, which in turn results in high pollutant concentrations).

Some of the lowest monthly averages of PM10 concentration (under 20 µg/m^3) were recorded and calculated at NT1 (April – September), SV1 and VS1 (April – June), BC2, BC3, IS3, NT2, NT 3 and SV2 (April – August). The lowest ever monthly averages were recorded and calculated at the station EM3 – Poiana Stampei (located in the mountains). These low value averages are mostly the result of intense atmospheric dynamics, high humidity and rainfall levels in the area, but the reduction of thermal energy consumption during the warm season is also to be taken into account.

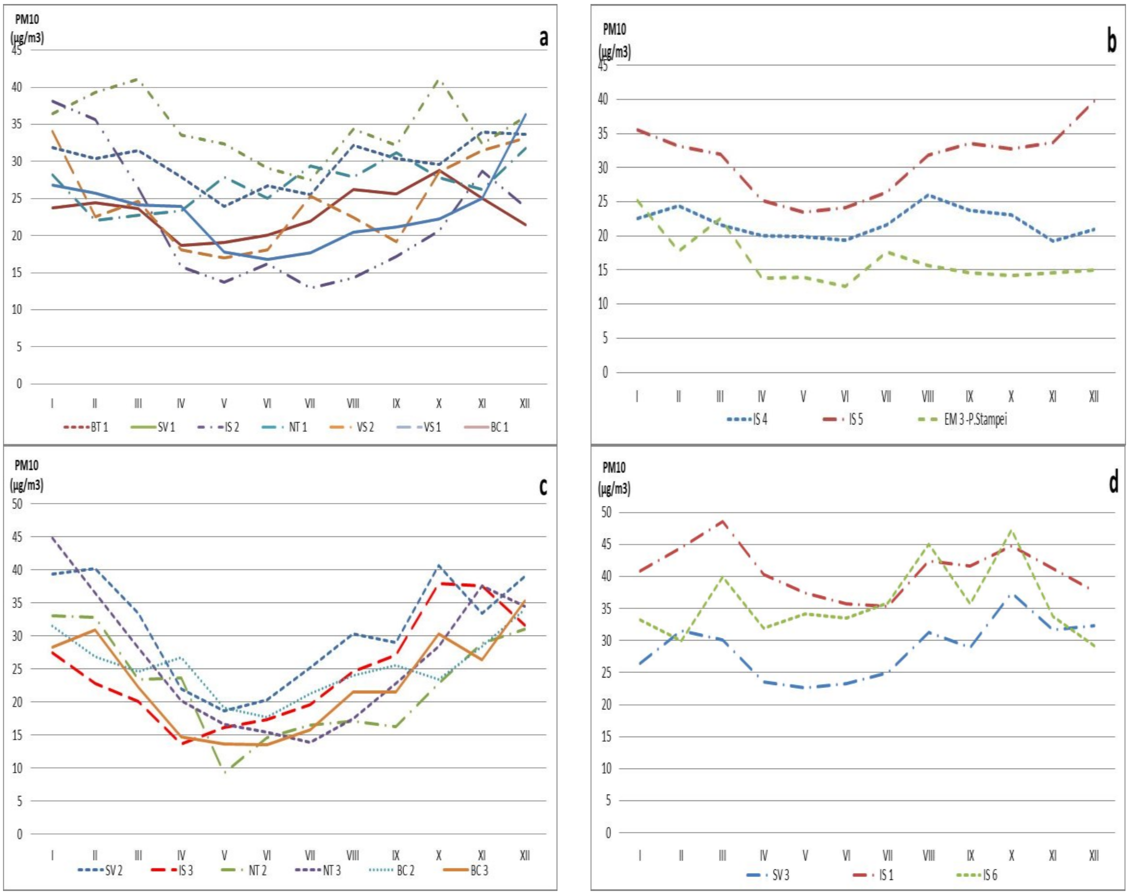

Figure 5. The annual values of PM 10 concentration at the EPA stations in the NE Region of Romania (a – urban; b – rural, suburban and EMEP; c - industrial; d - traffic) (2015 – 2019)

From a climate perspective, periods of great atmospheric stability have been detected in the NE Region of Romania. Fog is a common occurence too, often accompanied by wet frost. Furthermore, thermally-originated temperature inversions (nocturnal radiation inversions) are behind the increase in foggy days, which together with emissions from combustion lead to an increase in the PM10 concentrations in the mornings and evenings. It was noticeable that during the autumn months and at the beginning of winter (October, November and December) the population experienced persistent thermal inversions which generated high levels of PM10 concentrations in the ambient air, fuelled even more by the absence of significant rainfall. On the other hand, the role of precipitation in pollution control became obvious in the months of the cold season when the PM10 averages were very low, against the background of some important episodes of precipitation during this period. Moreover, the snow was even more helpful in cleaning away the suspended particulate matter from the ambient air.

To sum up, the particular issues relating to pollution in the region under study are also caused by a large occurence of days with still air conditions in certain areas. In other words, on such days the dispersion of pollutants is extremely low, not to mention that still air conditions are linked to lack of rainfall over long periods of time.

4.3. Maximum daily values of PM10 concentration at the monitoring points in the NE Region of Romania

Concentrations of suspended particulate matter smaller than 10 microns in diametre in the ambient air are evaluated by means of the daily limit value established gravimetrically (50 μg/m^3). This value is not to be exceeded more than 35 times/year (Table 6).

Table 6 Number of days exceeding allowed values of PM 10 concentration recorded at the EPA stations in the NE Region of Romania (2015 – 2019).

		2015		2016		2017		2018		2019		2015 - 2019	
station	Met.	No. days 50-100 μg/m^3	No. days ≥100 μg/m^3	No. days 50-100 μg/m^3	No. days ≥100 μg/m^3	No. days 50-100 μg/m^3	No. days ≥100 μg/m^3	No. days 50-100 μg/m^3	No. days ≥100 μg/m^3	No. days 50-100 μg/m^3	No. days ≥100 μg/m^3	No. days 50-100 μg/m^3	No. days ≥100 μg/m^3
SV 3	GRAV	8	-	1	-	-	-	25	-	27	-	61	-
BT 1	GRAV	25	2	17	-	21	-	25	1	20	1	108	4
SV 2	GRAV	-	-	-	-	24	2	35	3	35	1	94	6
SV 1	GRAV	8	-	1	-	1	-	9	-	7	-	26	-
EM 3	GRAV	-	-	-	-	-	-	1	-	-	-	1	-
IS 6	GRAV	5	-	10	-	23	-	89	10	50	2	177	12
IS 4	GRAV	3	-	8	-	7	-	5	1	2	-	25	1
IS 3	LSPM	18	8	10	2	12	-	31	2	8	2	79	14
IS 1	GRAV	69	4	37	3	94	5	119	7	60	1	379	20
IS 2	GRAV	-	-	-	-	41	2	67	2	41	1	149	5
IS 5	GRAV	34	5	27	-	32	3	29	-	14	1	136	9
NT 2	LSPM	-	-	4	-	24	-	8	-	9	-	45	-
NT 1	GRAV	1	-	2	-	8	-	6	1	7	2	24	3
NT 3	GRAV	-	-	8	-	19	6	18	-	20	1	65	7
VS 2	GRAV	-	-	-	-	-	-	-	-	-	-		-
VS 1	GRAV	-	-	-	-	-	-	10	-	10	-	20	-
BC 1	GRAV	12	3	3	-	-	-	8	-	-	-	23	3
BC 2	GRAV	28	7	6	-	-	-	10	-	8	-	52	7
BC 3	LSPM	13	2	19	-	12	1	22	-	2	-	68	3

The daily average concentrations of PM10 suspended particulate matter are directly influenced by weather factors: wind direction and speed, rainfall, air temperature, atmospheric pressure etc., and by the geographical factors characteristic of the area under study. The types of atmospheric thermal stratification and temperature inversions affect the particulate matter dispersion processes to a great extent. Another significant cause is the traffic of cars transiting the main city sectors on the main roads, outside cities along national roads and in the settlements located alongside. The hours when traffic in the vicinity of EPA stations gets more intense are between 07:30 – 19:00, resulting in high values of PM10 concentration being recorded. On Saturdays and Sundays the traffic gets lower, consequently lower values of PM10 concentration are recorded.

Upon analysis of the daily air quality specific rate indicated by the PM10 concentration in the air, a number of interesting comments can be made regarding the NE Region of Romania (Fig. 6). At times when the daily value of PM10 concentration ranges between 0 and 10 μg/m^3, the daily air quality specific rate is 1 (air quality is excellent), whereas when these daily values range between 10 and 20 μg/m^3, the specific rate is 2 (air quality is very good). Very good and excellent air quality (over 50 % of added values) were recorded at more than a few EPA stations in the area under study. Lower rates indicating high air quality were recorded at stations SV2 (32 %), BT1 (27 %), IS2 (23 %), which are urban or industrial-type stations. When the daily value of PM10 concentration falls between 20 and 30 µg/m^3, the daily specific rate is 3 (air quality is good), whereas values between 30 and 50 µg/m^3 trigger a daily specific rate of 4 (average air quality).

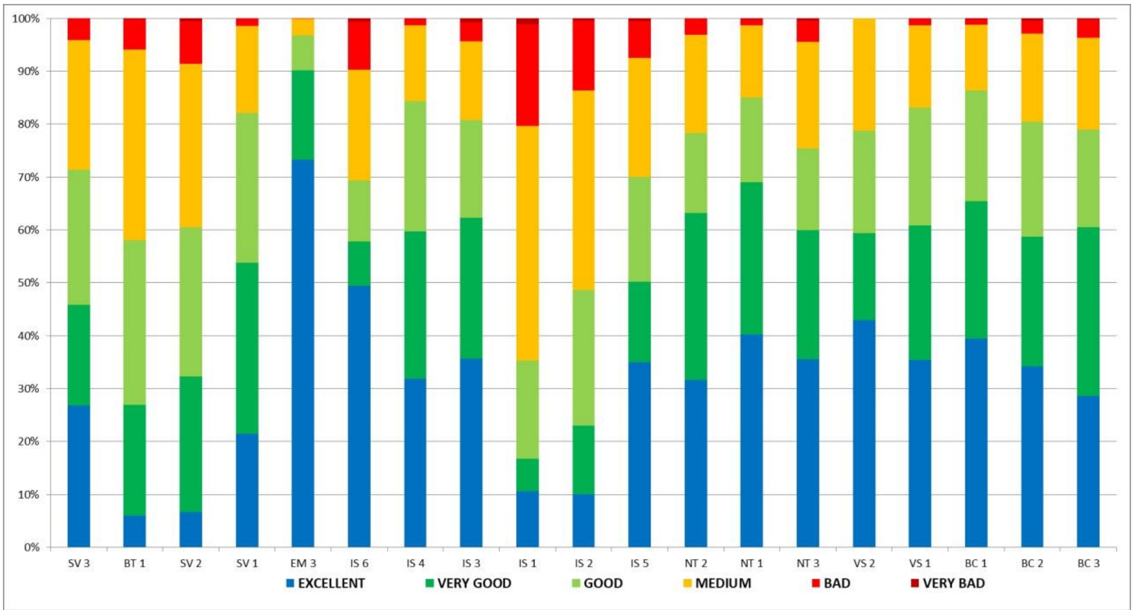

Figure 6 Total percentages of daily air quality specific rates corresponding to PM 10 concentrations in the air recorded at the EPA stations in the NE Region of Romania (2015 – 2019).

When the daily value of PM10 concentration falls between 50 and 100 µg/m^3, the quality specific rate is 5 (air quality is bad), and when the PM10 concentration exceeds 100 µg/m^3, the specific rate is 6 (air quality is very bad). Bad and very bad air quality were reported at the urban and traffic-type stations IS1 (20 %), IS2 (14 %), IS6 (10 %). The most days with PM10 concentrations over 50 µg/m^3 were recorded at IS1 – Podu de Piatră (119 days in 2018, 99 days in 2017, 61 days in 2019, 73 days in 2015 and 40 days in 2016). A significant count of days with PM10 concentrations over 50 µg/m^3 was also reported at IS6 – Bosia Ungheni (99 days in 2018 and 52 days in 2019), IS2 – Decebal Cantemir (69 days in 2018, 43 days in 2017 and 42 days in 2019) and SV2 – Suceava (38 days in 2018 and 36 days in 2019). These high concentration ranges were recorded at urban and traffic-type EPA stations. In the case of EM3 – Poiana Stampei station, there were no days with PM10 concentrations exceeding 50 µg/m^3 during years 2015 to 2019. Other similar locations exhibiting low count of days with over 50 µg/m^3 PM10 were NT 1 – Piatra Neamț, SV1 – Suceava, VS1 – Vaslui – all urban stations (located downtown) as well as the rural-type station IS4 – Copou / Aroneanu. The highest count of days with PM10 concentrations exceeding 100 µg/m^3 was reported in 2018 (IS6 – 10 days, IS1 – 7 days, SV2 – 3 days), and the lowest count of same days was reported in 2016 and 2015 per entire region and period under study.

The highest values of concentrations of suspended particulate matter smaller than 10 microns in diametre in the ambient air (over 150 µg/m^3) evaluated by means of the daily limit value established gravimetrically were reported at IS1 – Podu de Piatră (193.83 µg/m^3 – November 5th, 2015), IS5 – Tomești (182.76 µg/m^3 – November 5th, 2015) and IS6 – Bosia Ungheni (170.38 µg/m^3 – October 19th, 2018) (Table 7).

High values of PM10 concentration on November 5th, 2015 were also recorded at BC1 – Bacău (100.59 µg/m^3), BC2 – Bacău (126.19 µg/m^3), BT1 (142.22 µg/m^3), SV1 – Suceava (94.85 µg/m^3) and SV2 – Suceava (97.57 µg/m^3). These high levels of PM10 were linked to some particular meteorological conditions on the entire continent as seen from a weather-synoptic perspective, that is they occurred on the background of accentuated thermal inversion.

Table 7 Maximum daily values of PM 10 concentration per different years recorded at the EPA stations in the NE Region of Romania (2015 – 2019).

Station Name	Type	Method	2015	2016	2017	2018	2019
SV 3	Traffic	GRAV	97.57	68.85	0	99.93	96.12
BT 1	Urban	GRAV	142.22	70.02	73.27	100.21	109.39
SV 2	Industrial	GRAV	0	0	60.32	127.00	97.95
SV 1	Urban	GRAV	94.85	61.78	41.61	91.76	68.14
EM 3	EMEP	GRAV	49.79	35.79	0	56.14	35.25
IS 6	Traffic	GRAV	91.34	77.1	60.33	170.38	131.36
IS 4	Rural	GRAV	50	64.77	27.53	100.36	50.00
IS 3	Industrial	LSPM	231.50	106.88	55.61	130.49	145.98
IS 1	Traffic	GRAV	193.83	131.73	87.94	132.19	101.26
IS 2	Urban	GRAV	0	0	68.47	130.62	149.45
IS 5	Suburban	GRAV	182.76	99.16	0	97.71	113.40
NT 2	Industrial	LSPM	0	87.95	90.2	97.18	91.21
NT 1	Urban	GRAV	62.42	72.67	79.22	127.45	96.76
NT 3	Industrial	GRAV	0	99.29	70.85	96.84	157.25
VS 1	Urban	GRAV	0	43.58	46.87	75.39	68.82
VS 2	Urban	GRAV	48.14	48.69	0	0	0
BC 1	Urban	GRAV	104.45	65.63	39.41	69.42	48.65
BC 2	Industrial	GRAV	126.19	74.52	48.83	83.66	94.95
BC 3	Industrial	LSPM	109.89	92.63	108.43	99.50	63.65

After considering the daily data collected at the EPA stations in the NE Region of Romania over the period 2015 – 2019, we identified days with high and extremely high PM10 concentrations whose expressed values exceeded 100 µg/m^3. We correlated the specific time intervals with the EPA stations, thus pinpointing 14 similar space-time intervals in Moldova, in which values over 50 µg/m^3 and even exceeding 100 µg/m^3 were recorded:

a. the EPA stations in all 6 counties in Moldova (Suceava, Botoșani, Neamț, Iași, Bacău, Vaslui): April 5 – 10, 2016, January 3 – 8, 2018 (January 3 - 5 – north of Moldova and January 6 – 8, – south of Moldova), December 1 – 4, 2018, January 27 – 28, 2019, December 16 – 21, 2019.

b. the EPA stations in 5 counties in Moldova (Suceava, Botoșani, Neamț, Iași, Bacău and/or Vaslui): January 19 – 21, 2017, February 1 – 3, 2017, October 17 – 21, 2017, November 18 – 21, 2017, January 24 – 25, 2018, March 2 – 5, 2018, October 18 – 21, 2018, February 10 – 11, 2019.

c. the EPA stations in the County of Iași: September 7 – 17, 2016.

Concerning the EPA stations in the County of Iași, it should be noted that the period of April 5 – 10, 2016 saw favourable conditions for dry build-up of Saharan dust. From a dynamic weather perspective, a Sahara-origin hot air mass occurred in the NE Region during the above mentioned interval. In the south-east and east of the continent there was a broad anticyclone and the north-west experienced a well-defined trough. In such conditions, the Saharan dust reached the north-east region of Romania on a mostly south-east trajectory, at first transported to the forward part of an Atlantic low-pressure system, then to the forward part of the Mediterranean cyclone. Also to be noted is the fact that the above mentioned period was extremely low in precipitation. In this way, this North Africa-originated dust was easy to spread in the ambient air on moderately or intensely windy days, resulting in significant increase in PM10 concentration recorded at all the air quality monitoring stations in Iași City.

In the case of exceeding values indicating PM10 concentration in the month of September 2016, the weather was very hot all over Romania during the time period with PM10 exceeding values and temperatures went higher than 5°C over the multiannual average for the given time interval. Increased temperature and scarce rainfall not only during the above mentioned period of time, but also during the preceding interval led to particulate matter accumulation in the atmosphere. In addition, the south of Italy experienced a cyclone nucleus in the given interval, which triggered several instances of intense dust transport from North-East Africa. The dust reached as far as Romania and stayed in the air in circumstances predominantly anticyclonic and lacking rainfall. Even so, the Saharan dust transport was only moderately intense, leading to the interpretation that the high PM10 concentrations recorded during this period of time were rather the result of high traffic, as shown by the increased concentrations recorded at urban and traffic-type stations, in contrast with the rural and suburban stations that provided lower value recordings. The impact of traffic is brought out by such value distribution, especially as the well-known temperature inversions triggering higher values in the valleys were missing from the picture of such unusually hot interval (*Source: www.meteomoldova.ro - Conf. univ. dr. Lucian Sfîcă, „A. I. Cuza‟ University, Iaşi / Climatology – Faculty of Geography and Geology*).

5. Conclusion

The multiannual concentrations of PM10 pollutant recorded at the EPA stations in the NE Region of Romania fell within legally accepted values (under 40 μg/m^3), with a few exceptions. The highest multiannual averages of PM10 concentration (years 2015 – 2019) were calculated in the case of traffic-type or industrial-type stations Iași 1 (40.90 μg/m^3), Iași 6, Iași 2, Iași 5 and Suceava 2. The lowest multiannual averages of PM10 concentration were recorded at the EMEP station Poiana Stampei (16.70 μg/m^3) as well as at the urban stations Neamț 1, Bacău 1, Vaslui. The year with the highest average of PM10 concentration was 2018, whereas the year with the lowest one was 2015 (notice that many stations were under maintenance, malfunctioning or out of order).

The highest monthly averages of PM10 concentration were calculated in the cold season months (especially October, November, February, March), whereas the lowest ones were recorded in the spring-end and summer months (May – August). High monthly averages of PM10 concentration (over 40 µg/m^3) were determined based on daily recordings at the urban or traffic-type stations IS1 (all year round), SV3 (February, October), IS6 (August, October), IS2 (March, October), SV2 (January, February, November). The main reasons for such increased averages are: the large number of mobile sources (emissions generated by car traffic), residential sources (generating combustion to produce thermal energy), meteorological conditions triggering the occurrence and persistence of particulate matter in the atmosphere (still air and fog conditions typical of the cold season lead to particulate matter build-up on the ground surface, thus causing high pollutant concentrations).

The lowest monthly averages of PM10 concentration (under 20 µg/m^3) were determined at stations NT1 (April – September), SV1 and VS1 (April – June), BC2, BC3, IS3, NT2, NT 3 and SV2 (April – August). The lowest ever monthly averages of PM10 concentration were recorded at station EM3 – Poiana Stampei (located in conditions of relative isolation in relation to pollutant sources and on the medium altitude mountain floor – approximately 900 m). These low PM10 concentration averages are explained by intense air ventilation, high amount of precipitation, consistent length of rain and snow intervals, as well as by the reduction of thermal energy consumption during the warm season.

The highest count of days with PM10 levels over 50 µg/m^3 (bad air quality) was collected at IS1 – Podu de Piatră (119 days in 2018, 99 days in 2017), IS6 – Bosia Ungheni (99 days in 2018), IS2 – Decebal Cantemir (69 days in 2018) and SV2 – Suceava (38 days in 2018), all urban and traffic-type EPA stations. Station EM3 – Poiana Stampei did not exhibit any day with PM10 values over 50 µg/m^3 during the period of time between years 2015 – 2019. A low count of days with PM10 levels over 50 µg/m^3 was recorded at the urban stations (located downtown) NT 1 – Piatra Neamț, SV1 – Suceava, VS1 – Vaslui and the rural station IS4 – Copou / Aroneanu.

The highest count of days with PM10 levels exceeding 100 µg/m^3 (very bad air quality) was recorded in year 2018, whereas the lowest count of such days was determined in the years 2016 and 2015. The highest values of concentrations of suspended particulate matter smaller than 10 microns in diametre in the ambient air evaluated by means of the daily limit value established gravimetrically were reported at IS1 – Podu de Piatră (193.83 µg/m^3) and IS5 – Tomești (182.76 µg/m^3) on November 5th, 2015.

The hereabove analysis presented the multiannual, annual and in some cases daily evolution of PM10 suspended particulate matter concentration as recorded at various stations in the NE Region of Romania and provided quantitative, real and precise data on the time shifting and space distribution of the air pollution degree caused by PM10 suspended particulate matter and the quality of ambient air.

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