

Assessment of air pollution due to ozone in the North-East Region-Romania

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Abstract: The results of air quality monitoring in automatic stations in the North-East Region-Romania (RNER) region in the period 2009 - 2020 indicated that the declared ozone concentration values were generally lower than those regulated by law - information threshold $> 180 \mu\text{g}/\text{m}^3$ as required by Directive 2008/50/EC and Law 104/2011. Air quality in RNER relative to ozone concentration was good, with only one exceeding of the information threshold (NT1 - 28 April 2009, 12:00 - $180,28 \mu\text{g}/\text{m}^3$). The highest multi-annual averages (2009 - 2020) were calculated for IS 4 stations ($59,5 \mu\text{g}/\text{m}^3$), EM 3 ($53,4 \mu\text{g}/\text{m}^3$) and SV 1 ($50,3 \mu\text{g}/\text{m}^3$). High values were generally recorded at urban-type stations, where urban characteristics, heavy road transport and the presence of airports generated high ozone concentrations. The lowest multi-annual ozone concentrations were calculated for stations VS 1 ($41,5 \mu\text{g}/\text{m}^3$), BC 2 ($41,9 \mu\text{g}/\text{m}^3$), IS 3 ($42,0 \mu\text{g}/\text{m}^3$) and BC 3 ($42,7 \mu\text{g}/\text{m}^3$). Low values were generally registered at industrial stations, where the atmospheric calm and the frequency of urban haze favored the accumulation of pollutants on the soil surface, limiting the formation of ozone. Ozone pollution is viewed with concern by environmental specialists in RNER, due to the fact that this element has a negative impact on people and the environment.

1. Introduction

Tropospheric ozone is a strong oxidant with a characteristic odor of bluish color and very toxic. This is located approx. 90% in the stratosphere known as “good ozone” and approx. 10% in the troposphere called “bad ozone” (Meteo.md, 2017).

Ozone is formed naturally in the atmosphere by electrical discharges and under the catalytic action of ultraviolet solar rays. There are studies that emphasize the correlation between climate elements (air temperature, atmospheric precipitation, wind regime) and ozone concentration (Baur et al., 2004; Salve et al., 2007; Khiem et al., 2010).

The increase in ozone concentrations usually occurs under the influence of favorable weather conditions (late spring and early summer) for photochemical ozone production in the micro- and topoclimate layer. Ozone levels typically reach values around $45\text{--}60 \mu\text{g}/\text{m}^3$ in the afternoon, and during the night to morning due to the downward movements of the air that drives ozone from top to bottom, towards the basal atmospheric layer and the fact that this downward movement does not compensate for the absence of UV radiation, ozone levels remain low (Kalabokas et al., 2020).

Local geographical factors and meteorological factors/elements greatly influence the genesis, spatiality and temporality of ozone levels in the atmosphere of different places. Local meteorological factors, which have a strong influence on surface ozone concentrations, have interannual variability and are expected to change in the future. This statement is based on studies carried out on this subject, which have covered various areas in Europe: France, the Balkans, the Iberian Peninsula, the Mediterranean Basin, Central Europe, Eastern Europe (Chatzianastassiou et al., 2017; Castell et al., 2008; Cichowicz et al., 2017; Otero et al., 2018). Observations of solar radiation flux density at the earth's surface have shown an upward trend in Europe since the mid-1980s, known as solar "lighting". In general, the impact of increased radiation flux on surface ozone has accentuated the reactions of ozone production by photochemical processes, but also the emissions of biogenic volatile organic compounds (BVOC). Changes in the balance of interaction between aerosols and radiation have positively influenced trends in ground surface ozone levels during summers and have led to partial amortization of the effects of reducing emissions of ozone precursors after 1990 (Oikonomakis et al., 2018).

There is an interrelationship between vegetation and ozone. Following the process of photosynthesis, above the areas occupied by vegetation, large quantities of oxygen and VOCs are released, with an important role in ozone formation. On the other hand, high ozone levels reduce the ability of plants to perform photosynthesis and prevent carbon dioxide absorption. Ozone also hinders plant reproduction and growth, resulting in lower harvests and reduced biomass production (Lamaux et al., 1994; Hunova et al., 2011).

By artificial means, ozone is a secondary pollutant formed by photochemical reactions (UV) in chain from precursors such as nitrogen oxides, volatile organic compounds and carbon monoxide, in which an important role is played by free radicals hydroxyl (OH), organic peroxide and hydroperoxyl (HO₂) (Mihăilă, Piticar, 2021). Land use changes have a positive impact on ozone production by changing ecosystems. In addition, the heat emitted by the surface of the urban land influences the dynamics of the flow of this gas in the atmospheric boundary layer and dynamizes its transport. The sources of precursor gases which contribute to the formation of ozone are mobile (they generate exhaust gases, NO_x, CO and C_nH_m - hydrocarbons, especially present during peak hours of motor vehicle traffic when they are running at a low speed and burns are not complete) and stationary (chemical plants, oil refineries, dye-works, gas stations, etc.) (Mihăilă, Piticar, 2021). Land transport is an important source of emissions of nitrogen oxides, carbon monoxide and volatile organic compounds. The highest absolute contributions of ozone precursor mobile sources occur around Paris, Southern England, Moscow, Athens, Po Valley, Western Germany, central Serbia (Markovic et al., 2008; Malinovic-Milicevic et al., 2008; Psiloglou et al., 2013; Mertens et al., 2020).

Measures to improve air quality, which were implemented in Europe in the 1990s, have led to a reduction in concentrations of ozone precursors. Long-term trends in medium and maximum ozone concentrations are largely controlled by decreasing precursor emissions, while seasonal oscillations and increasing sensitivity of ozone to temperature depend predominantly on regional climate conditions (Lee et al., 2014; Boleti et al., 2020).

Tropospheric ozone is part of the group of greenhouse gases (along with CO₂, CH₄, N₂O), and it is also incriminated for indirect participation in the formation of acid rain and when it is present in the soil, it behaves as a component of "photochemical smog". Tracking of tropospheric ozone concentration is important, known to be a compound harmful to vegetation, animal health and human health (Fenech et al., 2018). Ground level ozone can be dangerous, especially for people with respiratory problems (it causes irritation, lung inflammation, cough, aggravates asthma, pneumonia and bronchitis)

(Jimenez-Horneo et al., 2010; Monforte, Ragusa, 2018). The relationship between exposure of the population to varying environmental concentrations of ozone and public health has been investigated. On the basis of demographic indicators, links have been established between ozone levels and morbidity/mortality of the population (Kassomenos et al., 2018).

There is a growing awareness of human communities regarding the role of ozone as a short-term climate pollutant and that this could be an important direction to follow in the integrated mitigation of climate change, with an impact and in improving air quality. The review of the ozone issue requires the current understanding of the processes that regulate tropospheric ozone on a global/local scale based on measurements as well as models. There is a clear and current need to treat ozone throughout the spatial scalar range (from hemisphere/cross-border to regional/local level), by using both large-range equipment (satellites) and sensors that accurately determine local ozone levels (Monks et al., 2015).

For Romania, there are few studies that analyze the annual and daily regime of ozone concentration. The evolution of tropospheric ozone, the relationship with air temperature and NO_x was highlighted for the Ciuc Depression (Szep et al., 2016; Petres et al., 2018). The daily and annual regime of tropospheric ozone in the municipality of Suceava were described and explained based on the correlations between ozone concentration and local meteorological parameters (Mihăilă et al., 2018). The temporal variability of surface ozone and its precursors (NO_x) was analyzed in the area of the municipality of Iasi, with an emphasis on the influence of meteorological elements on these two important air pollutants (Amihăiesei et al., 2019).

The main sources of pollution that contribute to the genesis of ozone in Romania, and implicitly in the counties of RNER highlight the fact that the energy sector contributes significantly to emissions of ozone precursor pollutants, followed by the transport and agriculture sectors (solvent use contributes significantly with emissions of Non-methane volatile organic compound (NMVOC) (Report on the state of the environment in Romania, 2018; 2019).

The study aims to analyze the spatial distribution and ozone concentration regime for RNER, with the division of areals where air quality needs to be maintained and the identification of those areals where air quality needs to be improved.

The objectives of this paper are: i) analysis of ozone concentrations in the period 2009– 2019 for RNER, ii) highlighting the relationships among meteorological factors (temperature, precipitation, humidity, atmospheric pressure, wind) and the spatio-temporal distribution of ozone concentrations, iii) identification of the contribution of ozone pollution to the general pollution of the analyzed region.

In the Member States of the European Union (including Romania), the long-term target (2020) was set at a concentration of 120 µg/m³ O₃ as a maximum daily 8 - hour mean (European Environment Agency - EEA, 2009). The level at which the population is informed about ozone is set at an hourly concentration of 180 µg / m³ of O₃, while the alert level has been set at an hourly value of 240 µg / m³ of O₃ (EEA, 2009). Policies that reduce emissions from mobile sources can lead to substantial benefits for human health.

2. Materials and Methods

2.1. Study area

The study area - RNER comprises 6 counties: Suceava, Botoşani, Neamţ, Iaşi, Bacău, Vaslui. The surface of this region covers an area of 36.850 km². The relief is varied as it includes all relief levels and landforms in Romania. In the western counties (Suceava, Neamţ and Bacău) there are mountain subunits belonging to the Eastern Carpathians,

but also subunits belonging to the Moldavian Subcarpathians. All six counties are composed of subunits of the Moldavian Plateau. In the eastern counties of the region (Botoșani, Iași and Vaslui) there are territories belonging to the Moldavian Plain and the Prut Meadow. The counties of Suceava, Neamț, Iași and Bacău are crossed by the Moldavian and Siret Corridors.

The population of RNER registered a number of 3.923.221 inhabitants on July 1, 2020. The majority of the population lives in rural areas. The largest cities of the region are Iași (362.142 inhabitants), Bacău (196.883 inhabitants), Botoșani (122.311 inhabitants), Suceava (116.404 inhabitants), Piatra Neamț (115.273 inhabitants) and Vaslui (97.067 inhabitants). The density of public roads is 36,3 km/km², being higher in the counties of Iași, Bacău and Suceava, which are crossed by a number of European corridors (E85, E576, E583, E574).

2.2. Research methodology

The research algorithm used by us consisted of: *i*) collecting various information, including by consulting articles and scientific papers on the topic and region studied, *ii*) statistical processing of data held, *iii*) graphical and cartographic transposition of ozone concentration values and *iv*) comparative analysis and interpretation of statistical, graphical and cartographic data on the causal, distributive and evolutionary components. This methodological approach allowed us to obtain quantitative, real and accurate information on the temporal evolution and spatial distribution of ambient air quality, in relationship with their causality.

2.3. Ozone determination method/STAS (Romanian standard)

The reference method for ozone sampling and measurement is that provided in the SR EN 14625 standard: "*Ambient air. Standard method for the measurement of the concentration of ozone by ultraviolet photometry*". UV Photometry / Spectrometry instrumentally measures the light transmitted by a colored (visibly) or colorless (ultraviolet) solution working with a monochromatic light source. In the UV field, the human eye not perceiving light, only spectrophotometry is used. The chemiluminescence technique, associated with flow injection analysis, is particularly sensitive for the determination of environmental pollutants such as ozone, sulfur compounds or phosphorus, etc.

2.4. Means of determination

The UV-VIS spectrophotometer (ultraviolet-visible) is used to determine concentrations of certain precipitation compounds, whose analytical principle is photolorimetry or turbidimetry. These optical/spectral methods allow the determination of the composition, concentration, structure of different substances and materials, including an extremely diverse category of analysis methods and techniques. The Lambert-Beer law is the basis of colorimetric and photolorimetric determinations.

Model GUX-113 Ozone Analyzer made by DKK TOA Japan is another ozone measuring device used by EPA stations in RNER and its measuring principle is also UV photometry.

2.5. Monitoring stations network

In Moldavia, the air quality monitoring system - the ozone parameter, includes 15 stations for assessing the influence of different types of sources on air quality as well as the assessment of background level, far from any source.

There are 5 industrial stations in RNER (SV2, IS3, NT2, BC2, BC3), 7 urban stations (BT1, SV1, IS2, NT1, VS2, VS1, BC1), 1 suburban station (IS5), 1 rural station (IS4) and 1 EMEP type station (EM3), arranged from N to S and from V to E (Table 1 and Figure 1).

Table 1. Location of ozone measuring stations in RNER (2009–2019)

Nume of the station	Type of station	Adress of the station	Latitude	Longitude	Altitude
BT 1 – Botoșani1	Urban	Botoșani, B-dul M. Eminescu, 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 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 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, 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
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

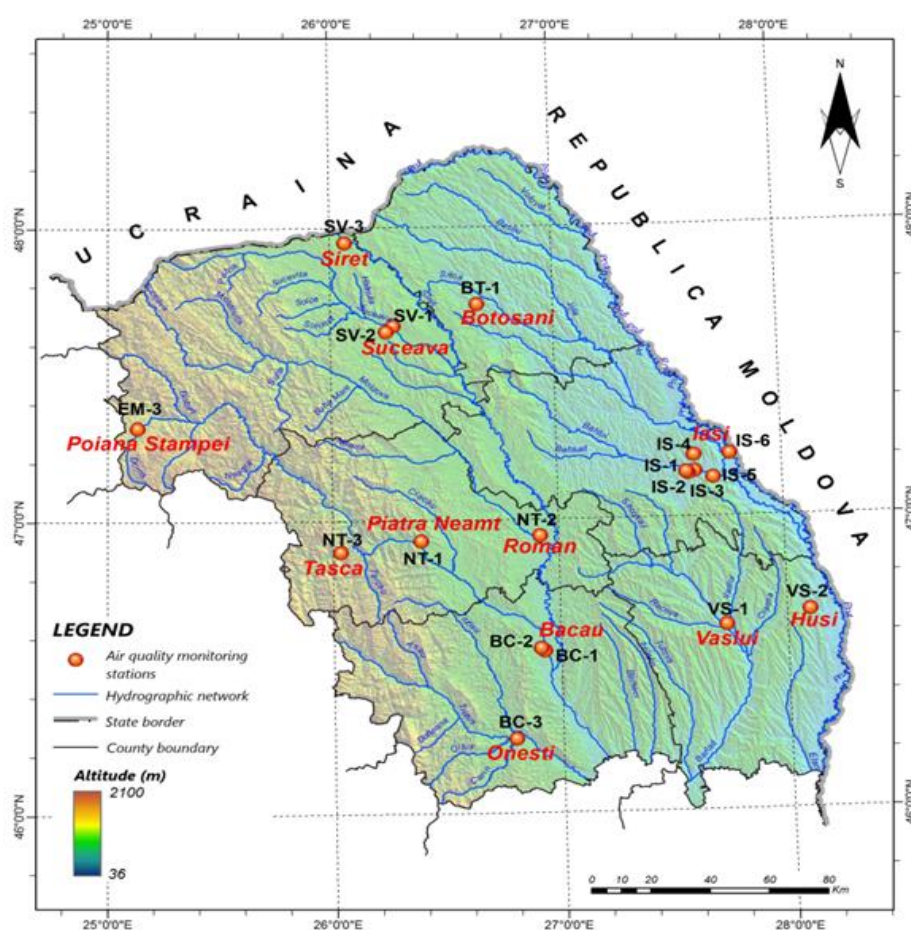


Figure 1. The geographical map of RNER and the air quality monitoring stations

Table 2. Data capture (%) of ozone concentration at EPA stations in RNER (2009–2020)

Code station	Type station of	2009	2010	2011	2012	2013	2014	2015	2016	2017	2018	2019	2020	Mean
SV 3	Trafic	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
BT 1	Urban	97.3	84.7	95.6	95.1	89.6	94.3	91.5	97.5	95.6	97.8	99.2	98.9	94.8
SV 2	Industrial	81.4	35.1	0.0	7.7	0.0	0.0	1.6	17.8	98.4	65.8	76.2	71.6	37.9
SV 1	Urban	84.1	80.8	91.8	90.7	94.3	95.6	98.9	88.8	98.4	80.8	75.6	92.6	89.4
EM 3	EMEP	9.6	84.7	82.2	75.4	64.4	56.7	81.9	73.5	73.7	92.9	49.9	61.5	67.2
IS 6	Traffic /	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
IS 4	Rural	98.1	81.4	97.5	96.7	94.3	93.7	83.8	59.3	91.5	93.4	87.4	88.8	88.8
IS 3	Industrial	97.5	98.1	95.3	95.9	99.2	98.1	98.6	87.2	98.9	97.3	43.3	0.0	84.1
IS 1	Traffic	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
IS 2	Urban	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	50.4	98.9	12.4
IS 5	Suburban	0.0	95.6	96.4	94.0	83.0	92.1	87.4	92.6	37.8	98.9	98.1	74.6	79.2
NT 1	Urban	92.1	66.6	98.1	92.6	96.7	96.2	76.4	92.1	99.7	81.6	64.9	71.6	85.7
NT 2	Industrial	35.1	0.0	16.2	84.2	5.8	0.0	0.6	47.5	98.1	99.2	98.9	66.1	46.0
NT 3	Industrial	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
VS 2	Urban	0.0	0.0	74.3	85.3	46.6	50.4	21.9	83.1	16.7	0.0	0.0	0.0	31.5
VS 1	Urban	92.9	98.9	95.1	53.8	18.6	0.0	52.9	82.8	89.3	96.2	98.4	93.2	72.7
BC 1	Urban	94.3	96.7	61.6	87.4	97.3	79.2	81.1	95.9	90.4	45.5	63.0	93.2	82.1
BC 2	Industrial	85.5	81.1	91.8	91.0	85.8	93.7	91.8	89.1	99.7	98.4	99.7	99.2	92.2
BC 3	Industrial	90.4	95.6	95.2	93.7	96.7	94.5	95.9	96.5	98.9	77.3	70.1	96.7	91.8
LEGEND			Total lack of data				Insufficient data for validity					Valid data capture		

2.6. Data provided and the period analyzed

The air quality monitoring system allows environmental authorities to take prompt action to reduce or eliminate pollution episodes, to inform, warn and protect the population in case of exceedances of ozone concentration. The results of air quality monitoring in the automatic stations belonging to the RNMCA on the territory of RNER, in the period 2009 - 2020, indicated levels of ozone concentrations lower than the limit - values/ information threshold ($> 180 \mu\text{g}/\text{m}^3$) provided by Directive 2008/50/EC and Law 104/2011.

3. Results

3.1. Sources of pollution with ozone precursors in RNER

The main stationary sources that generate ozone precursors (chemical plants, oil refineries, painters, dye-works, dry cleaning facilities, gas stations) are represented by: i) Bacău industrial platform (SC Sofert SA Bacău, SC Letea SA Bacău), Onești chemical platform (SC Chimcomplex SA Borzești, SC Rafo Onești) in Bacău County; ii) Botoșani industrial platform, SC TERMICA Botoșani in Botoșani county; iii) Iași industrial Platform (SC Fortus SA Iași, SC Antibiotice Iași), CET I Iași, CET II Holboca Iași in Iași County; iv) Săvinești chemical platform, Tașca Cement factory - Bicaz in Neamț county; v) Suceava industrial Platform (SC Ambro SA Suceava), Termica Suceava in Suceava County; vi) Vaslui industrial platform in Vaslui county. Some of these operators have reduced/ceased their activity, but on the respective sites other production activities that generate ozone precursors are developed.

The mobile sources (generate exhaust gases, nitrogen oxides, carbon monoxide and hydrocarbons) are the means of transport. The region is crossed by a number of European roads: Bucharest – Bacău – Suceava – Siret (E85), Suceava – Vatra Dornei – Dej (E58), Bacău – Brașov (E574), București – Bârlad – Albița (E581), Roman – Tg. Frumos – Iași – Sculeni (E583) and national and county roads, respectively.

3.2. Interannual regime and the multiannual average distribution of ozone concentration in the monitoring points of this pollutant in RNER

Solar radiation via the UV component is the determining factor for the ozone generation process, to a greater extent than the air temperature. The high positive correlation between temperature and ozone is determined by high radiation values,

which impose high temperature values, the temperature being the agent of ozone-forming solar radiation, especially during hot periods. Air temperature acts as an important catalyzer, directly correlating its interannual / annual / diurnal course with that of solar radiation. In the annual regime, a clear correlation among radiation, air temperature and ozone concentration is observed. In the diurnal regime, there is a delay in the production of the maximum ozone concentration compared to that of the temperature, caused by the delay of VOC emissions by the vegetation in the forested areas surrounding the cities.

The evolutionary and distributive situation of the annual average concentration values calculated for air quality monitoring stations in RNER for ozone (using the valid annual values) is presented in Table 3.

Table 3. Annual and multiannual mean values of ozone concentration at EPA stations in RNER (2009–2020)

Code station	2009	2010	2011	2012	2013	2014	2015	2016	2017	2018	2019	2020	2009–2020
BT 1	45.8	46.0	50.2	48.0	44.6	42.0	44.5	46.7	39.5	44.9	34.1	29.8	43.0
SV 1	52.6	54.5	55.4	60.3	41.1	39.3	54.5	48.2	44.7	51.9	53.3	50.4	50.3
EM 3	34.4	58.8	54.9	58.0	66.3	50.7	46.4	50.7	48.5	49.0	53.1	43.6	53.4
IS 4	58.3	61.7	56.7	65.4	50.8	47.6	68.6	63.6	55.7	66.1	60.2	64.0	59.5
IS 3	42.5	41.1	37.7	47.6	42.0	38.5	42.8	44.6	37.3	46.3	46.3	-	42.0
IS 5	-	42.4	37.7	54.0	39.3	31.5	48.8	47.6	40.4	49.1	44.2	37.8	43.8
NT 1	66.0	22.2	27.1	41.4	43.6	38.5	49.3	51.9	43.4	49.2	49.1	50.8	45.6
VS 1	40.2	36.0	33.7	51.3	52.2	-	63.3	42.7	46.5	43.2	46.6	43.4	41.5
BC 1	32.1	40.7	33.2	48.4	48.8	40.7	41.0	48.7	39.1	54.4	43.6	44.4	42.8
BC 2	37.2	40.3	31.0	52.6	51.7	34.8	42.9	37.5	39.6	48.5	42.1	44.4	41.9
BC 3	39.1	35.4	23.3	46.6	46.1	40.7	44.6	50.1	38.8	51.8	45.2	45.7	42.7

* 53.3 – annual value that does not meet valid data capture; * -- missing data due to revisions, faults in data recording equipment

The highest multiannual average (2009 - 2020) was calculated for IS 4 station (59,5 $\mu\text{g}/\text{m}^3$). Throughout the studied interval, Iași 4 station had high values (out of the 12 years of analysis, in 11 years the highest value of RNER was calculated for IS4). IS4 station currently includes (since 2016) Iași International Airport in the area of representation, in order to determine the influence of air traffic on air pollution with noxious substances resulting from the combustion process (<https://www.anna.aero/databases>).

Based on the data recorded and processed following the monitoring, it was concluded that one of the causes for both this station and the other urban stations is the heavy air traffic at the international airports: George Enescu Airport (Bacău), Iași International Airport and Ștefan cel Mare Airport (Suceava), located in the metropolitan area of the respective cities. Aircraft eliminate in the lower and upper atmosphere (upper troposphere and stratosphere) pollutants from the combustion of fuels (nitrogen oxides, hydrocarbon residues, carbon oxides) that affect the ozone balance. According to the statistical data provided by INS Eurostat, the highest passenger flows and the highest number of flights were recorded at the airport of Iași, followed by Bacău and Suceava (Table 4). Other high multi-annual averages were calculated for EM 3 (53,4 $\mu\text{g}/\text{m}^3$) and SV 1 (50,3 $\mu\text{g}/\text{m}^3$) stations. High values have been recorded at urban stations, where the characteristics of the active surface (high altitudes, urban characteristics, etc.) cause high ozone concentrations.

Table 4. Passenger traffic statistics and number of flights for the year 2019

Airport	Passangers	No. of flights
<i>Bacău</i>	468.383	3.303
<i>Iași</i>	1.312.611	11.656
<i>Suceava</i>	428.974	2.569

The lowest multi-annual averages of ozone concentration were calculated for VS 1 (41,5 $\mu\text{g}/\text{m}^3$), BC 2 (41,9 $\mu\text{g}/\text{m}^3$), IS 3 (42,0 $\mu\text{g}/\text{m}^3$) and BC 3 (42,7 $\mu\text{g}/\text{m}^3$). Low values have generally been recorded at industrial stations, where atmospheric calm and fog conditions favor the accumulation of noxes at the soil surface, which makes ozone formation reactions more difficult. The highest daily multi-annual maximum value (2009 – 2020) was calculated for the NT 1 station (137,57 $\mu\text{g}/\text{m}^3$ – 2009). Other high values were calculated for IS 4 (128,15 $\mu\text{g}/\text{m}^3$ – 2018), IS4 (123,02 $\mu\text{g}/\text{m}^3$ – 2016). These maximum values were caused by heavy car and air traffic at international airports in RNER (Table 5).

Table 5. Daily maximum values of ozone concentration for the years monitored at EPA stations in RNER (2009–2020)

Code station	2009	2010	2011	2012	2013	2014	2015	2016	2017	2018	2019	2020	2009 - 2020
BT 1	87.34	79.18	102.04	92.63	81.35	89.43	86.22	87.51	72.08	95.19	61.29	57.76	102.04
SV 1	115.11	97.09	104.14	96.94	74.60	91.54	97.05	83.49	80.24	91.02	92.64	95.39	115.11
EM 3	55.63	109.90	101.60	109.24	100.92	99.60	80.68	86.55	76.85	88.67	86.26	90.59	109.90
IS 4	116.60	98.55	106.24	112.77	102.66	106.43	111.75	123.02	116.45	128.15	97.37	104.25	128.15
IS 3	84.06	77.97	69.52	106.90	82.70	78.05	96.84	92.79	70.34	99.85	91.02	-	106.90
IS 5	-	77.01	70.16	109.86	68.33	66.46	94.82	84.69	71.85	104.65	83.15	78.70	109.86
NT 1	137.57	52.38	47.88	116.40	95.30	68.95	88.98	104.47	70.98	90.94	102.51	95.87	137.57
VS 1	76.82	66.90	71.65	118.65	86.74	-	96.93	81.55	84.58	104.84	86.10	89.25	118.65
BC 1	65.18	78.31	76.03	100.67	94.81	93.93	79.84	90.54	79.59	87.40	73.98	80.47	100.67
BC 2	70.63	74.19	73.14	120.86	94.56	110.30	82.00	81.68	71.90	106.99	78.46	82.55	120.86
BC 3	73.62	91.64	48.46	94.63	82.90	87.79	83.90	101.80	81.87	117.73	97.72	92.07	117.73

3.3. Annual evolution and distribution (outlined on the basis of monthly average values) of ozone concentration at the monitoring points of this pollutant in RNER

The annual regime of solar radiation and air temperature impose intermonthly differences in ozone concentration. The warm season months, which are characterized by higher values of global radiation and air temperature, have higher values (by 15-30 $\mu\text{g}/\text{m}^3$) of ozone concentrations (Table 6 and Figure 2).

During an average year, many similarities, but also some differences can be established between the air temperature regime and that of the ozone concentration. The maximum values of ozone concentration occur at most stations in spring months (April - May, when the UV flow reaches maximum intensity against the background of maximum atmospheric transparency in the annual profile). Low ozone concentrations occur in the cold season months, especially in November and December, which are foggy months with stratiform clouds, short days and low UV values. Thermal maxima occur a little later, in July-August, between the maxima of the two correlated elements (ozone-temperature) existing a time lag of 1-2 months.

Table 6. Monthly average ozone values at EPA stations in RNER (2009 – 2020)

Code of station	I	II	III	IV	V	VI	VII	VIII	IX	X	XI	XII	Mean
BT 1	31.3	40.6	46.4	52.8	54.8	51.9	53.4	53.7	46.4	32.8	26.6	26.8	43.0
SV 1	44.7	54.0	64.3	62.8	63.2	57.7	56.8	53.1	48.1	44.2	35.4	43.0	50.3
EM 3	40.6	48.2	53.5	62.0	60.5	57.6	59.2	57.8	51.5	38.6	33.3	37.1	53.4
IS 4	45.5	52.0	60.0	69.0	70.2	72.5	73.0	77.8	65.2	49.2	39.4	38.0	59.5
IS 3	30.3	39.5	46.3	54.6	56.0	54.0	55.6	52.7	41.3	29.9	23.6	21.9	42.0
IS 5	33.2	42.0	48.2	54.1	52.1	54.7	53.8	55.0	44.7	31.0	27.5	23.1	43.8
NT 1	36.0	44.6	50.2	55.3	53.5	45.2	48.4	54.9	49.5	36.0	27.3	30.9	45.6
VS 1	33.3	44.3	50.6	55.8	55.2	51.7	51.6	52.2	43.2	29.9	25.2	27.1	41.5
BC 1	33.5	44.1	47.4	52.6	52.5	52.6	52.7	52.3	44.6	28.4	24.8	23.4	42.8
BC 2	35.1	44.8	48.1	52.1	51.5	49.4	48.6	48.7	42.5	31.2	25.3	23.3	41.9
BC 3	39.3	43.9	51.3	49.5	48.5	47.4	48.7	46.0	43.3	33.1	28.1	29.4	42.7

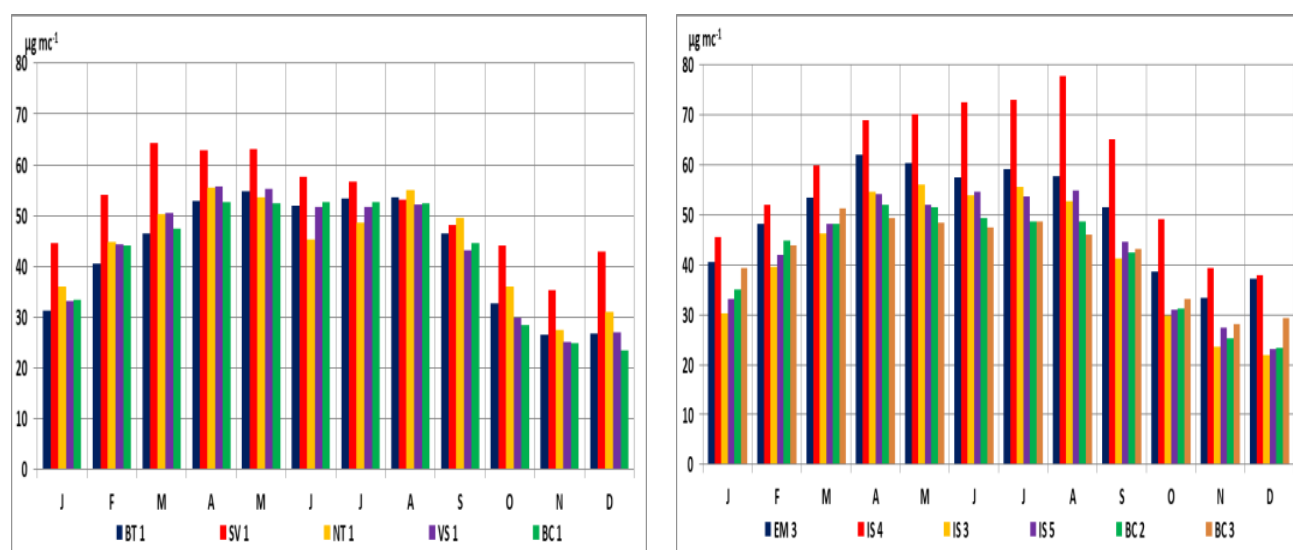


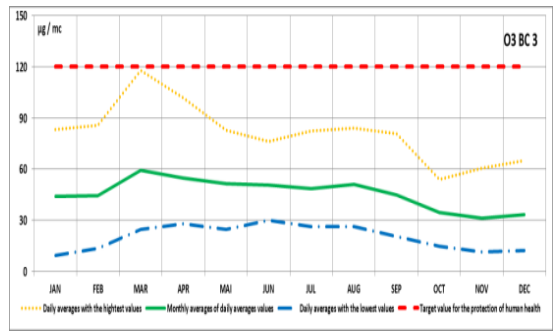
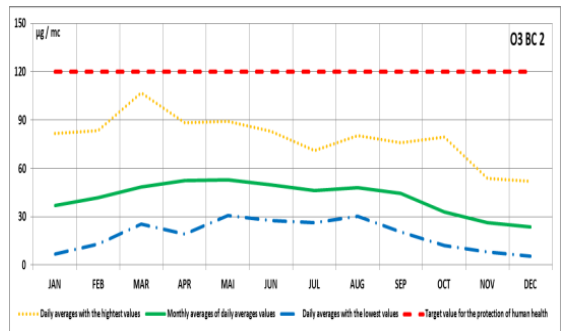
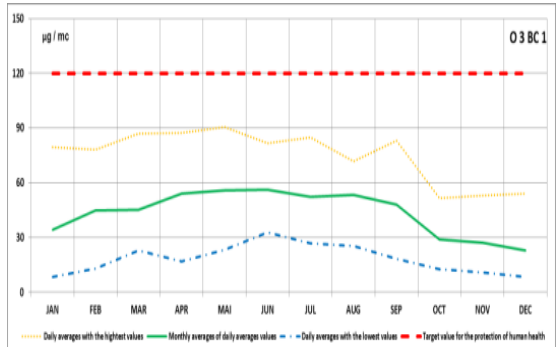
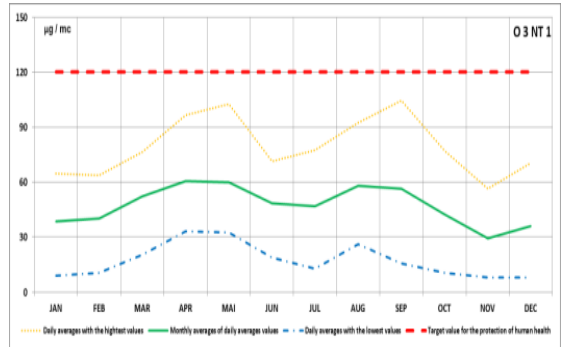
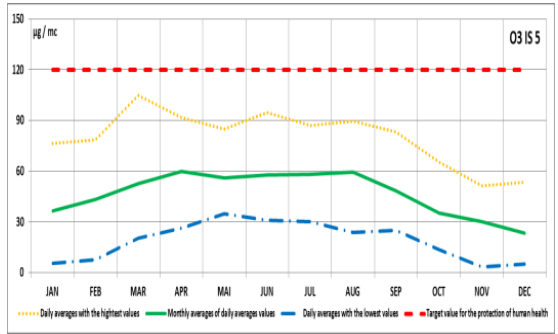
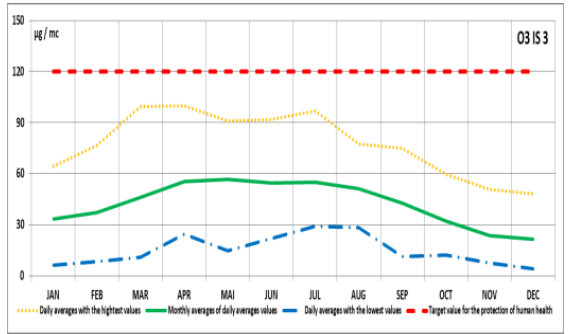
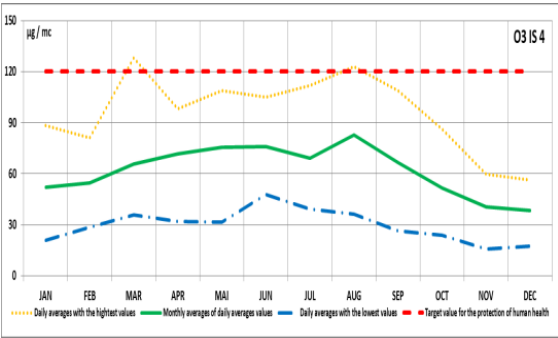
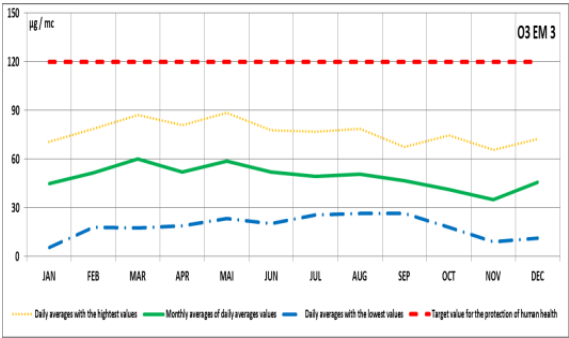
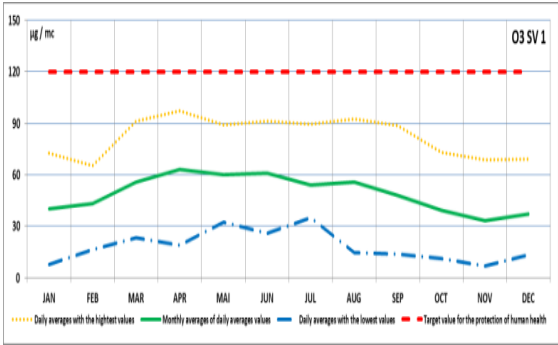
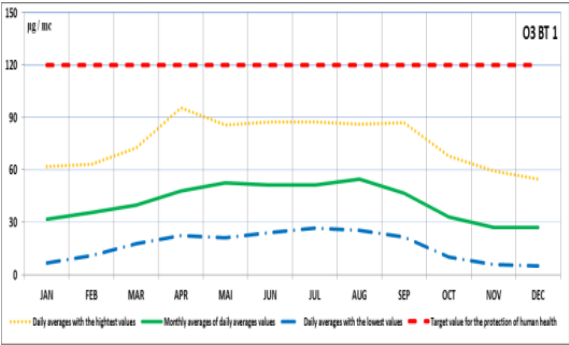
Figure 2. Annual regime of monthly average values of ozone concentration at EPA stations in RNER (2009 – 2020)

3.4. Daily average, maximum and monthly hourly maximum values of ozone concentration at the monitoring points of this pollutant in RNER

Within a year, the intermonthly course of the daily average and extreme values is similar to that of the monthly average. Important correlations were observed between the tropospheric ozone concentration and the air humidity, respectively the wind regime (Mihăilă et al., 2018; Amihăesei et al., 2019) (Table 7 and 8 and Figure 3 and 4). There is a strong reverse proportionality relationship between air humidity and tropospheric ozone concentrations over a year. During the year when the relative humidity reaches the lowest values (April–May), the ozone concentrations reach their maximum. In the months with the highest relative humidity (November–January), ozone concentrations reach the lowest values. The first explanation would be that ozone is removed from the air by absorbing water droplets. Moreover, in periods of high humidity (evening, night and morning, especially in the cold season), with pronounced cloudiness and precipitation, the intensity of sunlight decreases and this reduces photochemical processes in the production and decomposition of ozone. The humidity itself has, during these periods, a relevant impact on the ozone level, resulting in strong negative ozone – air humidity correlations.

Table 7. Maximum daily values of ozone concentration for each month at EPA stations in RNER (2009 – 2020)

Code station	I	II	III	IV	V	VI	VII	VIII	IX	X	XI	XII	Diurnal maximum
BT 1	62.0	82.5	102.0	96.4	85.6	87.4	92.6	89.6	87.0	73.0	59.3	56.6	102.0
SV 1	94.3	96.9	102.0	115.1	90.0	93.2	96.1	104.1	97.1	78.5	80.5	91.0	115.1
EM 3	85.1	95.5	101.6	109.8	105.0	109.9	109.2	98.4	90.6	78.0	85.1	81.0	109.9
IS 4	92.8	99.7	128.2	116.6	108.8	105.0	111.8	123.0	109.0	86.2	65.2	68.6	128.2
IS 3	73.5	106.9	99.3	99.9	91.0	92.0	96.8	90.7	81.2	59.5	50.6	55.2	106.9
IS 5	77.0	79.1	104.7	91.7	85.0	94.8	109.9	101.7	91.2	78.7	52.3	64.2	109.9
NT 1	73.3	116.4	106.1	137.6	104.4	102.8	88.9	92.4	104.5	77.1	56.4	76.6	137.6
VS 1	66.9	118.7	104.8	89.3	96.9	90.5	91.0	81.1	84.6	62.2	61.7	69.4	118.7
BC 1	80.5	100.7	94.8	94.2	94.6	81.8	88.6	89.1	83.1	71.7	53.1	64.8	100.7
BC 2	87.7	120.9	107.0	88.3	92.9	83.0	79.4	82.6	75.9	79.4	78.4	69.0	120.9
BC 3	86.5	89.3	117.7	101.8	82.8	76.1	94.6	90.9	80.8	91.6	60.8	72.0	117.7



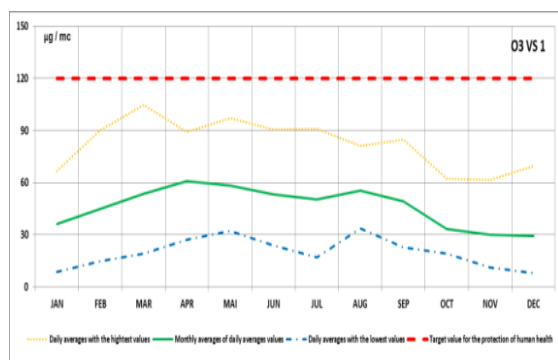


Figure 3. Annual course of average, maximum and minimum diurnal values of ozone concentration for each month at EPA stations in RNER (2009 - 2020)

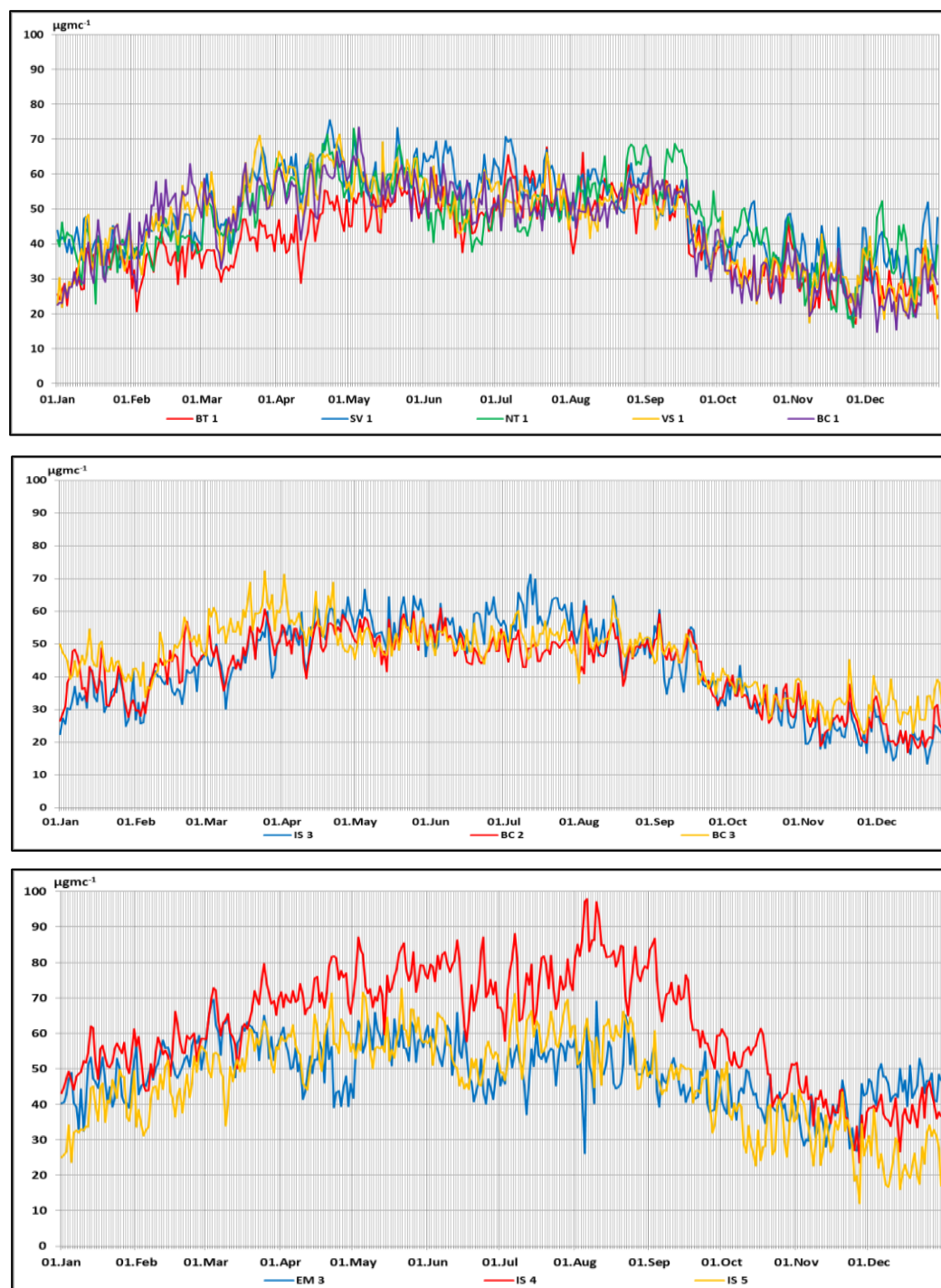


Figure 4. Annual regime of ozone concentration outlined based on the diurnal average values of this parameter at the EPA stations in RNER (2009 - 2020)

More frequent atmospheric calm and lower wind speed in urban areas have been factors that have contributed to the accumulation of ozone precursors and the exceeding of the specific threshold for this pollutant. The most likely cause of the positive correlation between the two parameters (ozone and wind) is the transport of ozone precursors from the local industrial sources of NO_x and VOC (industrial platforms of cities) and from the natural sources of VOC (forests around cities). Horizontal air movement can cause significant changes in ozone concentration in urban or non-urban areas. Concentrations higher than 60 µg/m³ were measured when the wind had a speed higher than 5 m/s from the south and southwest direction. In summer, ozone concentrations are high regardless of the wind direction, but an ozone concentration peak occurs in wind speed ranges between the thresholds of 4 and 7 m/s. The wind on the southern and southwestern component is related to tropical air advections. High maximum air temperature and low humidity are specific features of these advections. In winter months, even if east winds have speeds higher than 7 m/s, ozone concentrations are low, as these periods are characterized by heavy fog, low radiation and minimum temperatures.

The correlations established between the ozone concentration and the duration of the Sun's brightness are statistically significant during the days of the warm season of the year and in the afternoons of the days of this season.

Anthropogenic emissions (mobile - exhaust gas, NO_x, CO, hydrocarbons; stationary - chemical plants, oil refineries, dye-works, dry cleaning facilities, gas stations) induce cyclicity in ozone concentrations. There is also a weekly periodicity of urban ozone levels, ozone correlating negatively with anthropically generated nitrogen oxides (NO_x) which are strong ozone dispersers in urban areas. A high concentration of ozone was observed on Saturday-Sunday as a result of low road traffic. Ozone concentrations show weekend values above the averages of the other days of the week (Amihăesei et al., 2019).

The correlations established between the ozone concentration and the duration of the Sun's brightness are statistically significant during the days of the warm season of the year and in the afternoons of the days of this season.

The hourly maximums of ozone are higher in spring days (Table 8) when a combination of natural and anthropogenic factors favoring the genesis of ozone meet on short term and punctually. These situations and values are, however, rare.

The annual regime of ozone diurnal means illustrates that during hot season days, ozone concentration is higher and the highest concentrations of this pollutant have occurred at the IS4 station (Figure 4).

Table 8. Hourly maximum values of ozone concentration for each month at EPA stations in RNER (2009 - 2020)

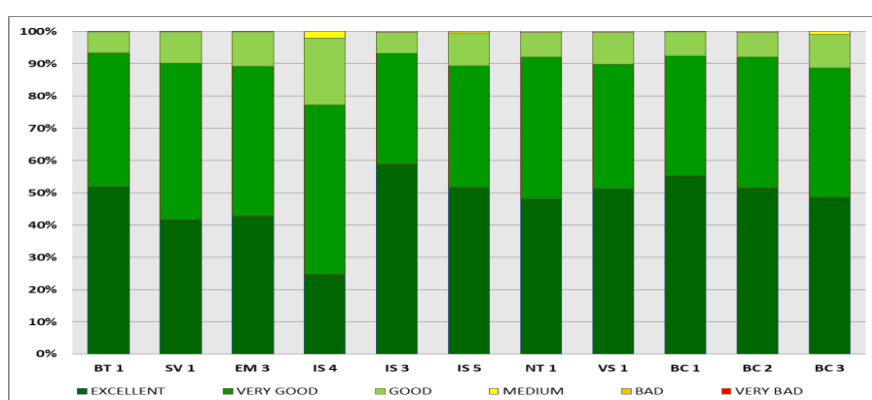
Code station	I	II	III	IV	V	VI	VII	VIII	IX	X	XI	XII	Hourly maxim
BT 1	91.7	108.0	139.7	148.6	125.8	122.2	141.9	125.9	124.9	112.3	79.2	120.8	148.6
SV 1	119.5	138.4	132.6	157.2	134.9	130.9	148.0	135.4	142.4	119.4	111.7	114.6	157.2
EM 3	97.7	124.9	134.0	153.0	155.4	148.7	149.4	145.5	121.3	119.2	109.6	101.0	155.4
IS 4	127.2	111.5	151.4	143.7	127.7	131.8	144.4	149.7	144.4	116.2	78.9	92.4	151.4
IS 3	112.4	125.1	152.1	134.2	135.6	128.0	151.6	142.5	134.8	101.9	73.7	75.3	152.1
IS 5	117.5	117.9	151.6	131.2	126.3	145.0	148.2	140.4	135.4	109.3	76.6	107.2	151.6
NT 1	103.7	174.5	142.9	180.3	157.7	163.7	144.6	140.2	144.6	115.1	93.2	99.5	180.3
VS 1	100.1	168.4	127.5	135.0	125.9	119.1	128.5	125.0	125.5	103.9	83.1	93.5	168.4
BC 1	108.6	151.0	152.6	137.8	124.2	122.4	142.4	130.6	124.9	107.3	91.1	85.2	152.6
BC 2	114.0	162.9	168.9	129.8	123.0	128.6	146.1	156.5	127.9	117.1	126.0	105.3	168.9
BC 3	110.4	121.4	169.5	135.5	121.9	113.1	134.4	134.7	126.4	113.9	103.7	99.0	169.5

Analyzing centrally by years and stations the specific daily index of air quality given by the ozone concentration in the atmosphere, a few interesting observations can be made for RNER (Table 9 and Figure 5).

Table 9. The specific index corresponding to ozone (according to Law 104/2011)

Ozone concentration range ($\mu\text{g} / \text{m}^3$)	Quality index	Specific index
0-40	Excellent	1
40-80	Very good	2
80-120	Good	3
120-180	Medium	4
180-240	Bad	5
>240	Very good	6

An excellent and very good air quality (over 80% of summed and mediated values) has been recorded and calculated at most EPA stations in the areal under study. The lowest proportion of indices indicating a superior air quality was recorded and calculated at IS4 station (rural type station, which monitors air traffic at Iași International Airport). The highest proportion of indices indicating a superior air quality was recorded and calculated at IS3 station (industrial type station, where polluting industrial activities have been stopped or gradually replaced by other economic activities).

**Figure 5.** Centralized multiannual proportions of daily values of quality indices corresponding to the concentration of ozone in the atmosphere at EPA stations in RNER (2009 - 2020)

In the period 2009 - 2020, there were rare situations when the air quality index was medium, at the stations Iași 4 – 2,16% (rural type station near Iași International Airport), Bacău 3 – 0,98% (industrial type station near Onești Industrial Platform), Iași 5 – 0,45% (suburban type station near CET II Holboca Iași) and Neamț 1 – 2,16% (urban type station). The Bad quality index was recorded only once in the analyzed interval (NT 1 - April 28, 2009, 12:00 - 180.28 $\mu\text{g} / \text{m}^3$), and the Very bad quality index indicated by the ozone concentration, was not registered at the RNER stations.

3.5. Hourly average values of ozone concentration for each month at the monitoring points of this pollutant in RNER

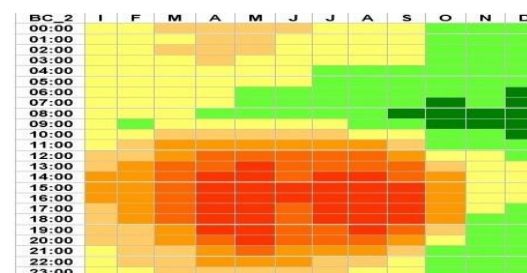
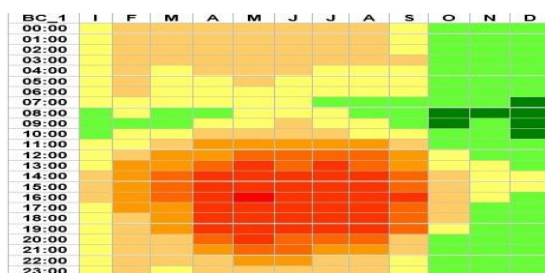
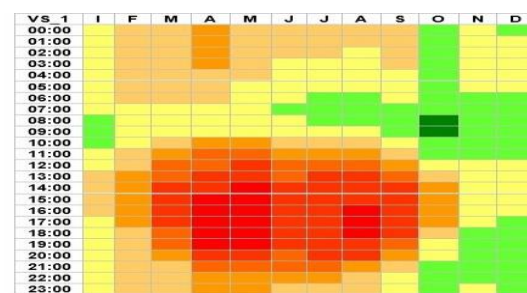
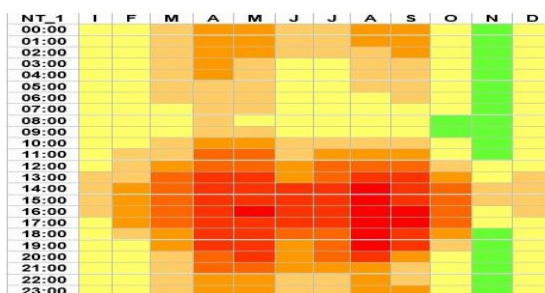
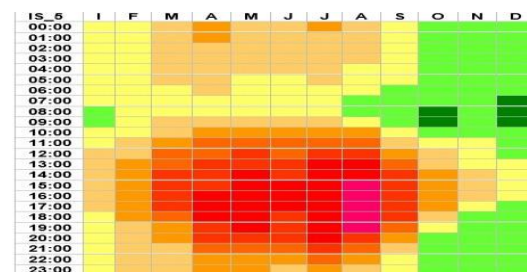
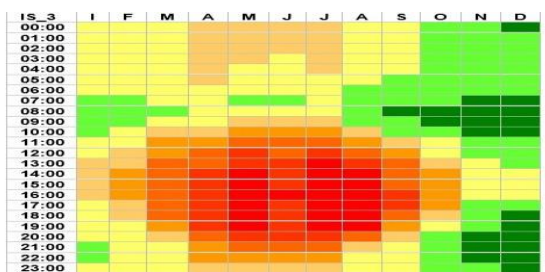
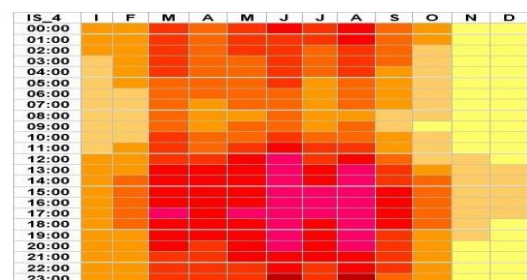
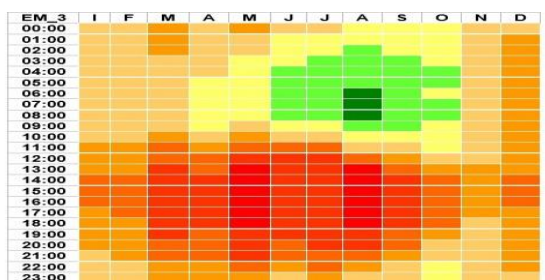
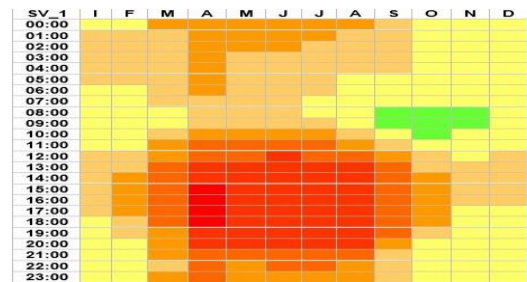
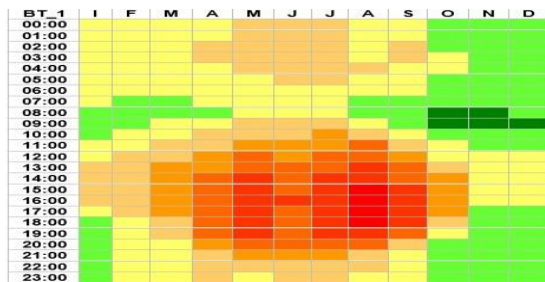
The annual profile of ozone had the highest hourly concentrations in the spring-summer period, especially in April-May. During a day, the critical time interval in which ozone had the highest values was from 11⁰⁰-20⁰⁰ in the spring-summer season and 11⁰⁰-18⁰⁰ in the rest of the year. We considered both the full diurnal cycle and the main critical diurnal interval (11⁰⁰-18⁰⁰), when ozone values were highest throughout the year [27, 28] (Figure 6).

The diurnal evolution of the duration of the Sun's brightness and of the tropospheric ozone concentration are positively and strongly correlated. The diurnal maximum of the Sun's brightness duration occurs most frequently when the Sun crosses the meridian of the place or immediately post-meridian (12⁰⁰-13⁰⁰), approximately 3-4 hours before the diurnal maximum of ozone, which is usually recorded during the afternoon hours

(16⁰⁰-17⁰⁰). The duration of the Sun's brightness in the time interval 8⁰⁰-17⁰⁰ best correlates with the ozone level determined in the time interval 11⁰⁰-20⁰⁰.

The diurnal course of ozone level and temperature shows for all the months of an average year a relatively good overlap in the time interval 10⁰⁰-11⁰⁰ and 19⁰⁰-20⁰⁰. Within this interval, the ozone concentrations which are higher than the multiannual average (approximately 50 µg/m³) correspond to temperatures > 15 to 20 °C.

The higher wind speed in the middle of the day and in the afternoon, influences the ozone maximum, bringing air masses from nearby forests (low in NO_x and rich in VOCs). Under conditions of polluted urban air volumes, water vapor in the atmosphere can act by forming the OH radicals in the regeneration of photochemically decomposed ozone.



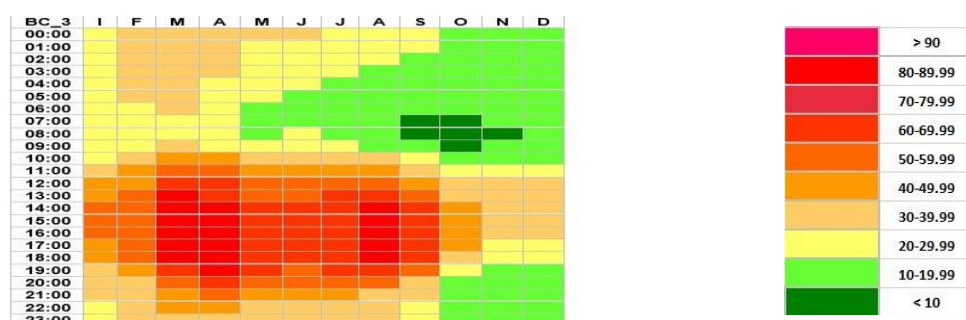


Figure 6. Hourly average values of ozone concentration for each month at EPA stations in RNER (2009 – 2020)

4. Conclusions

The results of the air quality monitoring in the automatic stations belonging to the RNMCA on the territory of RNER, in the period 2009 - 2020, indicated that the values of recorded ozone concentrations were generally lower than the information threshold ($> 180 \mu\text{g} / \text{m}^3$) provided of Directive 2008/50/EC and Law 104/2011.

In space, no areals with Bad and Very bad quality index of ozone concentration were registered. During period under study (2009 - 2020) there were very rare situations when the air quality index was Medium, due to air traffic (IS 4) and activities within industrial platforms (BC 3, IS 5, NT 1). As a result of the data processed, at most of the stations in the areal concerned, the air quality index was Excellent and Very good. High ozone concentrations were recorded at urban type stations due to the characteristics of the active surface and the economic and social profile of the areas of representativeness, to which is added in some cases the heavy air traffic at international airports in the metropolitan area of large cities of RNER. Low values were generally recorded at industrial stations, where atmospheric calm and urban haze favor the accumulation of solid and gaseous pollutants on the soil surface, an excess humidity that limits the transparency of the atmosphere for UV radiation and ozone genesis.

In time, the maximum monthly, diurnal and hourly values of ozone concentration have been recorded at most stations in April-May, when the UV flow reaches its maximum intensity against the background of maximum atmospheric transparency. The lowest monthly, diurnal and hourly ozone concentrations occurred mainly in November and December (foggy months, with stratiform clouds, short days and low UV values). Anthropogenic emissions (mobile - exhaust gases, NO_x , CO, hydrocarbons; stationary - chemical plants, oil refineries, dye-works, dry cleaning facilities, gas stations), induce weekly cyclicity in ozone concentrations. The diurnal profile of ozone shows the highest hourly concentrations in the time interval 1100-2000 in the days of the hot season and 1100-1800 in the days of the cold season, due to the influence of the duration of the Sun's brightness on anthropogenic activities.

Ozone, according to the air quality indices calculated for RNER, has taken second place among air pollutants after PM_{10} sedimentable dusts. Ozone pollution may become a real problem in the current climate context in RNER, as with rising air temperatures and falling atmospheric precipitation, ozone concentrations are also rising.

This element may cause damage to all components of the environment, particularly in urban environments, when surface ozone concentrations increase due to photochemical reactions dynamized by the increase of the caloric and thermal potential of the atmosphere and by the atmospheric pollution whose growth is difficult to control.

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