Atmospheric levels of PM₁₀ in relation to meteorological elements. Case study: Moldova Region, Romania

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ABSTRACT: The major goal of our research was to find statistically relevant mathematical correlations between PM 10 levels in the atmosphere and the meteorological complex over the Moldova - Romania region. The period of analysis was from 2009 to 2020. We worked on daily data on PM10 levels and meteorological parameters from a network of 25 monitoring stations. The correlations between PM10 levels and air temperature, solar radiation, wind and precipitation were negative. The correlations between PM10 concentrations, atmospheric pressure and relative humidity were positive. However, over the medium term (12 years), the overall climatological correlations between PM10 concentrations and meteorological elements (assessed using Pearson coefficients) had values below ±0.5. This suggests that PM10 levels are not primarily influenced by meteorological elements but rather are closely associated with daily and annual patterns of socioeconomic activities. At time intervals of a few days up to 1-2 weeks, in certain favorable geographical situations, the correlative links between PM10 levels and meteorological elements can become strong and relevant (the Pearson index has in these temporal and spatial contexts values above or below 0.9 units).

KEY WORDS: PM₁₀, air quality, quality indices, Moldova – Romania region.

1. Introduction

High concentrations of atmospheric pollutants have a significant impact on the health of the population, the most harmful effect being caused by particulate matter and nitrogen dioxide. Extensive research has been conducted in this direction, spanning global, regional and local level. Numerous articles have been published in specialized journals aiming to disclose the factors driving this process, and outlining the required the measures to mitigate pollution, for example, *Atmospheric Chemistry and Physics, Environmental Monitoring and Assessment, Natural Hazards and Earth System Sciences*, etc.

There are studies published in specialized journals that cover nearly the entire Europe, addressing various topics related to: the analysis of PM10 concentrations in close correlation with chemical processes and emission sources (*Grivas et al., 2004; Kiesewetter et al., 2015; Hoffer et al. al., 2020; Liaskoni et al., 2023*); the identification of the relationship between climatic factors and the evolution of PM10 concentrations (*Houthuijs et al., 2011; Markovic et al., 2008; Bielawska, Wardencki, 2014; Largeron, Staquet, 2016; Gašparac et al., 2020; Tsyro et al., 2022*); the influence of human-related factors on the variations in the concentration of mobile dust suspension (*Houthuijs et al., 2011; Gualtieri et al., 2015; Matthias et al., 2021; Borlaza et al., 2022; Gorrochategui et al., 2022*)

In Romania, there are studies investigating: the impact of PM10 pollution episodes on the health of Bucharest residents and the surrounding environment (*Raicu, Iorga, 2009; Proorocu et al., 2014*); the role of meteorological factors in the PM10 concentration regime for two monitoring stations located in the green perimeters of Bucharest (*Bodor et al., 2020*); air pollution with PM2.5 and respectively with PM10 in a comparative study for the city of Târgu Jiu (*Căpățînă et al., 2016*); the role of meteorological parameters in explaining the variations of PM10 concentrations on the territory of our country (*Maco B.A. et al., 2016*) ; the effects of meteorological conditions on atmospheric pollution with PM10 and on human health in the city of Brăila (*Bălănică et al., 2019*);

In the case of Moldova Region, until 2010, there were fewer studies conducted (*Gugiuman*, *Cotrău*, 1975; Erhan, 1979), but in the recent years their number has increased. These studies were focused on the emission sources of mobile dust – PM10 and the influence of meteorological conditions on the variation of PM10 concentrations in the urban agglomerations of Suceava, Piatra Neamţ, Iaşi, Botoşani (*Lazurcă*, 2015; Sfîcă et al., 2018; Roșu et al., 2020; Nistor et al., 2020). In Suceava municipality, *Lazurcă*, 2015 and Nistor et al., 2020 looked at the influence of meteorological factors (temperature, precipitation, humidity, atmospheric pressure, wind direction and speed) on the distribution of PM10 concentrations and air quality In Iaşi metropolitan area, studies (*Sfîcă et al., 2018*) explored the role of climatic and meteorological factors on the temporal and spatial variability of PM10 particles and the connection between thermal inversions and pollution conditions. In the case of Piatra Neamţ city, Roșu et al., 2020 work showed that the excellent air quality related to the low values of PM10 concentrations increased the tourist attraction of the city.

The main aim of the present study is to assess the spatio-temporal distribution of PM_{10} at the air quality monitoring stations across the Moldova Region and to identify the stations and intervals where the air quality index exceeds the daily limit value allowed by Law 104/2011, in close connection with climatic elements (air temperature, atmospheric pressure, relative humidity, solar radiation, wind speed).

The objectives of the present work aim at *i*) the spatial and temporal analysis of PM_{10} concentrations over the period 2009 – 2020 for the Moldova Region, *ii*) the identification of the stations and intervals in which the air quality index was average, bad and very bad and *iii*) highlighting the relationships between spatio-temporal distribution of PM_{10} concentrations and meteorological elements (air temperature, precipitation, humidity, atmospheric pressure).

2. Study area

Moldova region is located in the north-eastern and eastern part of Romania, close to the border with Ukraine and the Republic of Moldova. It stretches between 48°15'55.893"N and 45°22'6.52"N northern latitude and between 24°58' 16.925"E and 28°17'2.602"E east longitude (Figure 1, Table 1).

3. Methods

In general, for measuring PM_{10} dust, which represents the dimensional fraction of toxicological interest in the urban aerosol, 2 methods are applied: the GRAV gravimetric method (reference method) and the LSPM nephelometric method (automatic measurements, for informational purposes; the recorded exceedances can be later validated/invalidated by the gravimetric method).

The reference method for sampling and measuring PM_{10} is the one provided in the EN 12341 standard - "Ambient air. Standardized method for gravimetric measurement to determine the mass fraction of PM_{10} or $PM_{2.5}$ of suspended particles". Most APM stations use the gravimetric method, but some use the nephelometric method (the data used in this study were analysed by the gravimetric one). The stations that use only the nephelometric method are: IS-3, NT-2, BC-3, GL-5.

The research methods used consisted of: *i*) collectiong general information about PM₁₀ by reviewing scientific articles and publications, *ii*) analysing and comparatively interpreting statistical data (using the Pearson correlation coefficient, PCA – Principal Component Analysis), *iii*) graphically representing PM₁₀ concentration values analongside their corresponding quality indices and *iiii*) interpretating graphic and cartographic material.

These methods are essential for analysing PM_{10} suspended particles concentrations because they provide quantitative, real and accurate data regarding the temporal evolution and spatial distribution of air quality, particularly in relation to their causality.

The air quality monitoring system from Moldova includes 25 stations for evaluating the influence of different types of pollution sources on air quality. Therefore, in this region, one can find: 1 *EMEP-type station* (EM3), *8 industrial-type stations* (SV-2, IS-3, NT-2, NT-3, BC-2, BC-3, GL-4, GL-5), *8 urban type stations* (BT-1, SV-1, IS-2, NT-1, VS-2, VS-1, BC-1, GL-2), *4 traffic type stations* (SV-3, IS-1, IS-6, GL-1), *2 suburban type stations* (IS-5, GL-3) *2 rural type stations* (IS-4, VN-1) (Figure 2, Table 1).

The potential health negative impacts of mobile dusts in suspension, especially on individuals with asthma, cardiovascular and respiratory diseases, children, and the elderly, is directly contingent on the size of PM_{10} suspended particles. It is very important to follow the specific daily quality index corresponding to the pollutant PM_{10} , in order to not exceed the limit values for the protection of human health approved in Law no. 104 of June 15, 2011 (Table 2).



Figure 1 Map of the Moldova Region showing the location of APM station studied.

 Table 1 Geographical characteristics of the of APM station locations within the Moldova Region.

		l atitude					Location details /
Station	Station type	Longitude	Alt m	TAU	Relief unit	Relief subunit	pollution sources
EM-3	EMEP	47°19'28"N 25°08'03"E	910	Poiana Stampei	Eastern Carpathians	Suhard Mountains	car traffic E58
SV-1	Urban	47°38'58"N 26°14'59"E	373	Suceava	Suceava Plateau	Dragomirnei Plateau	downtown
BT-1	Urban	47°44'27"N 26°39'31"E	168	Botoșani	Moldavian Plain	Cozancea Hill	downtown
NT-1	Urban	46°56′03″N 26°23′20″E	357	Piatra Neamț	Moldova Subcarpathians	Bistritei Valley	downtown
IS-2	Urban	47°09'03"N 27°34'54"E	42	lași	Moldavia Plain	lasi Coast, Bahlui Valley	downtown
BC-1	Urban	46°33'50"N 26°54'37"E	169	Bacău	Siret Corridor	Bistrita Valley – Siret Valley	downtown
VS-1	Urban	46°37′56″N 27°43′49″E	107	Vaslui	Barlad Plateau	Barlad Valley	downtown
VS-2	Urban	46°40'30″N 28°05'47″E	80	Huși	Barlad Plateau	Prut Valley	the edge of the city, car traffic E581
GL-2	Urban	45°25'53"N 28°03'17"E	33	Galați	Lower Siret Plain	Danube Valley	downtown
SV-2	Industrial	47°40'09"N 26°16'53"E	289	Suceava	Suceava Plateau	Suceava Valley	Burdujeni neighbourhood, industrial platform AMBRO – CET
NT-2	Industrial	46°56′53″N 26°55′04″E	207	Roman	Suceava Plateau	Confluence Moldova – Siret Valley	Industrial platform Roman (UMARO, Sugar factory)
NT-3	Industrial	46°54′11″N 26°02′15″E	558	Tașca	Eastern Carpathians	Bicaz Valley	Industrial platform Heidelberg Cement
IS-3	Industrial	47°09'28"N 27°36'45"E	64	lași	Moldavian Plain	Bahlui Valley	Tătărași neighbourhood, industrial platform Chimia
BC-2	Industrial	46°33'19"N 26°55'32"E	157	Bacău	Siret Corridor	Bistrita Valley – Siret Valley	Cornişa neighbourhood, industrial platform LETEA
BC-3	Industrial	46°15′24″N 26°47′33″E	200	Onești	Moldova Subcarpathians	Trotus Valley	industrial platform CAROM
GL-4	Industrial	45°24'43"N 28°00'20"E	45	Galați	Lower Siret Plain	Danube Valley – Siret Valley	Car traffic E87
GL-5	Industrial	45°49'05"N 27°26'21"E	31	Tecuci	Barlad Plateau	Barlad Valley	Car traffic E581
SV-3	Traffic	47°57′21″N 26°04′06″E	302	Siret	Suceava Plateau	Siret Valley	Car traffic E85
IS-1	Traffic	47°09'24"N 27°34'29"E	40	lași	Moldavian Plain	Bahlui Valley	Car traffic E58
IS-6	Traffic	47°12′56″N 27°46′07 E	34	Ungheni	Moldavian Plain	Prut Valley	Economic region Ungheni – Rep. Moldova
GL-1	Traffic	45°25'14"N 28°01'06 E	51	Galați	Lower Siret Plain	Danube Valley	Car traffic E87
IS-4	Rural	47°12′47″N 27°36′39″E	186	Aroneanu	Moldavian Plain	Ciric Valley	International Airport Iași
VN-1	Rural	45°41'49"N 27°12'48"E	45	Focșani	Lower Siret Plain	Lower Siret Plain terrace	Industrial platform Focșani
IS-5	Suburban	47°08'09"N 27°41'35"E	37	Tomești	Moldavian Plain	Bahlui Valley	Metropolitan region Iași, CET Holboca
GL-3	Suburban	45°28'22"N 28°01'57" E	68	Galați	Lower Siret Plain	Danube Valley – Brates Lake	Car traffic DN26



Figure 2 Map showing the types of APM stations used in this study.

Idule 2 Concentration ranges for Pivi10 (µg/III) (according to Law 104/2011 and Order 1010/2020	Table	2 Concentration	ranges for PM ₁₀	$_{0} (\mu g/m^{3}) (a$	according to	Law 104/2011	and Order	1818/2020
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Concentration range for suspended particles (μg / m³) (Law 104 / 2011)	Quality index	Specific Index	Concentration range for suspended particles (μg / m³) (Order 1818 / 2020)	Quality index	Specific Index
0 - 10	excellent	1	0 - 20	Good	1
10 - 20	very good	2	20 - 40	acceptble	2
20 - 30	good	3	40 - 50	moderate	3
30 - 50	medium	4	50 - 100	bad	4
50 - 100	bad	5	100 - 150	very bad	5
>100	very bad	6	150 - 1200	extremely bad	6

In Romanian legislation, the specific index corresponding to PM_{10} was recently updated, according to Order no. 1818 / 2020 (Table 2). Limit values for the protection of human health were established for the PM_{10} pollutant to 50 μ g/m³ – for the daily limit value and 40 μ g/m³ for the annual limit value. The specific index corresponding to PM_{10} is established by fitting the arithmetic mean of the hourly values, recorded in the last 24 hours (daily averages), in one of the

concentration ranges listed in Table 2. In our study, we adopted the fitting from Table 2 according to Law 104/2011, because the analysis period overlaps this interval.

The data used in this study for PM₁₀ pollution cover the interval between 2009 to 2020. The data were obtained from the County Environmental Protection Agencies of Moldova. According to Annex 4 of Law no. 104/2011, the monitoring data quality objective regarding the minimum data capture for the 1-year averaging period is 90%. Given that the 90% capture requirement does not include data loss due to routine calibration, checks and maintenance, valid data captures of at least 75% are considered compliant (Table 3). For the studied region, it is observed that the highest percentage of valid data is reached at the stations: BC-2, BC-3, IS-1, IS-4. The lowest percentage of valid data was recorded at the stations: GL-5, GL-2, IS-2, VS-2.

PM 10	2009	2010	2011	2012	2013	2014	2015	2016	2017	2018	2019	2020	AVERAGE
BC-1	0.0	0.0	0.0	85.2	81.0	94.6	86.3	82.6	15.8	90.6	50.7	80.6	55.6
BC-2	80.9	88.1	94.5	86.6	85.8	91.8	77.0	67.9	13.7	91.8	95.7	96.1	80.8
BC-3	80.1	84.8	58.3	83.0	81.2	44.6	63.6	96.7	96.2	95.1	67.6	93.6	78.7
BT-1	0.0	0.0	32.5	96.2	91.9	98.4	88.2	99.2	95.7	98.6	98.7	97.0	74.7
EM-3	0.0	80.2	90.2	96.2	67.4	60.5	18.8	12.0	0.0	75.2	26.1	48.7	47.9
IS-1	94.6	45.5	79.3	97.0	80.6	53.8	83.5	75.8	93.6	98.1	91.4	95.7	82.4
IS-2	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	86.4	98.6	93.3	98.6	31.4
IS-3	34.6	52.0	67.9	95.6	80.6	67.0	77.1	25.8	97.8	92.6	63.0	89.7	70.3
IS-4	8.1	77.6	95.4	99.4	98.4	93.5	65.4	53.7	81.0	87.4	87.3	92.7	78.3
IS-5	0.0	0.0	0.0	0.0	95.4	88.0	67.3	76.9	33.6	88.9	66.0	94.2	50.9
IS-6	0.0	36.8	12.0	85.1	85.3	87.4	16.5	17.6	66.1	91.8	65.6	60.1	52.0
NT-1	0.0	0.0	24.5	99.5	99.4	83.3	48.8	41.1	94.8	97.0	85.0	88.8	63.5
NT-2	32.8	41.9	55.2	70.1	43.6	0.0	0.1	42.5	95.3	93.8	98.6	91.9	55.5
NT-3	0.0	63.5	0.0	40.7	0.0	0.0	0.1	41.7	49.1	93.0	98.9	91.4	39.9
SV-1	0.0	0.0	0.0	83.1	87.9	88.3	75.5	79.7	77.9	91.8	87.5	86.9	63.2
SV-2	88.9	86.4	80.2	85.2	88.6	0.0	0.1	0.0	94.8	97.8	97.8	93.7	67.8
SV-3	0.0	75.6	86.3	87.0	80.2	83.0	79.2	49.4	0.0	78.0	94.1	36.3	62.4
VS-1	92.6	95.3	77.8	49.1	63.4	19.4	0.0	42.4	39.2	95.7	92.2	86.9	62.8
VS-2	0.0	0.0	72.0	84.2	95.7	93.4	90.6	27.8	0.0	0.0	0.0	0.0	38.6
GL-1	39.1	0.0	0.0	26.1	78.3	72.0	74.2	58.1	75.5	91.3	95.1	95.6	58.8
GL-2	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	60.7	90.4	94.3	97.0	28.5
GL-3	28.0	22.1	19.6	70.2	7.3	0.0	66.0	0.0	61.2	81.7	94.6	94.0	45.4
GL-4	46.0	34.9	38.7	63.3	27.3	23.8	66.9	0.0	77.4	86.1	86.1	53.7	50.3
GL-5	66.8	15.0	0.0	0.0	0.0	0.0	0.0	0.0	86.0	91.3	28.0	33.8	26.7
VN-1	44.6	56.5	75.4	67.5	55.6	30.5	0.0	3.5	79.0	92.3	95.4	97.3	58.1

Table 3 Data capture (%) for the pollutant PM₁₀ at APM stations in Moldova (2009 – 2020).

4. Results and discussion

4.1. Analysis of air quality in Moldova based on PM₁₀ levels

The concentrations of suspended particles with a diameter of <10 microns in the surrounding air were assessed using the annual limit value (40 μ g/m³). The average annual concentrations of PM₁₀ from the APM stations in the Moldova Region were, in general, within the legally allowed values (below 40 μ g/m³). The highest multiannual average (2009 – 2020) was calculated for station IS-1 (41.2 μ g/m³). This average exceeds the allowed annual limit value. Throughout the studied interval, the IS-1 station (traffic-type) had values above or close to the annual allowed limit, due to the intense road traffic on the Stone Bridge and railway traffic (the railway tracks pass under the bridge). Other high multi-year averages were calculated for stations IS-2 (32.5 μ g/m³), IS-6 (32.1 GEOREVIEW 33.1 (95-117)

 μ g/m³), SV-2 (31.6 μ g/m³) which are traffic-type stations or industrial. The lowest multiannual averages of atmospheric levels of PM₁₀ were calculated for the EM-3 (16.9 μ g/m³; cross-border station, located at the altitude of 910 m) and the stations in the city of Galati: GL-2 (14.9 μ g/m³), GL-3 (16.7 μ g/m³) and GL-4 (17.5 μ g/m³) (after the year 2000, the activity of the Arcelor Mittal Galati steel plant was constantly reduced, the production gradually decreased and thus, the pollution level were drastically reduced <u>https://ro.wikipedia.org/wiki/</u>Combinatul_siderurgic_din_Gala%C8%9Bi) (Table 4).

In the Moldova Region, between 2009 and 2020, the highest monthly averages of PM10 concentrations were registered during the cold season (October - March), while the lowest averages were visible for the spring and summer months (May - August). *Industrial-type* APM stations recorded relatively high and uniform PM₁₀ values throughout the year, with a maximum during January or October (approx. 35-40 μ g/m³) and a minimum during May - July. At *urban-type* and *traffic-type* APM stations, PM₁₀ concentrations had variable values during the year, with a maxima during March and October – November and a minima during summer months. At the *rural* APM stations, the monthly averages show little variability throughout the year, between 15-25 μ g/m³. The variation of PM₁₀ mobile dust concentrations was also reduced at the *EMEP type* APM station (EM-3), but the average monthly values were lower throughout the year, when compared to those from the rural stations. *Suburban* APM stations recorded monthly averages of mobile dust, intermediate in value, between urban and rural all year round (over 25 μ g/m³).

Table 4 Average annual concentrations for the PM10 pollutant at APM stations in the Moldova Region	۱
(2009 – 2020).	

JAN	FEB	MAR	APR	ΜΔΥ	ILIN			SED	ОСТ	NOV	DEC	VEAD
20.4					1014	JOL	AUG	JLF	001		DLC	TEAN
20.4	19.9	20.4	17.6	14.1	12.9	14.8	15.8	14.1	16.1	18.9	17.8	16.9
25.3	28.1	24.0	19.1	17.5	19.3	21.7	24.1	23.5	27.0	26.7	24.1	23.4
41.4	37.0	35.6	28.9	21.5	21.3	25.7	28.5	29.7	35.0	35.1	38.9	31.6
31.0	33.4	32.2	23.5	21.0	21.7	25.1	27.2	26.3	31.9	32.1	30.1	27.9
30.5	31.9	30.1	27.5	22.0	23.0	25.2	28.8	27.7	30.8	33.6	31.9	28.6
30.6	29.9	23.1	16.9	13.4	15.5	15.9	16.9	19.3	24.1	28.2	24.3	21.5
32.9	28.3	26.3	21.4	12.1	13.7	16.0	16.4	16.2	23.5	27.5	29.5	22.0
40.8	36.6	27.8	22.6	14.5	16.6	16.6	20.9	21.0	27.1	37.6	35.2	26.4
23.4	24.8	23.9	20.3	17.1	16.7	18.1	19.0	17.9	22.1	23.6	24.7	21.0
27.4	25.0	22.8	22.0	17.7	16.8	19.1	21.0	21.6	23.0	25.6	26.4	22.4
31.0	26.9	22.8	17.0	13.0	13.2	14.8	18.0	17.6	24.1	27.2	33.9	21.6
41.2	43.1	53.6	43.9	36.9	34.4	35.7	38.8	41.4	45.0	44.1	36.0	41.2
38.1	37.7	39.0	30.6	26.0	26.5	26.7	31.3	29.7	38.7	34.5	30.6	32.5
31.0	26.6	24.0	18.0	14.4	17.7	19.7	20.8	22.6	35.5	33.9	30.0	24.5
23.6	25.2	23.0	20.6	18.0	18.0	21.5	24.2	22.2	23.2	22.0	22.4	22.0
33.1	32.2	32.1	27.1	23.5	22.8	26.0	31.6	30.4	35.2	31.1	28.4	29.4
27.0	28.4	34.6	32.7	27.5	27.9	36.2	38.1	35.9	41.9	30.0	25.1	32.1
26.4	23.5	23.0	19.7	16.5	18.1	19.5	21.1	18.8	24.7	29.2	28.9	22.5
26.5	24.8	24.0	22.7	22.7	21.1	27.7	25.5	29.1	28.4	28.6	29.4	25.9
24.2	21.9	19.1	17.7	16.2	15.6	18.1	19.4	18.4	20.1	20.4	21.6	19.4
22.3	19.9	19.3	18.1	16.1	17.4	19.0	17.7	18.0	21.4	21.9	20.9	19.3
17.7	12.6	13.9	13.3	12.1	11.4	14.3	15.7	14.8	18.3	18.6	16.4	14.9
18.5	14.7	17.8	14.4	12.5	12.9	16.8	18.1	17.0	21.5	18.9	17.2	16.7
19.2	17.9	18.9	15.5	14.2	14.8	17.5	16.5	16.9	19.0	20.2	19.7	17.5
21.4	25.5	22.9	15.5	12.7	13.9	18.9	21.2	18.6	21.8	25.5	18.8	19.7
	20.4 25.3 41.4 31.0 30.5 30.6 32.9 40.8 23.4 27.4 31.0 41.2 38.1 31.0 23.6 33.1 27.0 26.4 26.5 24.2 22.3 17.7 18.5 19.2 21.4	20.4 19.9 25.3 28.1 41.4 37.0 31.0 33.4 30.5 31.9 30.6 29.9 32.9 28.3 40.8 36.6 23.4 24.8 27.4 25.0 31.0 26.9 41.2 43.1 38.1 37.7 31.0 26.6 23.6 25.2 33.1 32.2 27.0 28.4 26.4 23.5 26.5 24.8 24.2 21.9 22.3 19.9 17.7 12.6 18.5 14.7 19.2 17.9 21.4 25.5	20.4 19.9 20.4 25.3 28.1 24.0 41.4 37.0 35.6 31.0 33.4 32.2 30.5 31.9 30.1 30.6 29.9 23.1 32.9 28.3 26.3 40.8 36.6 27.8 23.4 24.8 23.9 27.4 25.0 22.8 31.0 26.9 22.8 41.2 43.1 53.6 38.1 37.7 39.0 31.0 26.6 24.0 23.6 25.2 23.0 31.0 26.6 24.0 23.6 25.2 23.0 33.1 32.2 32.1 27.0 28.4 34.6 26.5 24.8 24.0 24.2 21.9 19.1 22.3 19.9 19.3 17.7 12.6 13.9 18.5 14.7 17.8 19.2 17.9 18.9 21.4 25.5	20.4 19.9 20.4 17.6 25.3 28.1 24.0 19.1 41.4 37.0 35.6 28.9 31.0 33.4 32.2 23.5 30.5 31.9 30.1 27.5 30.6 29.9 23.1 16.9 32.9 28.3 26.3 21.4 40.8 36.6 27.8 22.6 23.4 24.8 23.9 20.3 27.4 25.0 22.8 22.0 31.0 26.9 22.8 17.0 41.2 43.1 53.6 43.9 38.1 37.7 39.0 30.6 31.0 26.6 24.0 18.0 23.6 25.2 23.0 20.6 33.1 32.2 32.1 27.1 27.0 28.4 34.6 32.7 26.4 23.5 23.0 19.7 26.5 24.8 24.0 22.7 24.2 21.9 19.1 17.7 26.5 24.8	20.4 19.9 20.4 17.6 14.1 25.3 28.1 24.0 19.1 17.5 41.4 37.0 35.6 28.9 21.5 31.0 33.4 32.2 23.5 21.0 30.5 31.9 30.1 27.5 22.0 30.6 29.9 23.1 16.9 13.4 32.9 28.3 26.3 21.4 12.1 40.8 36.6 27.8 22.6 14.5 23.4 24.8 23.9 20.3 17.1 27.4 25.0 22.8 17.0 13.0 41.2 43.1 53.6 43.9 36.9 38.1 37.7 39.0 30.6 26.0 31.0 26.6 24.0 18.0 14.4 23.6 25.2 23.0 20.6 18.0 33.1 32.2 32.1 27.1 23.5 27.0 28.4 34.6 32.7 27.5 26.4 23.5 23.0 19.7 16.5 2	20.419.920.417.514.112.925.328.124.019.117.519.341.437.035.628.921.521.331.033.432.223.521.021.730.531.930.127.522.023.030.629.923.116.913.415.532.928.326.321.412.113.740.836.627.822.614.516.623.424.823.920.317.116.727.425.022.822.017.716.831.026.922.817.013.013.241.243.153.643.936.934.438.137.739.030.626.026.531.026.624.018.014.417.723.625.223.020.618.018.033.132.232.127.123.522.827.028.434.632.727.527.926.423.523.019.716.518.126.524.824.022.722.721.124.221.919.117.716.215.622.319.919.318.116.117.417.712.613.913.312.111.418.514.717.814.412.512.919.217.918.	20.419.920.417.614.112.914.825.328.124.019.117.519.321.741.437.035.628.921.521.325.731.033.432.223.521.021.725.130.531.930.127.522.023.025.230.629.923.116.913.415.515.932.928.326.321.412.113.716.040.836.627.822.614.516.616.623.424.823.920.317.116.718.127.425.022.822.017.716.819.131.026.922.817.013.013.214.841.243.1 53.6 43.936.934.435.738.137.739.030.626.026.526.731.026.624.018.014.417.719.723.625.223.020.618.018.021.533.132.232.127.123.522.826.027.028.434.632.727.527.936.226.524.824.022.722.721.127.724.221.919.117.716.518.119.526.524.824.022.722.721.127.724.221.919	20.419.920.417.614.112.914.815.825.328.124.019.117.519.321.724.141.437.035.628.921.521.325.728.531.033.432.223.521.021.725.127.230.531.930.127.522.023.025.228.830.629.923.116.913.415.515.916.932.928.326.321.412.113.716.016.440.836.627.822.614.516.616.620.923.424.823.920.317.116.718.119.027.425.022.817.013.013.214.818.041.243.153.643.936.934.435.738.838.137.739.030.626.026.526.731.331.026.624.018.014.417.719.720.823.625.223.020.618.018.021.524.233.132.232.127.123.522.826.031.627.028.434.632.727.527.936.238.126.423.523.019.716.518.119.521.126.524.824.022.722.721.127.725.5	20.419.920.417.614.112.914.815.814.125.328.124.019.117.519.321.724.123.541.437.035.628.921.521.325.728.529.731.033.432.223.521.021.725.127.226.330.531.930.127.522.023.025.228.827.730.629.923.116.913.415.515.916.919.332.928.326.321.412.113.716.016.416.240.836.627.822.614.516.616.620.921.023.424.823.920.317.116.718.119.017.927.425.022.817.013.013.214.818.017.641.243.153.643.936.934.435.738.841.438.137.739.030.626.026.526.731.329.731.026.624.018.014.417.719.720.822.623.625.223.020.618.018.021.524.222.233.132.232.127.123.527.936.238.135.926.423.523.019.716.518.119.521.118.826.52	20.419.920.417.614.112.914.815.814.116.125.328.124.019.117.519.321.724.123.527.041.437.035.628.921.521.325.728.529.735.031.033.432.223.521.021.725.127.226.331.930.531.930.127.522.023.025.228.827.730.830.629.923.116.913.415.515.916.919.324.132.928.326.321.412.113.716.016.416.223.540.836.627.822.614.516.616.620.921.027.123.424.823.920.317.116.718.119.017.922.127.425.022.817.013.013.214.818.017.624.141.243.153.643.936.934.435.738.841.445.038.137.739.030.626.026.526.731.329.738.731.026.624.018.014.417.719.720.822.635.523.625.223.020.618.018.021.524.222.223.233.132.232.127.123.527.936.2 </td <td>20.4 19.9 20.4 17.6 14.1 12.9 14.8 15.8 14.1 16.1 18.9 25.3 28.1 24.0 19.1 17.5 19.3 21.7 24.1 23.5 27.0 26.7 41.4 37.0 35.6 28.9 21.5 21.3 25.7 28.5 29.7 35.0 35.1 30.5 31.9 30.1 27.5 22.0 23.0 25.2 28.8 27.7 30.8 33.6 30.6 29.9 23.1 16.9 13.4 15.5 15.9 16.9 19.3 24.1 28.2 32.9 28.3 26.3 21.4 12.1 13.7 16.0 16.4 16.2 23.5 27.5 40.8 36.6 27.8 22.6 14.5 16.6 16.6 20.9 21.0 27.1 37.6 23.4 24.8 23.9 20.3 17.1 16.7 18.1 19.0 17.9 22.1 23.6 27.4 25.0 22.8 17.0 13.0 13.</td> <td>20.4 19.9 20.4 17.6 14.1 12.9 14.8 15.8 14.1 16.1 18.9 17.8 25.3 28.1 24.0 19.1 17.5 19.3 21.7 24.1 23.5 27.0 26.7 24.1 41.4 37.0 35.6 28.9 21.5 21.3 25.7 28.5 29.7 35.0 35.1 38.9 31.0 33.4 32.2 23.5 21.0 21.7 25.1 27.2 26.3 31.9 32.1 30.1 30.5 31.9 30.1 27.5 22.0 23.0 25.2 28.8 27.7 30.8 33.6 31.9 30.6 29.9 23.1 16.9 13.4 15.5 15.9 16.9 19.3 24.1 28.2 24.3 32.9 28.3 26.3 21.4 12.1 13.7 16.0 16.4 16.2 23.5 27.5 29.5 40.8 36.6 27.8 22.6 14.5 16.6 16.6 20.9 21.0 27.1 37</td>	20.4 19.9 20.4 17.6 14.1 12.9 14.8 15.8 14.1 16.1 18.9 25.3 28.1 24.0 19.1 17.5 19.3 21.7 24.1 23.5 27.0 26.7 41.4 37.0 35.6 28.9 21.5 21.3 25.7 28.5 29.7 35.0 35.1 30.5 31.9 30.1 27.5 22.0 23.0 25.2 28.8 27.7 30.8 33.6 30.6 29.9 23.1 16.9 13.4 15.5 15.9 16.9 19.3 24.1 28.2 32.9 28.3 26.3 21.4 12.1 13.7 16.0 16.4 16.2 23.5 27.5 40.8 36.6 27.8 22.6 14.5 16.6 16.6 20.9 21.0 27.1 37.6 23.4 24.8 23.9 20.3 17.1 16.7 18.1 19.0 17.9 22.1 23.6 27.4 25.0 22.8 17.0 13.0 13.	20.4 19.9 20.4 17.6 14.1 12.9 14.8 15.8 14.1 16.1 18.9 17.8 25.3 28.1 24.0 19.1 17.5 19.3 21.7 24.1 23.5 27.0 26.7 24.1 41.4 37.0 35.6 28.9 21.5 21.3 25.7 28.5 29.7 35.0 35.1 38.9 31.0 33.4 32.2 23.5 21.0 21.7 25.1 27.2 26.3 31.9 32.1 30.1 30.5 31.9 30.1 27.5 22.0 23.0 25.2 28.8 27.7 30.8 33.6 31.9 30.6 29.9 23.1 16.9 13.4 15.5 15.9 16.9 19.3 24.1 28.2 24.3 32.9 28.3 26.3 21.4 12.1 13.7 16.0 16.4 16.2 23.5 27.5 29.5 40.8 36.6 27.8 22.6 14.5 16.6 16.6 20.9 21.0 27.1 37

Average daily concentrations of PM_{10} suspended particles are directly influenced by meteorological factors including: wind direction and speed, precipitation, air temperature, atmospheric pressure, etc. and the geographical factors specific to the studied area. The types of

the atmosphere thermal stratification and temperature inversions can greatly influence the pollutants accumulation. An important factor is represented by the cars that transits the central parts of the cities the national roads outside the city and the routes that transit the villages. During the day, the period when the traffic is high is between 07:30 and 19:00, which generates high values of the PM₁₀ particles concentration. On Saturdays and Sundays, the traffic is reduced and thus PM₁₀ particles register the lower values –, an interval also known as the "weekend effect" (*Nistor et al., 2020*).

4.2. Frequency of quality indices related to PM₁₀ levels



4.2.1. Multiannual frequency of air quality indices related to PM₁₀

The highest frequencies of quality indices 4 – 6 (average, bad and very bad) were recorded at the stations in the Iaşi metropolitan area (IS-1, IS-2, IS-6, IS-5). The lowest frequencies were recorded in the Galati metropolitan area and the EM-3 Poiana Stampei station (Figure 3).

Figure 3 Frequency (%) of daily air quality indices (classes 1-6) at each individual monitoring station for the PM10 indicator in the Moldova Region (2009-2020).

4.2.2. Multi-year frequency of PM_{10} -related air quality indices focused on quality indices of classes 6, 5 and 4

The population well-being is at risk when the particular daily quality index corresponding to suspended particles PM_{10} falls into classes 4 (medium), 5 (bad) and 6 (very bad). The analysis of the frequency of quality indices 6 (very bad), 5 (bad) and 4 (average) was carried out based on the station types (EMEP, industrial, traffic, urban, suburban, rural). This approach was necessary because depending on the station location there are different sources and regimes of the PM_{10} pollutant and the correlations what appear between the spatio-temporal distribution of PM_{10} concentrations and meteorological elements (air temperature, precipitation, humidity, atmospheric pressure) (Table 5).

The maximum values of PM₁₀ concentrations occur at industrial or traffic type stations, in the cold season months, under the conditions of an anticyclonic synoptic circulation or a high pressure field, against the background of accentuated thermal inversions, which generate stable, but cold and frosty weather (*Grant from the Ministry of Research, Innovation and Digitization, CNCS-UEFISCDI, project number PN-III-P1-1.1-TE-2021-0882, within PNCDI III*).

EMEP type station. At station EM-3 the frequency of average and bad quality indices is low registering 10.3% and 0.3%. Between 2009-2020, there were no cases in which PM10 dust concentrations exceeded 100 μ g/m³, the maximum value recorded was 63.1 μ g/m³.

	Alt.	Total no. of cases	No. of	cases v	with	Cumulativa	Frequen	cy % - o	f cases	Cumulativ	Maximum value PM ₁₀ /	
Station name	(m)		4	5 s	6	Cumulative	4	5 5	6	е	Date / Synoptic type	
			-	5	Ŭ	FMFP	-	5	Ū		eynopile type	
EM 3– Pojana Stampej	910	2116	218	7	0	225	10,3	0,3	0,0	10,6	63,1 / 15.12.2012 / Sa	
						Industrial	,	,			,, ,	
SV 2 – Suceava 2	289	2972	1008	260	36	1304	33,9	8,8	1,2	43,9	168,3 / 22.12.2010 / Sc	
IS 3 –Tătărași	64	3120	627	170	32	829	20,1	5,5	1,0	26,6	231,5 / 05.11.2015 / SEa	
NT 2 – Roman	207	2425	485	94	5	584	20,0	3,9	0,2	24,1	139,1 / 04.02.2011 / Wa	
NT 3 – Tașca	558	1799	437	96	7	540	24,3	5,3	0,4	30,0	177,2 / 01.02.2017 / H	
BC 2 – Bacău 2	157	3618	675	86	8	769	18,7	2,4	0,2	21,3	125,5 / 05.11.2015 / SEa	
BC 3 – Onești	200	3478	591	106	13	710	17,0	3,1	0,4	20,5	205,4 / 25.12.2009 / SWc	
GL 4 – Galați	45	2202	178	0	0	178	8,1	0,0	0,0	8,1	48,4 / 04.01.2020 / NWa	
GL 5 – Tecuci	31	1259	211	13	0	224	16,8	1,0	0,0	17,8	73,0 / 26.11.2009 / Wa	
Traffic												
SV 3 – Siret	302	2581	788	125	9	922	30,5	4,8	0,4	35,7	124,0 / 10.01.2013 / SWa	
IS 1 – Podu de Piatră	40	3649	1911	661	81	2653	52,4	18,1	2,2	72,7	285,7 / 04.03.2011 / H	
IS 6 – Bosia Ungheni	34	2300	746	258	18	1022	32,4	11,2	0,8	44,4	170,4 / 19.09.2018 / H	
GL 1 - Galați	51	2570	278	11	0	289	10,8	0,4	0,0	11,2	80,4 / 04.01.2020 / NWa	
						Urban						
BT 1 – Botoșani 1	168	3242	1121	222	16	1359	34,6	6,9	0,5	41,9	142,2 / 04.11.2015 / H	
SV 1 – Suceava 1	373	2793	593	63	9	665	21,2	2,3	0,3	23,8	155,7 / 10.02.2014 / SWc	
IS 2 – Decebal Cantemir	42	1374	511	181	7	699	37,2	13,2	0,5	50,9	149,5 / 19.12.2019 / Wa	
NT 1 – Piatra Neamț	357	2803	490	62	9	561	17,5	2,2	0,3	20,0	170,9 / 08.02.2014 / SWa	
VS 2 - Huși	80	1695	586	3	0	589	34,6	0,2	0,0	34,8	56,4 / 20.07.2011 / L	
VS 1 – Vaslui	107	2716	524	56	1	581	19,3	2,1	0,1	21,4	114,4 / 24.12.2009 / SWc	
BC 1 – Bacău 1	169	2463	356	25	3	384	14,5	1,0	0,1	15,6	102,1 / 08.11.2015 / NWa	
GL 2 – Galați	33	1248	90	5	0	95	7,2	0,4	0,0	7,6	63,4 / 16.11.2020 / Sa	
						Suburban						
IS 5 – Tomești	37	2277	718	59	5	782	20,8	1,7	0,1	22,7	182,8 / 05.11.2015 / SEa	
GL 3 – Galați	68	1986	359	32	0	391	14,1	1,3	0,0	15,4	61,8 / 04.01.2020 / NWa	
						Rural				-		
IS 4 – Aroneanu	186	3452	752	216	15	983	33,0	9,5	0,7	43,2	124,5 / 10.02.2014 / SWc	
VN 1 – Focșani	45	2539	144	11	0	155	7,3	0,6	0,0	7,8	83,7 / 13.01.2017 / SWa	

Table 5 Absolute frequency (number of daily cases) and relative (%) of quality indices of classes 4, 5 and 6 for the pollutant PM_{10} at stations in Moldova (2009-2020).

Acronyms: a – anticyclone, c – cyclone, H – maximum pressure region, L – minimum pressure region;

Industrial stations. The highest cumulative average frequencies, bad and very bad quality indicators were recorded at SV-2 Suceava station (43.9%), due to the industrial activity of the company *Romcarton* and *Ambro SA Suceava* (mercaptan pollution – these pollutants are added in the combustible gases, as they helps to signal the accidental loss of gas; they have an extremely unpleasant, suffocating, unbearable smell, which affects the respiratory tract), *Pro – Metal Mechanical Processing Suceava, Suceava Wastewater Treatment Plant, SC Termica SA* etc. (Table 5).

At the NT-3 station, the highest frequencies were recorded for specific indicators of average, bad, and very bad air quality (540 cases, respectively 30%), due to the industrial activity of the *Heidelberg Cement Factory*. In Moldova Region, one of the maximum values of the PM₁₀ pollutant was recorded here (177.2 μ g/m³ - February 1, 2017). The industrial activities on the *Bacău industrial platform*, even if in the last ten years the production decreased (*SC Sofert SA, SC Letea SA, Thermoenergy Group SA*) influence the air quality. At the BC - 2 station a number of 769 cases were registered for the average, bad and very bad quality index and a frequency of 21.3%. At the BC - 3 station, 591 cases with an average specific index, 106 cases with a bad specific index and 13 **GEOREVIEW 33.1** (95-117)

cases with a very bad specific index. These values are due to the emissions of pollutants: phenols, chlorine, hydrochloric acid (*SC CAROM SA*). The maximum value of 205.4 μ g/m³ of the pollutant PM₁₀ was recorded at this station on December 25, 2009. The cumulative frequency of 26.6% for average, bad and very bad quality indices at the IS-3 station is due to traffic from proximity, home heating in personal or centralized system, constructions and demolitions and reduced industrial activities: *SC Fortus SA, SC CET SA, SC Ceramica SA, SC Moldomobila SA* etc. The maximum value of PM₁₀ concentration at the IS-3 station exceeded 200 μ g/m³ (231.5 μ g/m³ on November 5, 2015).

Traffic type stations. At the IS-1 traffic type station, the highest cumulative frequencies were recorded for the specific average, bad and very bad air quality indices (72.7%), due to the heavy traffic that occurs in the vicinity of the Stone Bridge, and other anthropogenic activities including the home heating. To this one can add the relief configuration, the Bahluiului Valley, which favours the stagnation of cold air masses and the accumulation of PM₁₀ suspended dust concentrations. This is the only station in Moldova where the number of cases and the cumulative frequency of bad and very bad air quality indices exceeds 20% (Table 5). At the SV-3 station, high cumulative frequencies of average, bad and very bad quality indices were recorded (35.7%) due to the heavy road traffic on the European road E85 and the location in the Siretului valley. Cumulative frequencies of average, bad and very bad quality indices of 44.4% at station IS – 6 are due to industrial activities (more than 42 enterprises) across the Republic of Moldova border in the city of Ungheni, which is the 6th city as size in this country and the largest in the Central Moldova Development Region. At traffic-type APM stations, the maximum values of PM₁₀ concentrations exceeded the value of 100 μ g/m³ (IS-1 – 285.7 μ g/m³ on March 4, 2011, IS-6 – 170.4 μ g/m³ on September 19 2018, SV-3 - 124.0 µg/m³ on January 10, 2013). These elevated PM10 levels were associated with specific meteorological conditions from a meteo-synoptic perspective, manifesting on both a local and continental scale, and they coincided with pronounced thermal inversions.

Urban stations. Station BT - 1 had a higher number of cases for average, poor and very poorquality indices than the other stations. This is due to the fact that this station attains the longest record of valid data (3242) compared to SV - 1 (2753), NT - 1 (2803), VS - 1 (2716). The high frequency of these indices is due to human activities (demolitions, new constructions, sweeping, household shaking, fires), accidental fires, home heating in individual or centralized system, to which we can add gas emissions resulting from the combustion process (*Nistor et al., 2020*). At urban stations, the frequencies of average, bad and very bad specific indices increase from October to March, as air temperature values decrease and atmospheric pressure increases. These meteorological conditions favour the accumulation and stagnation of PM_{10} concentrations at ground level. Also, during this period, home heating systems are also activated (Table 5).

Suburban stations. The higher cumulative frequencies of the average, bad and very bad indices recorded at the IS – 5 Tomești station (22.7%) were primarily due to the fact that most houses are heated in individual system, such as wood or coal stoves. None of the counties of Moldova exceeded the threshold of 50% for houses with centralized heating. However, Iaşi County shows the highest percentage of 47%. In terms of air quality protection, the situation is critical, because winter thermal regime is more severe in Moldova (Table 5). The maximum value of PM_{10} concentrations exceeded 150 µg/m³ at IS-5 station (182.8 µg/m³ was recorded on November 5, 2015, a high value similar to that recorded at IS-3 station. This happened under a southeasterly anticyclonic circulation and also considering the configuration of the relief – Bahlui Valley which favours the stagnation of air masses. Wood stoves emit between 10 and 180 grams of carbon monoxide for every kilogram of wood burned. Carbon monoxide has serious effects on health because it mixes with the blood in the body (Report-RO-18-june-01.pdf (fppg.ro). Wood burning releases in the atmosphere aldehydes, polycyclic aromatic hydrocarbon compounds (known as

PAHs), volatile compounds (<u>https://www.meteorologiaenred.com/</u>). Coal is considered the most polluting source. Coal burning it releases suspended dust, carbon dioxide, nitrogen oxides, sulphur dioxide, persistent organic pollutants and a series of heavy metals - mercury, lead, arsenic, cadmium, with a negative impact on people's health and the surrounding environment.

Rural stations. At IS-4 station, there were high cumulative frequencies for specific average, bad and very bad air quality indices of 43.2%. This happens because air transports pollutants from the fuel combustion to the high atmosphere (upper troposphere and stratosphere)(Table 5). According to the statistical data provided by INSSE, the highest passenger flows and the highest number of flights were recorded in the lași, airport followed by Bacău and Suceava (https://insse.ro/cms/ro/content/transportul-aeroportuar-of-passengers-%C5%9Fi-m%C4%83rfuri-<u>%C3%AIn-the-year-2022</u>). This determined the relocation of the continuous air quality monitoring station (IS-4) from the centre of the city (Copou district), closer to airport (Aroneanu town). The main aim of this relocation was to evaluate the influence of air traffic on atmospheric pollution with pollutants resulting from the combustion process.

IV.2.3. Monthly / seasonal frequency and station types of air quality indices related to PM_{10} focused on quality indices of classes 6, 5 and 4

The frequency of daily air quality indices reveals that traffic-type stations record high values at bad and very bad indices and low values at excellent and very good indices due to pollutants emitted by road traffic. This occurred especially when car traffic was intense during morning, at noon and in the afternoon the population resumes its activity during the week – students at school, the movement of people to work, in the period before the holidays). The Car tires during the start/stop of the car moves the dust from the street and thus, constitute an additional source of emission of PM₁₀ mobile dust. The substances emitted by road traffic are numerous: mobile dust, carbon monoxide, nitrogen oxides, lead, aldehydes, carcinogenic organic substances (dioxin, benzopyrene), carbon dioxide, volatile organic compounds. By burning one litter of gasoline, 275 g of carbon monoxide, 13.5 g of nitrogen oxides, 24 g of aromatic hydrocarbons with a carcinogenic effect, 1.5 g of suspended particles are eliminated. (*Dițoiu and Holban, 2005*). EMEP and rural stations record low values of bad and very bad air quality indices, due to low road traffic (Figure 4).

In the cold season, the average, bad and very bad quality index values are high at all APM stations in Moldova. The very bad index is registered in the warm season (April - September) only at traffic type stations (0.22%). The bad index has values above 5% in the cold season (October – March) at industrial, traffic, urban and suburban type stations with values between 11% at traffic type stations and 6% at suburban type stations. In the cold season, the average quality index is higher at all APM stations, and the cumulative frequency of average, bad and very bad indices exceed 30% at traffic (42.3%), industrial (36.1%), urban type stations (35.5%) and suburban (29.9%). In the warm season, cumulative frequencies of over 80% of the excellent, very good and good indices were recorded at the APM stations in Moldova, with the exception of the traffic type stations (at the EM-3 station, the cumulative frequency of indices 1 - 3 is 96%).

All these values are closely related to the configuration of the relief (traffic and industrial stations are located in the valley) and to the synoptic situations. From a climatological point of view, periods of great atmospheric stability have been observed in the region of Moldova, when the phenomenon of fog appears, which is frequently accompanied by precipitation. Temperature inversions of thermal origin (nocturnal radiation inversions) are responsible for the greater

number of foggy days, which correlated with combustion emissions lead to increases in PM_{10} concentrations in the morning and evening. It was observed that in the autumn and early winter months (October, November and December) there were persistent thermal inversions that led to the maintenance of high levels of PM_{10} suspended particle concentrations in the atmosphere. On the other hand, against the background of the production of significant episodes of precipitation in the cold season (precipitation in the form of snow contributed even more to the removal of mobile particles from the atmosphere, having a depolluting role) the average values of PM_{10} concentrations were very low (*Nistor et al., 2020*).

The pollution problems faced by the study region also start from the high frequency of days with atmospheric calm at the local level. Thus, during these days, the dispersion of pollutants is very low, the calm being associated with the lack of precipitation for long periods of time.





EMEP – a

industrial – b











suburban - e



Figure 4 Frequency (%) of daily air quality indices (classes 1-6) for each month separately for the PM_{10} indicator at EMEP station - EM3 – Poiana Stampei - a and at *industrial* type stations – b, *traffic* – c, *urban* – d, *suburban* - e and *rural* - f from Moldova (2009-2020).

4.2.4. Frequency by month and station types of air quality indices related to PM_{10} focused on quality indices of classes 6 and 5

The value of 50 μ g/m³ of PM₁₀ concentrations represents the daily limit value for the protection of human health (Law 104/2011). Above this value, the air is considered bad (50 – 100 μ g/m³) or very bad (above 100 μ g/m³). Frequencies of very poor-quality index above 1% were recorded at IS-1, SV-2 and IS-3 stations and frequencies of poor quality index above 11% were recorded at IS-1, IS-2 and IS stations -6. The highest cumulative values of the bad and very bad indices exceeded 10%, with percentages ranging from 20.3% in IS-1 to 10.0% in SV-2. Very low frequencies of the very bad index were recorded at stations EM-3, GL-1, GL-2, GL-3, GL-4, GL-5, VS-2, VN-1 (0%) (Table 6).

The frequency of the specific air quality index bad and very bad was due to several causes: heavy traffic responsible for exhaust gas emissions for stations *IS-1, IS-6, SV-3 and GL-1,* industrial activities for stations *IS-3, SV-2, NT-3, BC-3, NT-2, BC-2, GL-4,* human activities (demolition, street sweeping, household sweeping, smoking), accidental fires, home heating in own or centralized system (heating with individual apartment gas centrals and especially with stoves fuelled with wood, coal, pieces of furniture, PAL, PFL, especially in the cold period of the year, starting from October - November until April). In this interval, due to the combustion process of fuels, the concentrations of pollutants PM₁₀, CO, NO_X, SO₂ increase, for urban type stations (*BT-1, IS-2, NT-2, SV-1*) and suburban type (*IS- 5*).

The cumulative monthly values of the bad and very bad quality indices reveal the following aspects:

- for the *urban type* stations, the months with the highest production frequencies of indices 5-6 (over 5%) were recorded in the months of the cold season, the exception being station IS-2, where they occurred almost all year (less in the months of June and July), due to the high density of blocks, buildings and institutions in the centre of cities and the human activities in close correlation with them (home heating in own or centralized system, car / rail traffic, economic, social and cultural activities);

- for the *industrial type* stations, the months with the highest frequencies of indices 5-6 were those of the cold season (January, February, March, October, November, December), because

most of the industrial type stations are located in the valleys of some rivers (Suceava, Bahlui, Siret, Trotuş, Bicaz, Bârlad), where the role of meteorological conditions is important (atmospheric calm and foggy conditions favour the accumulation of pollutants on the soil surface), thus determining high concentrations of PM₁₀ pollutants;

Quality index bad and very bad	APM Station	1	2	3	4	5	6	7	8	9	10	11	12	YEAR
EMEP	EM-3	1.0	0.6	0.9	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	1.5	0.3
	SV-1	5.5	7.5	2.4	0.0	0.0	0.0	0.0	0.0	0.4	4.1	6.7	5.2	2.6
	BT-1	15.1	12.6	8.6	3.5	0.0	0.4	1.5	3.0	4.5	9.3	12.7	14.0	7.3
	NT-1	12.0	7.1	1.4	0.0	0.0	0.0	0.0	0.0	0.0	1.5	5.5	4.6	2.5
	IS-2	30.1	19.6	28.3	14.4	7.4	0.0	0.0	10.7	7.7	22.6	12.2	15.9	13.7
UKBAN	BC-1	2.3	0.0	2.0	0.0	0.0	0.0	0.0	0.0	0.0	1.5	3.4	5.8	1.1
	VS-1	8.0	0.0	0.4	1.8	0.0	0.0	0.0	0.0	0.0	1.2	4.7	9.8	2.1
	VS-2	0.0	0.0	0.0	0.0	0.0	0.0	2.2	0.0	0.0	0.0	0.0	0.0	0.2
	GL-2	1.2	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	1.7	1.7	0.4
	SV-2	28.6	17.3	16.7	7.1	0.4	0.0	0.8	2.6	5.2	14.0	9.0	21.5	10.0
	NT-2	16.0	6.1	5.9	3.9	0.0	0.0	0.0	0.0	0.0	0.9	3.3	12.9	4.1
	NT-3	28.3	16.4	0.6	0.6	0.0	0.0	0.0	0.0	0.0	0.0	11.2	16.8	5.7
	IS-3	17.6	6.8	6.6	1.9	0.0	0.0	1.6	0.8	2.3	15.1	13.3	14.7	6.5
INDOSTRIAL	BC-2	8.5	1.5	2.6	1.4	0.0	0.0	0.0	0.9	0.0	1.8	5.1	9.1	2.6
	BC-3	12.6	4.6	1.3	0.3	0.0	0.0	0.0	0.0	0.0	3.6	5.0	15.7	3.4
	GL-4	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
	GL-5	5.8	0.0	0.9	0.0	0.0	0.0	0.0	0.0	0.0	0.0	4.3	2.2	1.0
	SV-3	15.4	8.0	7.5	0.5	0.4	0.0	0.0	0.5	1.3	9.9	9.2	10.8	5.2
TRAFFIC	IS-1	24.4	24.8	40.3	26.8	13.5	4.5	5.5	18.1	22.4	27.1	20.6	17.0	20.3
marrie	IS-6	11.1	10.2	21.0	13.4	5.6	2.5	8.7	19.3	15.8	18.0	10.4	7.2	12.0
	GL-1	1.5	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	1.7	0.4	1.3	0.4
RURAL	IS-4	3.2	5.8	3.2	1.3	0.0	0.3	0.6	0.3	1.7	1.6	3.6	1.9	1.9
NONAL	VN-1	7.4	1.9	0.5	0.0	0.0	0.0	0.0	0.0	0.0	1.9	0.0	3.4	1.3
SUBURBAN	IS-5	21.6	10.7	14.2	6.9	2.0	0.0	3.4	12.2	17.8	13.5	8.7	10.8	10.1
SUBURBAN	GL-3	1.2	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	2.9	0.6	1.5	0.6

Table 6 Cumulative frequency (%) by month and types of stations of bad and very bad quality index at APM stations in Moldova (2009 – 2022).

- for the *traffic type* stations, high frequencies for air quality indices 5-6 occurred all year round (except for the SV-3 station where they were recorded only in the cold season), due to the inadequate technical condition of the transport routes (deficit investments in road infrastructure) from the Moldova Region, combined with the delay of spring cleaning actions, against the background of unfavourable weather conditions, which led to the recording of high values of PM₁₀ concentrations;

- for the *suburban type* stations, the months with the highest frequencies of indices 5-6 were those of the cold season, because in the counties of Moldova, most homes were heated in an individual system (wood stoves, coal, gas plants), and in winter the thermal regime is more severe than other areas of Romania, affecting the air quality regime;

- in the case of *rural type* stations, the highest frequencies for indices 5-6 were recorded in the months of January, February, October and November, due to the activities specific to the autumn season around households and in the fields (burning of vegetable remains, garbage), heating of homes (central plants and stoves on wood, pellets, coal, gas) and transport activities (IS-4 station is located near the Iași International Airport, for monitoring pollutants from the burning of fuels used by airplanes).

4.3. Correlations between PM₁₀ concentration and meteorological elements

The Pearson correlation coefficient is often used to measure the statistical relationship and association between different time series of analyzed elements (in our case PM_{10} and GEOREVIEW 33.1 (95-117)

meteorological elements). The Pearson correlation coefficient provides us with valuable information regarding certain indicators we track (https://btprovider.com/ro/coeficient-corelatie-pearson-tableau/). For example, we can measure how close the connection is between the concentration levels of a pollutant (PM_{10} , NO_2 , CO, O_3) and the daily values of a meteorological element (air temperature, atmospheric pressure, atmospheric precipitation, relative humidity, solar radiation, speed the wind). The relationship of direct or inverse proportionality between these two components is given by the sign of the correlation index. The closer the value of the Pearson correlation coefficient is to 1 (in absolute value), the greater the "intensity" of the linear relationship between the 2 variables. The Pearson correlation coefficient (r) can vary between the thresholds of -1 and 1, and its values can be interpreted using the information in Table 7 (<u>https://phys.ubbcluj.ro/</u>).

Table 7 Thresholds of r and the type of correlation indicated by r in different situations.

Pearson's correlation coefficient (r)	Type of correlation
-0,25 < r < 0,25	Weak or no correlation
-0,50 < r < -0,25 // 0,25 < r < 0,50	Acceptable correlation
-0,75 < r < -0,50 // 0,50 < r < 0,75	Moderate to good correlation
r<-0,75 // r>0,75	Very good correlation
r = -1 // r = 1	Error

Table 8 Pearson correlation matrix (Air quality index I5-6) at APM stations in Moldova (2009 - 2020).

	CORRELATION MATRIX (PEARSON): PM_{10} + CLIMATIC ELEMETS (I 5 - 6)													
Name	Station type	т	UR	Рр	Р	R	W							
EM-3	EMEP	0.20	0.71	0.52	-0.02	-0.42	-0.78							
BC-2	Industrial	-0.15	0.11	-0.05	0.13	-0.19	-0.18							
BC-3	Industrial	-0.17	-0.04	0.14	-0.08	-0.24	-0.09							
GL-5	Industrial	0.21	0.18	0.20	-0.31	-0.16	-0.68							
IS-3	Industrial	-0.11	0.16	-0.09	-0.03	-0.10	-0.08							
NT-2	Industrial	-0.29	0.00	-0.01	0.12	-0.01	-0.17							
NT-3	Industrial	-0.22	0.04	-0.05	0.24	-0.04	-0.21							
SV-2	Industrial	-0.34	0.15	-0.05	0.10	-0.21	-0.09							
IS-4	Rural	-0.33	0.30	0.03	-0.35	-0.33	-0.18							
VN-1	Rural	-0.60	-0.10	0.31	0.13	-0.04	0.21							
GL-3	Suburban	-0.17	-0.02	-0.02	0.07	-0.22	0.40							
IS-5	Suburban	-0.26	0.15	0.00	0.12	-0.28	-0.14							
GL-1	Traffic	-0.24	0.41	-0.23	-0.38	-0.25	-0.36							
IS-1	Traffic	-0.26	0.18	-0.05	-0.14	-0.14	-0.26							
IS-6	Traffic	-0.03	0.06	0.01	0.00	-0.13	-0.28							
SV-3	Traffic	-0.30	0.09	0.02	0.08	-0.17	-0.25							
BC-1	Urban	0.13	0.08	-0.17	0.13	-0.09	0.03							
BT-1	Urban	-0.34	0.15	-0.06	0.06	-0.15	-0.14							
GL-2	Urban	0.89	0.44	-0.51	0.24	-0.74	-0.29							
IS-2	Urban	-0.33	0.23	-0.07	0.26	-0.29	-0.35							
NT-1	Urban	-0.33	0.28	0.19	-0.07	-0.32	-0.33							
SV-1	Urban	-0.19	-0.02	0.00	-0.18	-0.06	-0.07							
VS-1	Urban	-0.30	0.06	0.06	-0.01	-0.21	-0.25							
VS-2	Urban	-0.12	-0.84	0.14	-0.23	-0.96	0.60							
	Average	-0.15	0.12	0.01	-0.01	-0.24	-0.16							

Where: T - Air temperature; UR - Relative humidity; Pp - Atmospheric precipitation; P - Atmospheric pressure; R - Global radiation; W - Wind speed;

The highest values of the Pearson correlation matrix related to the bad and very bad air quality index are recorded at the GL-2 station (PM_{10} and air temperature: +0.89; PM_{10} and solar radiation: -0.74; PM_{10} and atmospheric precipitation: -0.51). Other high values are recorded at stations EM-3

 $(PM_{10} \text{ and relative humidity: +0.71; PM_{10} and atmospheric precipitation: +0.52; PM_{10} and wind speed: -0.78), VS-2 (PM_{10} and relative humidity: -0.84; PM_{10} and solar radiation: -0.96; PM_{10} and wind speed: +0.60). At the GL-4 station, no bad and very bad index values were recorded (Table 8). The values of the Pearson correlation matrix vary at the APM stations in the Moldova Region, due to the physical-geographic and socio-economic factors, there being no uniform correlation of this coefficient at the regional level, a fact confirmed by the average values.$

Correlative links of PM_{10} concentrations with meteorological elements are supported only at certain stations by Pearson correlation coefficients, which have values close to ±1. Most frequently, these coefficients have values less than ± 0.5, indicating that PM_{10} levels from the atmosphere are not strongly influenced by meteorological factors, but rather by the diurnal and annual rhythm of socio-economic activities (except for the cross-border station EMEP EM-3) (Figure 5).

The correlation between PM_{10} and meteorological elements (air temperature, air humidity, atmospheric precipitation, atmospheric pressure, wind speed, solar radiation) can be seen from the analysis of the PCA graphs (Principal Component Analysis) as follows:

- the sharp angles reflect strong positive correlations between PM_{10} and meteorological elements (Examples: strong positive correlative links were created between PM_{10} and relative humidity, respectively atmospheric precipitation - Table 8 and Figure 5 a, d, f);

- the obtuse angles and especially the opposite segments with which PM_{10} forms an angle of 180° or close to it reflect negative correlations between PM_{10} and meteorological elements (Examples: strong negative correlative links were created between PM_{10} and wind speed, respectively global radiation – Table 8 and Figures 5 a, b, c, e);

- right angles reflect the lack of correlations between PM₁₀ and meteorological elements.

The correlation between PM_{10} concentration values and air temperature is negative and shows an inverse proportional relationship between temperature and PM_{10} . In the hours and days when high temperature values were recorded, the levels of PM_{10} in the atmosphere decreased, and in the intervals with low temperature values, the concentrations of PM_{10} mobile dust increased. The same situation can be observed in the case of the correlation of PM_{10} pollutant values with solar radiation. Regarding the correlation between the concentration of PM_{10} mobile dust and atmospheric pressure, it is a positive one: when the atmospheric pressure is high, the concentration level of PM_{10} . The same is true of the correlation between relative humidity and the pollutant PM_{10} . The correlations between wind, precipitation and PM_{10} values are negative (inversely proportional): the higher the wind speed and the higher the amount of precipitation, the lower the concentrations of the PM_{10} pollutant (they are either dispersed by the wind or washed away by precipitation). PM_{10} concentrations register high values in the cold season, when the air temperature drops, the air is cold and dry and the atmospheric pressure is high, against the background of thermal inversions and also when the fog phenomenon prevails.

At each station where we analysed PCA, we were able to identify links between PM_{10} and physicalgeographical and socio-economic factors, resulting in different correlations. The EMEP type station is located at high altitude (910m), about 800m from the E58 road, with low economic activities (positive correlations between PM_{10} and relative humidity/atmospheric precipitation and negative correlations between PM_{10} and solar radiation and wind speed). The SV-1 urban station is in the Zamca neighbourhood, between the blocks, in the high area of Suceava city, with intense economic activities (positive correlations between PM_{10} with relative humidity and atmospheric



pressure and negative correlations between PM₁₀ with solar radiation and air temperature) (*Lazurcă*, 2015).

Figure 5 Principal Component Analysis PCA (Quality air index 5-6) – APM stations from Moldova (2009 - 2020).

The BC-2 industrial type station is in the Letea industrial area, approx. 150 m from the Bistrița valley, with intense economic activities (positive correlations between PM₁₀ with relative humidity and atmospheric pressure and negative correlations between PM₁₀ with solar radiation, air temperature and wind speed). The IS-1 traffic station is in the Podul de Piatră area, approx. 100 m from the Bahluiului valley and approx. 200 m from the Podul de Piatră roundabout, at the E58 intersection with the National Road (positive correlations between PM₁₀ with relative humidity and atmospheric precipitation and negative correlations between PM₁₀ with wind speed). The IS-5 suburban station is in Tomești, in the Bahluiului meadow, approx. 1.4 km from CET Holboca (positive correlations between PM₁₀ with relative numidity and atmospheric precipitation and negative humidity and atmospheric precipitation and negative correlation, air temperature and wind speed). The IS-5 suburban station is approx. 2 km N of Iasi International Airport, in the village of Aroneanu (positive correlations between PM₁₀ with relative humidity and atmospheric precipitation and negative correlations between PM₁₀ with relative humidity and atmospheric precipitation and negative correlations between PM₁₀ with solar radiation, air temperature and wind speed). The IS-4 rural station is approx. 2 km N of Iasi International Airport, in the village of Aroneanu (positive correlations between PM₁₀ with relative humidity and atmospheric precipitation and negative correlations between PM₁₀ with relative humidity and atmospheric precipitation and negative correlations between PM₁₀ with relative humidity and atmospheric precipitation and negative correlations between PM₁₀ with relative humidity and atmospheric precipitation and negative correlations between PM₁₀ with solar radiation, air temperature and wind speed).

Meteorological conditions can be critical in limiting or increasing the concentration of PM_{10} and can influence the crossing of certain thresholds that can affect human health.

5. Conclusions

The annual average concentrations of PM₁₀ from the APM stations in the Moldova Region were generally within the legally allowed values (below 40 μ g/m³). The highest multiannual average (2009 – 2020) was calculated for station IS-1 (41.2 μ g/m³). This average exceeds the annual allowable limit value and throughout the studied interval at this station values above or close to the annual allowable limit were recorded due to heavy road traffic. Other high multi-year averages were calculated for stations IS-2 (32.5 μ g/m³), IS-6 (32.1 μ g/m³) and SV-2 (31.6 μ g/m³), traffic-type stations or industrial. The lowest multiannual averages of atmospheric PM¹⁰ levels were calculated for EM-3 (16.9 μ g/m³; cross-border station, located at the altitude of 891 m) and the stations in the city of Galati: GL-2 (14.9 μ g /m³), GL-3 (16.7 μ g/m³) and GL-4 (17.5 μ g/m³) (after 2000, the activity of the ArcelorMittal Galați steel plant was reduced, production gradually decreased, and thus, the level of pollution decreased drastically).

The highest values of PM_{10} dust concentrations were recorded at industrial and traffic type stations, due to pollution sources (industrial activities, traffic) and also in the valley lanes of Suceava (SV-2), Siret (SV -3, NT-2, BC-2), Bahlui (IS-1, IS-2, IS-5), Bicaz (NT-3), Trotuş (BC-3) rivers, which due to the configuration of the relief favours the accumulation and stagnation of particles PM_{10} . The highest monthly averages of PM_{10} concentration were calculated for the cold season months (mostly October, November, February, March) and the lowest were calculated for the late spring and summer months (May – August). The maximum values of PM_{10} concentrations occurred at industrial or traffic type stations, during the cold season, under the conditions of an anticyclonic synoptic circulation or a high-pressure field, against the background of accentuated thermal inversions, which generate stable but cold weather and frosty.

In the period 2009 – 2020, the highest cumulative values of the bad and very bad indices exceeded 10%, (between 20.3% in IS-1 and 10.0% in SV-2). The highest frequencies of quality indices 4–6 (average, bad and very bad) was recorded at the stations in the Iaşi metropolitan area (IS-1, IS-2, IS-6, IS-5), and the smallest at the stations in the Galati metropolitan area (GL-1, GL-2, GL-3, GL-4) and at the EM-3 Poiana Stampei station. The frequency of the daily air quality indices shows that at the traffic type stations the bad and very bad indices had high values, due to the exhaust gas emissions resulting from the transport activity.

In the cold season, the average, bad and very bad quality index values were high at all APM stations in Moldova except EMEP and rural stations. The cumulative frequency of average, bad and very bad indices exceeded 30% at traffic (42.3%), industrial (36.1%), urban (35.5%) and suburban (29.9%) stations. The bad index recorded values above 5% (October – March) at industrial, traffic, urban and suburban stations (with values between 11% at traffic stations and 6% at suburban stations).

In the months of the warm season, the APM stations in Moldova recorded cumulative frequencies of over 80% of the excellent, very good and good quality indices, except for the traffic type stations. The very bad air quality index was recorded during April - September, only at traffic type stations (0.22%).

Correlative links of PM_{10} concentrations with meteorological elements are supported only at certain stations by Pearson correlation coefficients, which must have values close to ± 1. In our study, we observed that frequently, these coefficients have values less than ±0,5, indicating that the concentrations of suspended mobile particles PM_{10} in the atmosphere are not strongly influenced by meteorological factors, but rather by the diurnal and annual rhythm of socio-economic activities (with the exception of the cross-border station EM-3).

The correlation between PM_{10} concentrations and meteorological factors is strong at the EM – 3 station (because it is located at high altitude and there are no industrial, human and transport activities with major environmental impact) and much less at the industrial type stations, urban and traffic, because the anthropogenic factor is the most important.

There are negative correlations between PM_{10} concentrations and solar radiation, air temperature, wind speed and atmospheric precipitation. Positive correlations were established between PM_{10} mobile dust and atmospheric pressure, respectively relative humidity.

Research on the phenomenon of air pollution and the identification of the measures that must be implemented to maintain, protect and improve air quality has gained momentum due to the negative impact of PM_{10} suspended particles on air quality, human health and the natural environment. Therefore, a combined effort of stakeholders (local, regional, national), based on quantitative data obtained from research, seems to be the most effective and sustainable solution to prevent and limit the negative effects due to the increase in PM_{10} concentrations.

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