

OBSERVATIONAL STUDY OF MAJOR AIR POLLUTANTS OVER URBAN ROMANIA IN 2020 IN COMPARISON WITH 2019

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Abstract. This study aims to characterize urban air pollution in 33 cities across Romania in 2019 and 2020. The global COVID-19 pandemic that started in mid-March 2020 provided us with a unique opportunity to find out what would be the possible variation range of major air pollutants in Romanian urban areas under various strengths of anthropogenic activity. The time-series of atmospheric main pollutant mass concentrations, particulates PM₁₀ and PM_{2.5} and gaseous species NO₂, CO, SO₂, O₃, were statistically analyzed for each city for the representative month of each season in 2019 and 2020 and then used to estimate the pollutant impact on the surrounding regions. Results show significant decreases for most pollutants in most cities especially in spring of 2020, during the hard lockdown period. Maps of interpolated mass concentrations reveal regional significant differences with pollutant-specific hot- and cold-spots in Romania. This work constitutes a good database to characterize de urban air pollution in Romanian major cities and the spatial distribution of their impact around.

Key words: urban air pollution, seasonal variation, spatial distribution, COVID-19, East Europe.

1. INTRODUCTION

Atmospheric pollution represents an important environmental issue of global concern back to 1970s [1] and the anthropogenic activities are nowadays considered to be responsible for the elevated levels of air pollutants, either particulates of various sizes (PM_x-diameter smaller than $x \mu\text{m}$, where $x=10, 2.5, 1, 0.1$) or gaseous species (e.g., NO, NO₂, SO₂, CO, VOCs). Along with these pollutants, O₃ is also of high interest because of its numerous effects on climate, environment, and human health. O₃ is one of the most important secondary pollutants resulting from anthropogenic emitted precursors such NO_x, CO, and VOCs. Over 6000 cities in 117 countries are now monitoring the air quality and many efforts are made in addressing both the improvement of the air quality and to

mitigate the climate changes [2]. Despite of the progress made, people around the world still breathe unhealthy air, and mostly due to particulate matter PM and nitrogen oxides. In addition, other pollutants like carbon monoxide, sulfur oxides, various volatile organic compounds, etc. contribute to air degradation especially in industrial and urban agglomerations, where the respiratory health effects exacerbate, e.g. [3]. Moreover, regional discrepancies of air pollution effects are important to be revealed and quantified. In this respect, the very recent study of Gu et al. (2023) shows that eastern European countries suffered greater harmful health air pollution effects than other central or western European countries, in contrast to the emissions caused by eastern versus western/central countries [4]. Elevated levels of air pollutants not only affect human health but they impair the air quality and the visibility by modifying the atmospheric composition and leads further to climate changes because the air quality and the climate are highly interconnected through a plethora of ways [5]. Therefore, the systematic monitoring of air pollutants and continuous investigations is of essential importance.

In Romania, monitoring of major air pollutants using ground measurements started only about twenty years ago. The database of Romanian National Air Quality Monitoring Network (www.calitateaer.ro) and the European Air Quality Database (<https://www.eea.europa.eu/data-and-maps/explore-interactive-maps/up-to-date-air-quality-data>) provide freely the same data sets. Based on these measurements, coupled or not with other remote sensing observations, previous studies reported assessments or investigated various issues related to air pollution over Romanian territory, e.g. [6-10], or for Bucharest, e.g. [11-14] or in other various regions of the country, e.g. [15-17].

The lockdown associated to COVID-19 pandemic in 2020 represented an exceptional opportunity to investigate its effects on air quality at continental or regional scale, including Romania, e.g. [18]. The restrictions of social, non-critical traffic and industrial activities in 2020 can help understanding the variations of primary pollutants of most concern from lowest to highest levels that we may be facing on the Romania scale.

Therefore, present study investigates how the atmospheric pollution by PM_{10} , $PM_{2.5}$, NO_2 , CO , SO_2 , and the by-product O_3 varied over the Romanian territory in 2020 in comparison with 2019. We assessed comparatively these ground-based measured pollutant levels in 33 urban agglomerations suffering from different pollution sources, having different level of development, cities with regional role and potential influence at Romanian and European scale. The comparative 2019-2020 analysis was made in terms of lowest, average and highest levels. The detailed objectives were as follows: 1) to identify temporal (seasonal) variation of major air pollutants in all 33 urban areas in 2020 in comparison with 2019, as reference year; 2) to estimate spatial seasonal variation of pollutants at the

country scale; 3) to estimate the urban impact over their peri-urban areas determined by the hard lockdown measures in spring of 2020 in Bucharest, Brasov, Iasi. These three cities are in continuous attention of European Commission due to systematic exceedances of air quality limits, especially by PM_{10} and NO_x [19].

A description of the measurements sites and methodology used in this work is detailed in Section 2. Section 3 presents the results and discusses them. The main conclusions are exposed at the end of the paper.

2. MATERIALS AND METHODS

2.1. SELECTION OF THE URBAN AREAS

Cities or urban agglomerations were selected for present analysis based on the availability of data at monitoring stations, gathered from Romanian National Network of Air Quality Monitoring (www.calitateair.ro) and on their development level. Socio-economic statistical indicators (resident population, gross domestic product, level of industrialization of each city) used to select the cities were acquired from the National Institute of Statistics web portal (<http://statistici.insse.ro:8077/tempo-online/>) and from the General Secretariat of the Government (<https://data.gov.ro/en/dataset?tags=Parcuri+industriale>), under license CC BY 4.0. Generally, cities' economic development spans from Bucharest, the most developed and inhabited, to, for example, Giurgiu, city significantly less industrialized but an important road transportation hub for goods on western-eastern European countries' axis. Figure 1 shows the map of Romania, which has a continental climate with very warm to hot summers and relatively cold winters, and that is under climatic changes as results from analysis of observational data (1961–2013) in [20]. One can observe the selected cities and urban agglomerations provide a good spatial coverage of the entire country and they experience all regional climates, from warm oceanic climate at the Black Sea (Cfa by Köppen classification) to the cold continental Dfc and alpine climate ET. As a comparative example, Cluj - Napoca has a continental climate with warm summers, under the simultaneous influences of West Atlantic air mass circulations during winter and autumn and of the Apuseni Mountains (wet temperate continental, Dfb by Köppen classification), while Bucharest is characterized by a more humid continental climate in transition to humid subtropical climate (Dfa/Dfb to Cfa) with hot humid summers and cold winters but with more frequent milder periods during the last years.



Fig. 1 – Map of Romania and distribution of selected cities with ground-based measurements of air pollutants within the National Air Quality Monitoring Network.

2.2. PREVENTING THE SPREAD OF THE COVID-19 PANDEMIC IN 2020

First year of COVID-19 pandemic provided a unique opportunity for a comparison experiment of ambient air pollutant mass concentrations in a year with minimum impact of anthropogenic activities at country scale. The policy measures taken in Romania in order to limit the spread of SARS-COV2 virus and the virus multi-waves are presented in exhaustive details in [21,14]. Therefore, here we just mention that in present study the impact of hard lockdown measures can be seen in the air quality data in April 2020 because the hard lockdown in Romania was declared between middle of March to middle of May 2020.

2.3. AIR POLLUTANT MONITORING DATA

The study was conducted over two years, from 01.01.2019 to 31.12.2020 using daily means of particulate matter PM_{10} and $PM_{2.5}$ and of NO_2 , SO_2 , CO and O_3 in all 33 urban areas in the Romanian territory in Figure 1. Data was extracted from the database of the National Air Quality Monitoring Network. Each city has at least

one station for air quality measurement. Daily values of each pollutant per city were obtained by averaging data from all stations within the city, as in similar studies focused on the overview of air quality in small to medium-size cities, e.g. [22]. This method allows overcoming the design of the AQ monitoring network. For larger cities it has been found that the urban-suburban gap is rapidly narrowing especially in cities in fast development, e.g. [23]. However, in present study data at suburban stations were considered separately for the spatial analysis at the country scale. For the spatial analysis the inverse Euclidean distance method was used, one of various interpolation tools for environmental data analysis [24, 25]. In some cases, when no gravimetric data were available for PM_{10} or $PM_{2.5}$, hourly data collected with other accepted methods (e.g. LSPM, Sharp) extracted from the same source-database, were converted to daily means and used for analysis. Monthly mean values were calculated from daily mass concentrations. The temporal evolution was performed for the representative month (April, July, October, and December) of the four seasons (spring, summer, autumn, winter).

3. RESULTS AND DISCUSSION

3.1. DIFFERENCES IN MAJOR AIR POLLUTANT LEVELS IN URBAN AREAS

The minimum, mean and maximum levels of each pollutant by each city/urban agglomeration were first calculated. Figure 2a-e shows multi-panel seasonal variations for PM_{10} , NO_2 , SO_2 , CO and O_3 , respectively. Then, we estimated the mean changes in pollutant levels, synthesized in Figure 3. Results reflect both seasonal and substantial inter-city variations pointing to the local factors. Apart of their sources of various strengths (industrial activities, combustions, road traffic, etc.), other reason is also the level of development of the city, revealed by the gross domestic product (GDP) of the county where the city is located. At the country scale, the correlation between PM_{10} emission level and GDP is high and the level of atmospheric aerosol pollution from satellite data is above 0.60 as previously reported by Chiritescu et al. (2019), who analyzed data from 2009 to 2017 [26]. Figure 2a-e indicates that both particulate PM_{10} (and $PM_{2.5}$, not shown here as it is monitored in only 20 cities), and gaseous species NO_2 , CO and SO_2 had higher levels during cold seasons with the exception of O_3 , which shows an opposite behavior. Largest variations are observed in October 2019, from few $\mu g m^{-3}$ to more than $80 \mu g m^{-3}$, for PM_{10} in 25% of cities, and for NO_2 from few $\mu g m^{-3}$ to more than $40 \mu g m^{-3}$ in 16% of cities. Therefore, these two pollutants still represent an unsolved pollution issue in Romania at least for cold season. Variations became significantly lower during warm season.

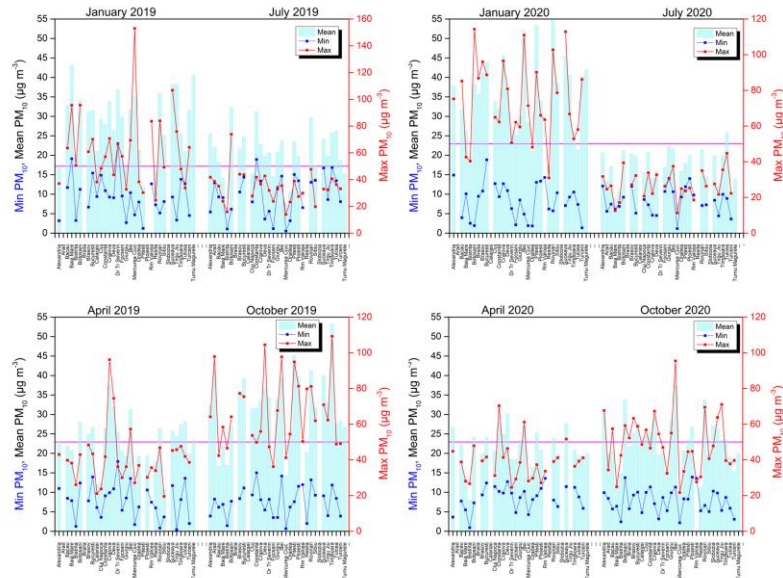


Fig. 2a – Minimum, mean and maximum seasonal levels of PM_{10} ($\mu\text{g m}^{-3}$) by city in 2019 (left) and 2020 (right). Horizontal (magenta) line represents the daily limit value set by the EU legislation (marked on the right axis). Cities are showed in alphabetical order in each set of graphs.

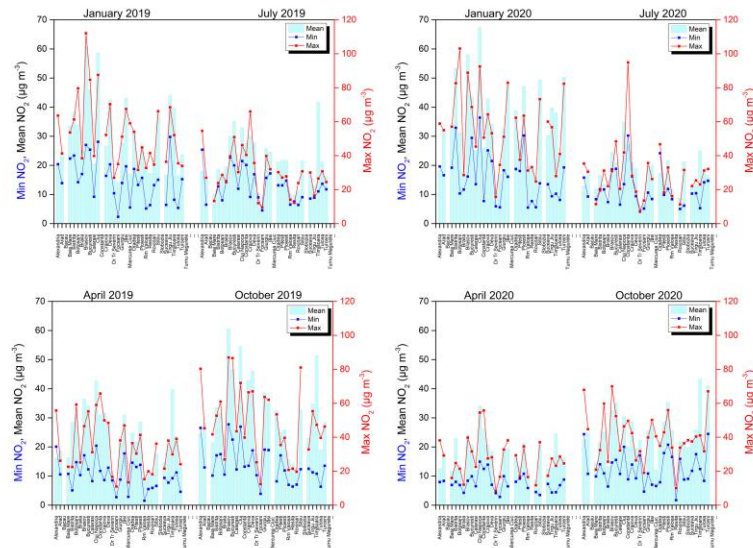


Fig. 2b – Minimum, mean and maximum seasonal levels of NO_2 ($\mu\text{g m}^{-3}$) by city in 2019 (left) and 2020 (right). Cities are showed in alphabetical order in each set of graphs.

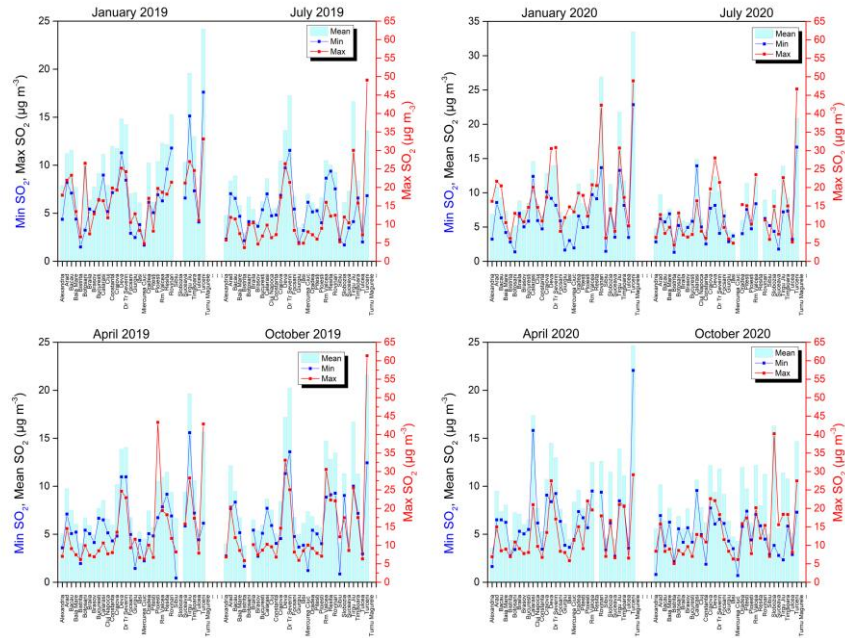


Fig. 2c – As in Fig. 2b but for SO₂ (µg m⁻³).

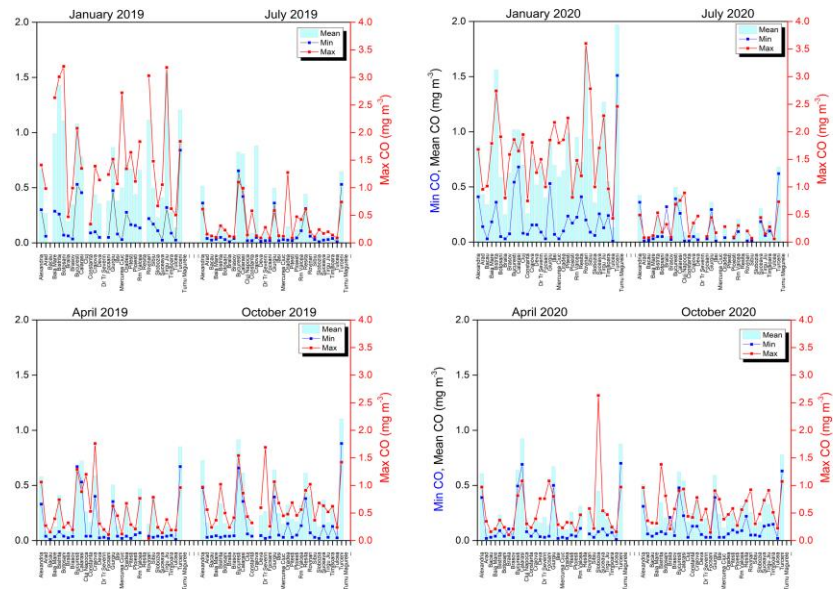


Fig. 2d – As in Fig. 2b but for CO (µg m⁻³).

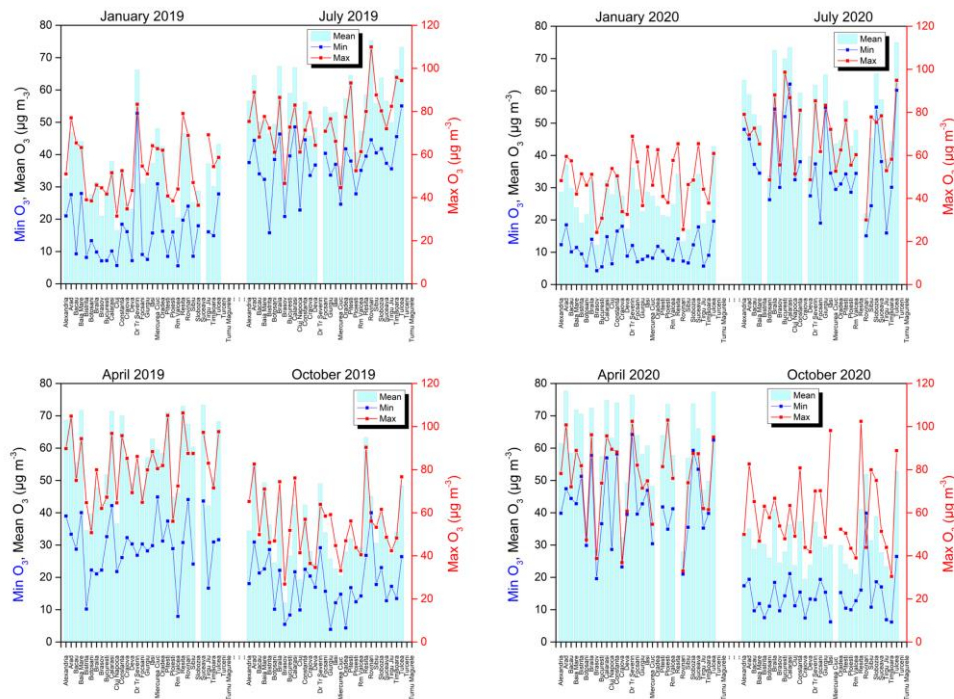


Fig. 2e – As in Fig. 2b but for O_3 ($\mu\text{g m}^{-3}$).

3.2. CHANGES IN POLLUTANT MEAN LEVELS IN URBAN AREAS

The daily mean concentrations of all pollutants in 2020 were compared with the daily mean concentrations of the years 2019. The vertical axis in Figure 3 has an increment of 1. Consequently, any vertical bar higher than 1 indicates an increase of pollutant levels, therefore a further amplification of urban effects over the surrounding areas, while a vertical bar below 1 represents a decrease of the mass concentrations of the corresponding pollutant in 2020, which corresponds to a reduction of urban effects. In January 2020 (no restriction measures), the pollutant levels were almost the same for SO_2 for 75% of cities (example exceptions are Miercurea Ciuc and Rovinari, where SO_2 in 2020 were about 2 times higher) and PM_{10} for 50% of cities (example exceptions: Pitesti, where PM_{10} in 2020 was 4 times higher, Alexandria - 2 times higher).

With respect to CO, we observe that 72% cities show higher mass concentrations in 2020 than in 2019. Some of these cities are located in the western part of the country (some are also characterized by increases in SO_2), while others are located in inter-alpine basins, Miercurea Ciuc, Sibiu, or in the North of the

country, as Suceava. During spring, summer and autumn seasons, in general, a pattern of lower mean mass concentrations of all air pollutants throughout 2020 compared to the 2019 in urban Romania is observed. The highest decreases are encountered in spring, and this is obviously a result of the decline of anthropogenic emissions during the lockdown. However, some exceptions can be noticed. Among the obvious exceptions are Rovinari, Botosani, and Oradea that present increases in PM_{10} levels than in April and July 2019; CO is enhanced in Constanta and Tirgu-Jiu in October 2020 and in Suceava and Slobozia in spring and summer 2020.

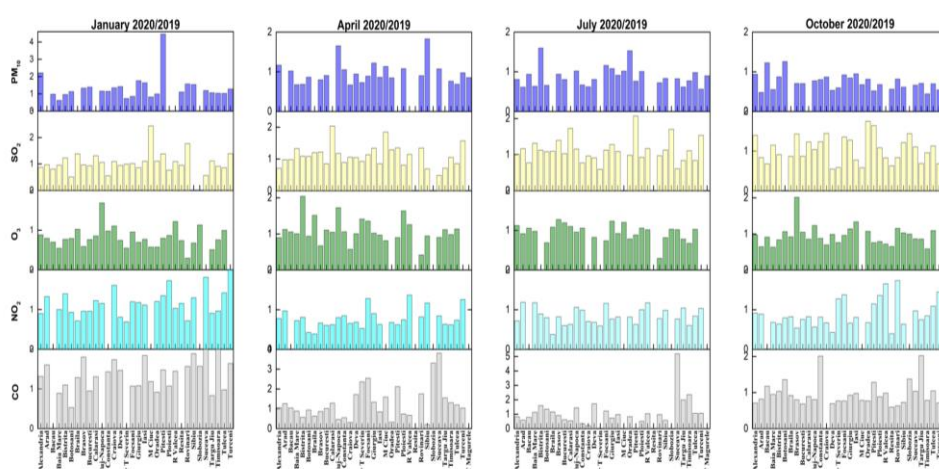


Fig. 2 – Specific pollutant ratios 2020/2019 for PM_{10} (dark blue), SO_2 (yellow), O_3 (green), NO_2 (light blue), and CO (grey) at country level in January, April, July and October (from left to right). Scale on the left has an increment of 1. Value above 1 (below 1) means increases (decreases) of pollutant levels in 2020 with respect to 2019 (right). Cities are showed in alphabetical order in each set of graphs.

In Poland were reported reductions of PM_{10} concentrations in April and May approximately by -23% and -18%, respectively and of NO_2 approximately by -10% to -19%, respectively [27]. In Spain, Baldasano (2020) reports higher reductions: NO_2 concentrations in Barcelona and Madrid, under COVID-19 lockdown during March 2020 decreased by 50% and 62%, respectively [28]. Other various European cities had different NO_2 reductions: London and Paris experienced reduction of about $24 \mu g m^{-3}$, $31 \mu g m^{-3}$ (about 48%, 50%) at traffic stations and $13 \mu g m^{-3}$, $16 \mu g m^{-3}$ (about 37%, 47%) at urban background stations, respectively. For Milan the reduction of NO_2 concentrations was of about $13 \mu g m^{-3}$ (about 35%). Obviously, all Romanian and European cities experienced pollutant reductions that depend largely on local factors and on the various levels of restriction measures which were country-dependent [29].

3.3. URBAN INFLUENCE OVER SURROUNDING AREAS AT ROMANIA SCALE

Further on, for the urban impact spatial distribution at Romania scale, we performed maps of interpolated spatial distributions of mean concentrations of all pollutants in the study area in Figure 4a-e. Graphical representations are shown for each pollutant maximum levels in 2019, maximum levels in 2020 and the minimum levels in 2020. The results are showed for the representative month of the each season so that the differences in the air quality levels to be evaluated under relevant contrasting conditions, both seasonal and from the point of view of measures taken again the spread of coronavirus disease.

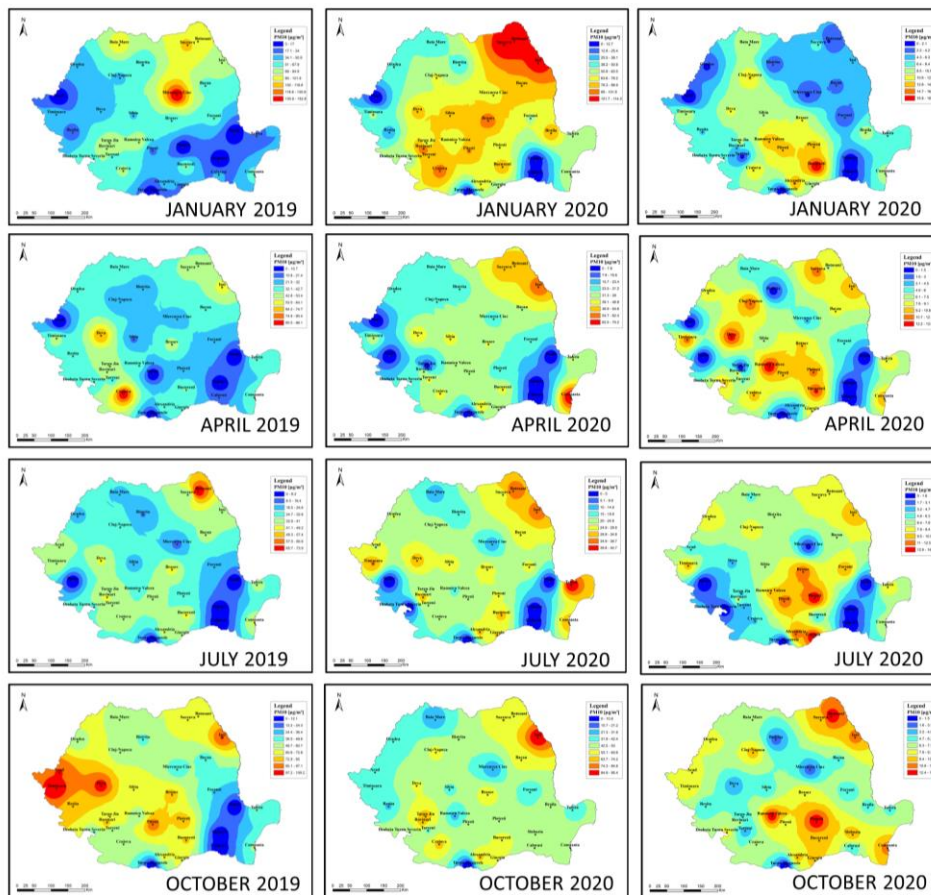


Fig. 4a – Spatial seasonal distribution of PM_{10} concentrations in urban areas and estimates of their effect on their surrounding regions in 2019 and 2020. Left side- PM_{10} maximum levels in 2019 (scale up to $153 \mu g m^{-3}$), center- PM_{10} maximum levels in 2020 (scale up to $114 \mu g m^{-3}$), and right side- PM_{10} minimum levels 2020 (scale up to $14 \mu g m^{-3}$).

Moreover, due to significant differences in seasonal pollutant levels calculated from very different measured mass concentrations in 2019 and 2020, in order to reveal the pollutant-specific hot- and cold-spots at the country scale, Figures 4a-e present various representation scales, each Figure caption and Figure legend mention clearly the scale used, together with the measurement unit ($\mu\text{g m}^{-3}$ for all pollutants, with the exception of CO, where the measurement unit is that generally accepted of mg m^{-3}).

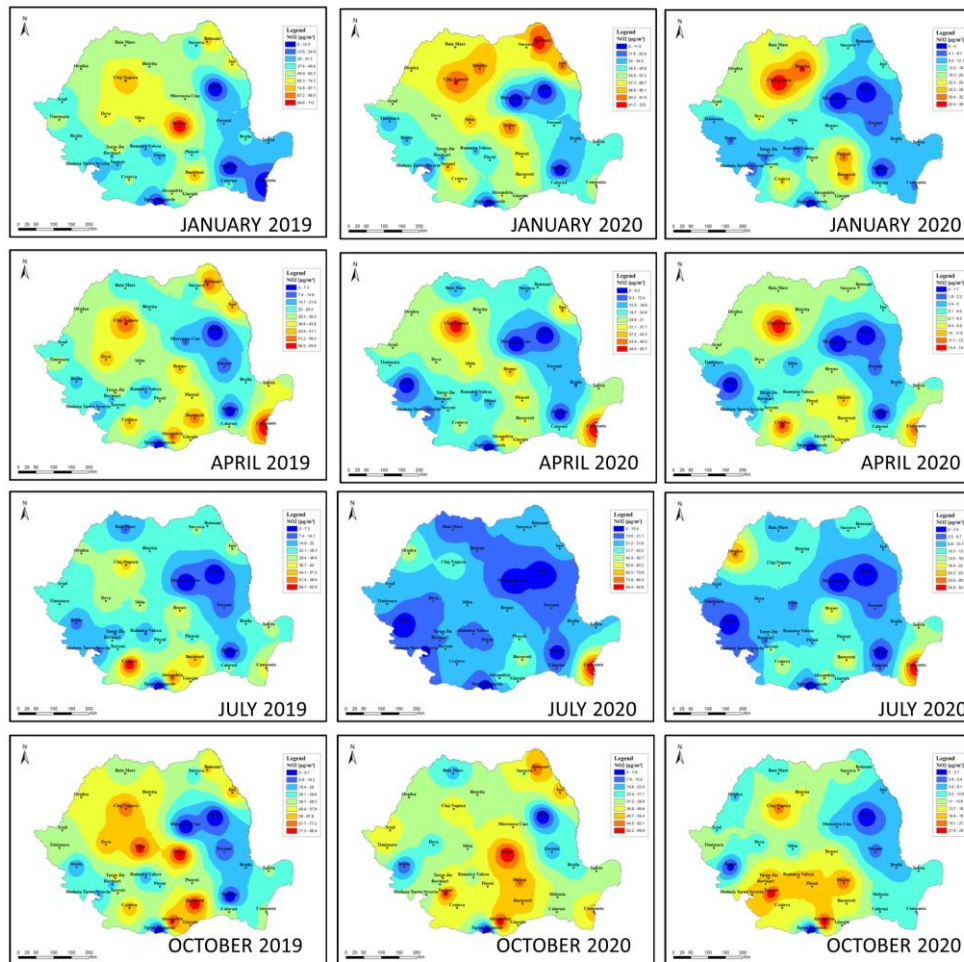


Fig. 4b – As in Figure 4a but for NO₂ concentrations. Left side-NO₂ maximum levels in 2019 (scale up to 112 $\mu\text{g m}^{-3}$), center-NO₂ maximum levels in 2020 (scale up to 103 $\mu\text{g m}^{-3}$), and right side-NO₂ minimum levels 2020 (scale up to 36 $\mu\text{g m}^{-3}$).

The representations in Figure 4a-e are very useful to reveal the spatial impact of the pollution sources, blue color can be interpreted as no significant impact on surroundings.

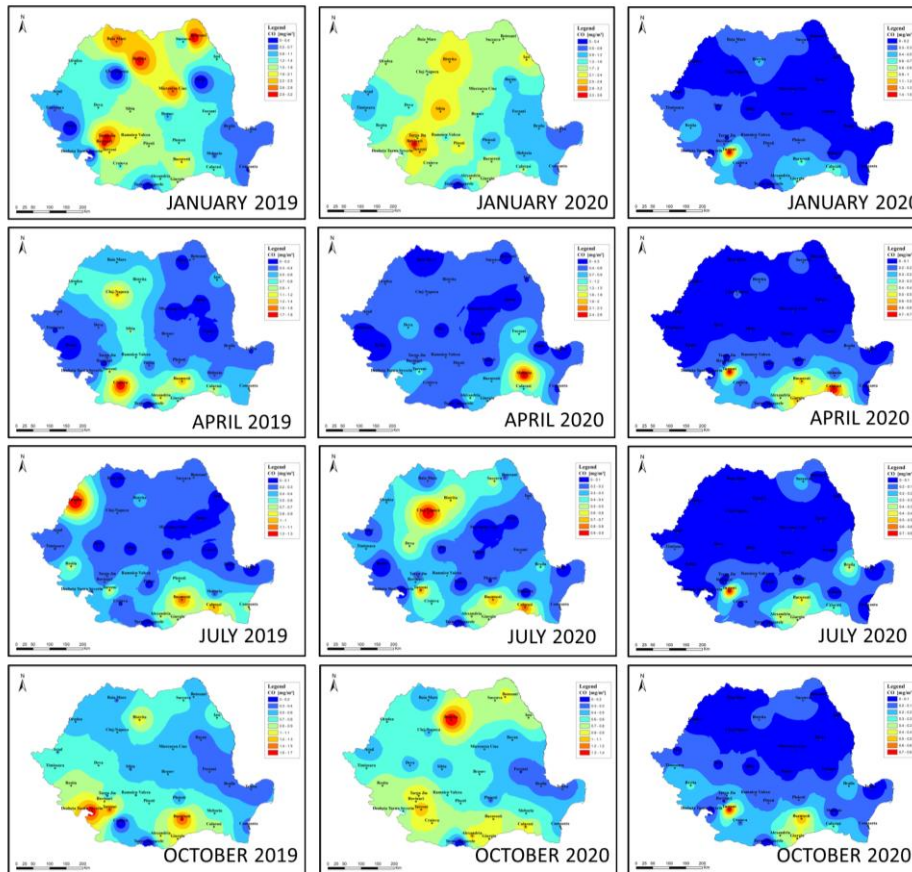


Fig. 4c – As in Figure 4a but for CO concentrations. Left side-CO maximum levels in 2019 (scale up to 3.2 mg m^{-3}), center-CO maximum levels in 2020 (scale up to 3.6 mg m^{-3}), and right side-CO minimum levels 2020 (scale up to 1.5 mg m^{-3}).

In 2019, maximum mass concentrations of all pollutants, represented on the left part of Figure 4a-e, had higher levels than the concentrations in 2020. Consequently, the PM_{10} spatial seasonal distribution indicate hot spots at Miercurea Ciuc in January 2019 (mass concentrations up to $153 \mu\text{g m}^{-3}$), at Timisoara, Deva and Iasi in October (mass concentrations up to $110 \mu\text{g m}^{-3}$), and intermediate levels of PM_{10} at Suceava and Pitesti, Rovinari. During April and July 2019, PM_{10} mass concentrations were lower than the above mentioned. Craiova and Suceava

showed, however, some elevated concentrations, up to $90 \mu\text{g m}^{-3}$, and $73 \mu\text{g m}^{-3}$, respectively.

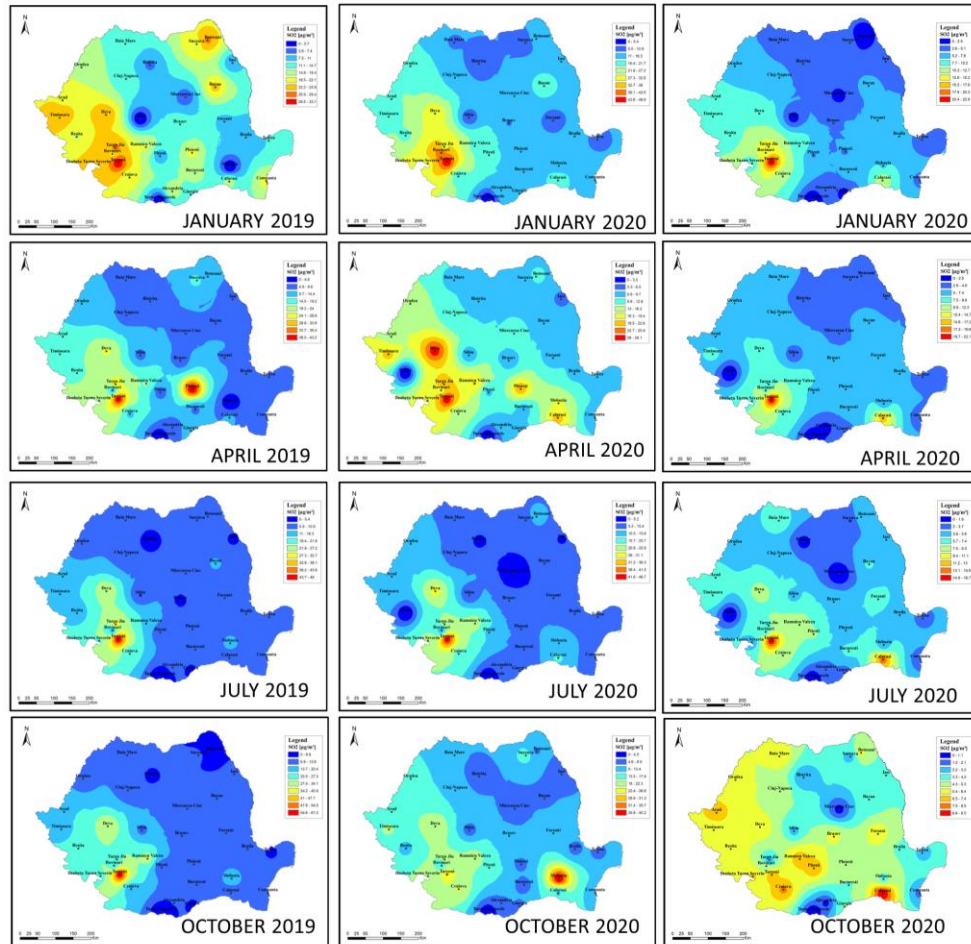


Fig. 4d – As in Figure 4a but for SO_2 concentrations. Left side- SO_2 maximum levels in 2019 (scale up to $61 \mu\text{g m}^{-3}$), center- SO_2 maximum levels in 2020 (scale up to $49 \mu\text{g m}^{-3}$), and right side- SO_2 minimum levels 2020 (scale up to $23 \mu\text{g m}^{-3}$).

The impact of urban pollution is obvious in October, most probably due to start of the cold season, when to the anthropogenic activities during summer, the supplementary heating activities and agricultural land clearing add to the pollution. NO_2 maximum levels in 2019 show some hot-spots (levels up to $103 \mu\text{g m}^{-3}$) at Bucharest, Brasov and intermediate levels at Sibiu, Craiova, Constanta ($70\text{--}80 \mu\text{g m}^{-3}$). Few cold spots are also visible in the East. CO concentrations in both 2019

and 2020 reveal a hot spot area in the south western Romania, in the Rovinari-Turceni area, where coal, mostly lignite, extraction open-pit mines and important thermal power plants operate using this fossil fuel. This is correlated very well with the SO_2 concentrations (Fig. 4d) showing the dominance of emissions from Rovinari-Turceni industrial complexes and it was also revealed in other very recent study [30] reporting the results from six field campaigns performed in this area in period 2018-2020.

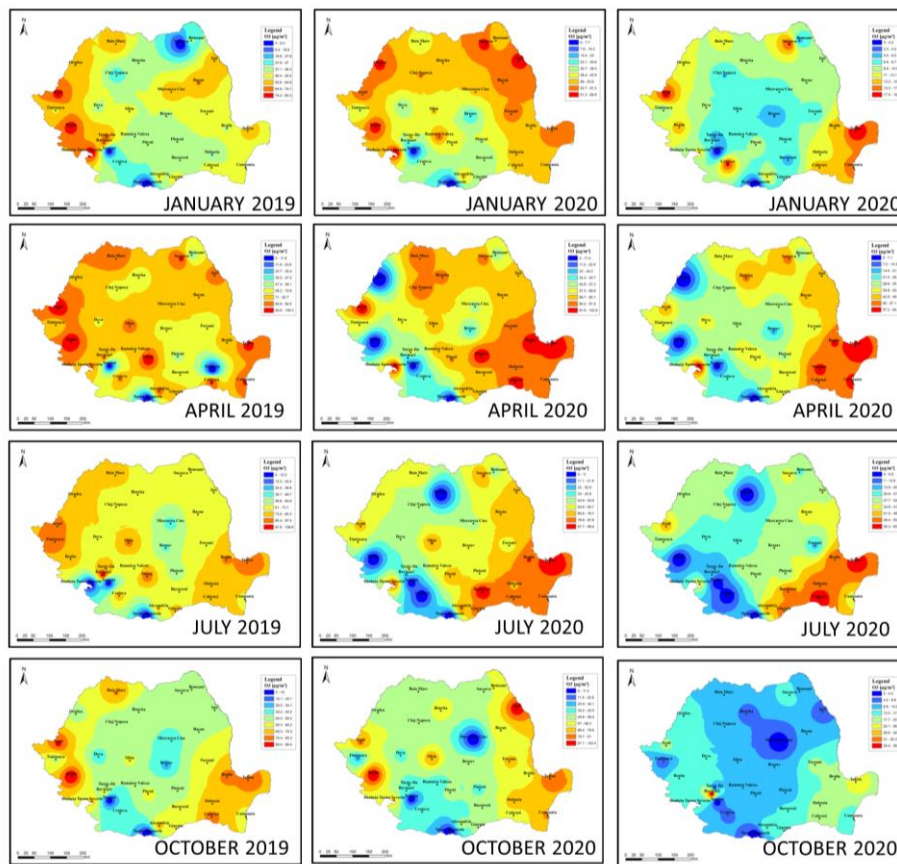


Fig. 4e – As in Figure 4a but for O_3 concentrations. Left side- O_3 maximum levels in 2019 (scale up to $110 \mu\text{g m}^{-3}$), center- O_3 maximum levels in 2020 (scale up to $103 \mu\text{g m}^{-3}$), right side- O_3 minimum levels 2020 (scale up to $64 \mu\text{g m}^{-3}$).

The maps in Figure 4e highlight to a good extent the negative correlation between NO_2 and O_3 behavior. For all major pollutants, Fig 4a-e shows that during 2020 lower mass concentrations (in both peak and minimum values) were registered, and the hot- and cold spots areas remain the same. The maximum level of calculated

seasonal mass concentrations is mentioned in both the legend and the Figure caption for each pollutant.

3.4. IMPACTS OF LOCKDDOWN ON AIR QUALITY-THREE SELECTED AREAS

A change-point analysis was performed on time series of daily mass concentrations of NO_x and PM₁₀ in selected larger cities in order to estimate the impact of implementation measures against COVID-19 spreading [31]. Figure 5 shows the situation in Bucharest 2020 which is known to be directly affected by the traffic NO_x emissions [11]. It reveals a significant AQ improvement during the hard lockdown period (between 75th - 135th Julian days), when NO_x atmospheric levels dropped to their lowest level of 23.75 $\mu\text{g m}^{-3}$, followed by a new level of 37.5 $\mu\text{g m}^{-3}$, when some of the relaxation measures were taken (until to about the 244th Julian day) and they further increased up to a mean level of 61.23 $\mu\text{g m}^{-3}$ by about the end of the year.

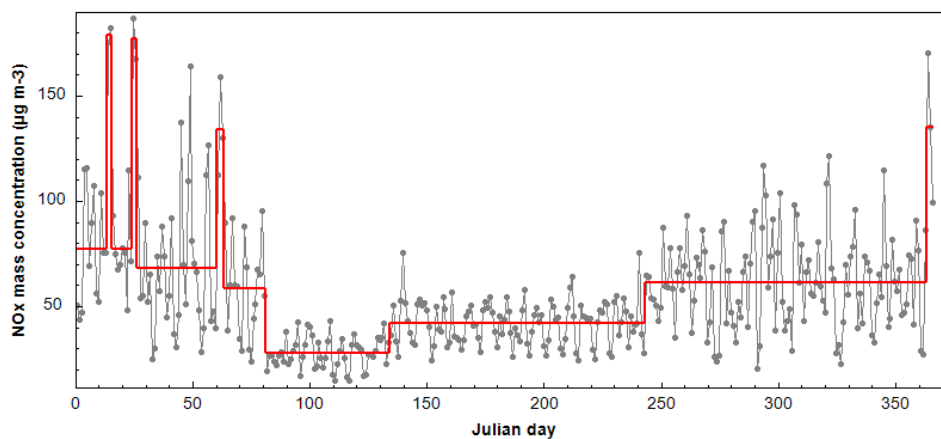


Fig. 5 – NO_x mass concentrations in Bucharest urban area in 2020 and the identified mean levels by change-point analysis

This behavior was also very recently reported by Putaud et al. (2023) for some cities in Western Europe, depending on the local anti-COVID-19 spreading measures taken [32]. In Central West Europe the decreases were up to even 30-40% for NO₂, while for PM₁₀ were lower (10-20%). The same study also reports some cases like Prague, Czech Republic, where PM₁₀ levels increased during the local lockdown, while NO₂ levels almost remained the same. Obviously, throughout the entire Europe, as it was also found in present study, PM₁₀ and NO₂/NO_x levels started to increase as the relaxation measures began to be applied. In general, the behavior in Bucharest was encountered over the Romanian territory,

but with various levels of atmospheric mass concentrations. In some cases these three steps in atmospheric mass concentrations could not be revealed, probably due to the local factors, like topography, for example (in intra alpine valley, the atmospheric mixing is significantly reduced). Present study also allows estimation of individual air pollutant urban impact (UI) on its surrounding area. UI has been estimated by difference between the pollutant-specific urban levels and the corresponding mass concentrations in the suburban area. Results for three cities, Bucharest, Brasov, and Iasi, are summarized in Figure 6.

Clearly, for all cities, the urban impact during the lockdown decreased and the cities and their surroundings areas benefited from the imposed restrictions. The improvements of the air quality presented local specificities: Brasov benefited most in terms of NO_2 and $\text{PM}_{2.5}$, followed by Bucharest and Iasi (for NO_2 and PM_{10}). In terms of SO_2 , almost no impact of a city over the suburban area was observed and this seems to remain constant over the year 2020. Suburban areas of all cities present higher mass concentrations of O_3 . Therefore, O_3 -UI is negative with the exception of Bucharest during lockdown period. When O_3 mass concentrations increase in some regions, it means the oxidizing capacity of the atmosphere is enhanced and this may lead to the production of secondary aerosol; this could be partly the reason for the differences between cities shown Figure 6.

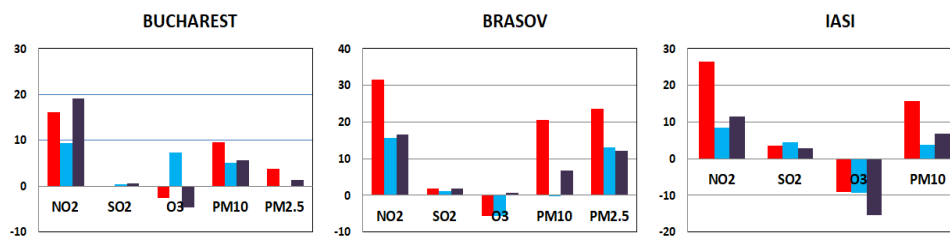


Fig. 6 – Urban impact on suburban areas for Bucharest, Brasov and Iasi in 2020 (red bars-before lockdown, blue bars-during lockdown, dark purple bars-after lockdown).

Present results point to the narrowing of the urban-suburban differences and this seems to be due to the rapid development of the suburbs around the medium to larger-size cities. This type of result was recently found in Ploiesti [33] but also worldwide, e.g. [22].

3. CONCLUSIONS

Current study was focused to assess and provide sufficient information on the variation range of ambient concentrations of major air pollutants PM_{10} , $\text{PM}_{2.5}$, NO_2 , CO , SO_2 , O_3 in the urban Romania. In the context of decreasing emissions of

major pollutants recently showed by Maranzano (2022) (pollutant emissions analyzed by 2017) at European scale [34] and in Romania [7, 19], analyzed by Nastase et al. (2018) - pollutant emissions data analyzed up to 2014, and by Iorga (2021) - emissions data analyzed up to 2018, present study shows (1) very different and heterogeneous pollution levels at the Romanian scale, (2) evidences pollutant-specific hot- and cold-spots across the country, and (3) quantifies comparatively the intra- and intercity variations in different seasonal regimes. The research also examined the impact of the lockdown measures imposed due to the COVID-19 pandemic on pollutants levels. Specific results were discussed at their appropriate section in context with other very recent reports worldwide, and mostly in Europe.

Present study has a slight limitation determined by the inconvenience of air quality type observations, which have a local character, by station type. We overcame this inconvenience as much as possible by averaging all measurements at the urban scale, but we must point to the necessity of a denser monitoring network in order to obtain a more accurate image of spatial concentration distribution. Additionally, the maps in Figure 4a-e are pictures of urban pollution impact distribution over the surrounding areas and they are subject to temporal evolution, where some random fluctuations due to meteorological factors must be considered. These random fluctuations were not taken into account here. Consequently, these maps showing spatial distribution of concentrations should be considered as an approximate urban influence over surrounding areas which are valid for certain periods, but subject to atmospheric changes [35]. However, the results are very useful to reveal the spatial impact of the pollution sources. Moreover, they are very useful when designing the local strategy development for limitation of air pollution in various degrees polluted environments.

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