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Temporal trends in the spatial-scale contributions to black carbon in a Middle Eastern megacity

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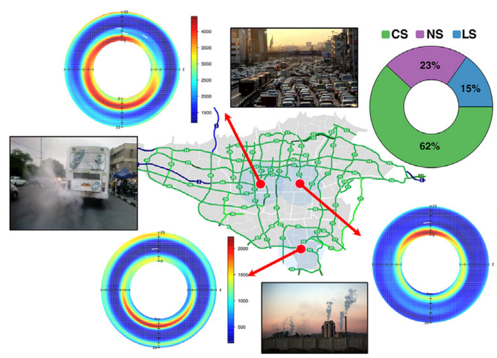
HIGHLIGHTS

- High-resolution changes in BC concentrations were examined to distinguish various spatial-scale contributions.
- City-scale BC emissions in Tehran are a major contributor to BC exposures.
- BC emissions on the neighborhood scale only contribute a minor fraction (~15%) to total BC concentrations.
- 62% of BC is part of the city emissions, and around 23% is transported from local surroundings.
- City-scale emissions contribute to wider pollution plume expansions and larger-scale transport.

GRAPHICAL ABSTRACT



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ABSTRACT

The analysis of high-resolution changes in black carbon (BC) concentrations was examined to distinguish and quantify various spatial-scale contributions to BC concentrations from nearby sources within 1 km distance to ranges of emission sources distributed over a larger city scale spanning tens of kilometers. Our analysis illustrated that BC emissions on the neighborhood scale only contribute a minor fraction (~15%) to total BC concentrations in the megacity of Tehran. Approximately 62% of the total black carbon is part of the city emissions, and around 23% is transported into the city from local nearby surroundings. Our analysis in highly polluted areas, including industrial and traffic hotspots in Tehran, demonstrated that the contributions of the urban mixture were relatively smaller than the average (~56%) in highly polluted areas; however, larger local-scale (~30%) contributions were observed in these areas. Our analysis in traffic hotspot areas also demonstrated significantly smaller contributions of BC from neighborhood surroundings (~9%). These results imply that the city-scale BC emissions in Tehran are a major contributor to BC exposures even in locations with local high-emitting sources. Polar annulus analysis of BC from city-scales in Tehran showed a mixture of hotspot locations ranging from north to easterly directions implying that city-scale emissions contribute to wider pollution plume expansions and larger-scale transport and vertical mixing corresponding to mixtures of emitters located further away.

1. Introduction

Air pollution situations caused by both gaseous and particulate matter (PM) components are considered as one of the most challenging

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environmental health concerns in Iran (Anenberg et al., 2012; Karimi and Shokrinezhad, 2021; Kermani et al., 2021). During the past recent years, Iran experienced severe and continuous air pollution situations, affecting millions of people in many cities. Air quality issues specifically, in regions with a high density of population, have been associated with both acute and chronic adverse human health impacts, causing cardiovascular and pulmonary diseases, asthma, allergy, or even associated with premature human mortality (Fazlzadeh et al., 2021; Heger and Sarraf, 2018; Krzyzanowski et al., 2005; Lelieveld et al., 2015; Sicard et al., 2019). The annual economic health costs associated with air pollution in Iran were estimated at approximately $2.6 billion.

Iranian authorities and provincial municipalities have implemented multiple air pollution control strategies to reduce air pollutants and $PM_{2.5}$ concentration (Atash, 2007; Heger and Sarraf, 2018; Taksibi et al., 2020). For instance, Tehran Municipality suspended some of the major businesses and restricted older trucks and buses for transportation. They also support the manufacture of vehicles with the latest technology to reduce air pollution, which will switch some of the 3400 old buses that commute in the streets today (Heger and Sarraf, 2018). Measurements at major areas of the city also showed that the daily average concentrations of PM would easily surpass the environmental standards.

Studies showed that acute and chronic exposure to $PM_{2.5}$ was responsible for approximately 4.2 million deaths globally (Cohen et al., 2017; Krzyzanowski et al., 2005). Reports of the World Health Organization (WHO) showed that particulate matters emitted from combustion-related sources (i.e., especially those from traffic emission) are more concerned with detrimental human health impacts (Luoma et al., 2021). Traffic emission sources contain complex mixtures of gaseous and fine particulate, including toxic compounds (Bond et al., 2013; Luoma et al., 2021).

Black carbon (BC), the carbonaceous part of PM, is a good indicator and a side-product of pollution from incomplete combustion and traffic emission sources (Luoma et al., 2021). In epidemiological studies, BC is considered as an indicator of fine particulate air pollution, and it is often associated with significant acute and chronic health outcomes (i.e., cardiovascular diseases) (Anenberg et al., 2012; Baumgartner et al., 2014; Janssen et al., 2011). BC can absorb solar radiation in all wavelength regions and reduces the albedo of reflecting surfaces (i.e., ice sheets and snow), and is therefore considered as one of the most significant warming agents in climate change (Luoma et al., 2021; Ramanathan and Carmichael, 2008; Stocker, 2014). Because BC is mainly emitted from combustion sources, its spatial variability could be significant in urban areas within a city (Van den Hove et al., 2020). Therefore, reducing BC emissions would have quite immediate results on the radiative forcing and decelerating the extent of global warming and improving air quality (Ding et al., 2016). Nevertheless, comprehensive studies conducted on BC in Tehran are still considerably uncertain, and our understanding of BC sources and their spatial and temporal variations are still poorly constrained in terms of being sufficiently comprehensive and quantifiable.

Source apportionment analysis in the city of Tehran showed that the majority of BC is emitted from diesel vehicle fleets in the city (Arhami et al., 2017; Esmaeilirad et al., 2020). Furthermore, the only available supplementary analysis conducted by Taheri et al. (2019) supported the hypothesis of the effects of heavy-duty diesel vehicles (HDDVs) on the BC fraction of $PM_{2.5}$ in Tehran. Their analysis showed that BC emission in Tehran is mainly from local sources, and their concentrations were primarily affected by emissions of diesel vehicles, including heavy-duty trucks (Taheri et al., 2019). Although these analyses were valuable in understanding BC emission sources in Tehran, contributions from different spatial scales are still unknown, hindering the comprehensive interpretations from measurements in all monitoring sites in Tehran (Watson and Chow, 2001). Studies showed that BC concentrations could vary substantially even in small geographical scales and nearby locations (Brantley et al., 2014). This significant difference could be attributed to the different source emissions occurring at various spatial scales (i.e., local (~100 m–1 km), neighborhood (~1–5 km), city (~5–50 km), regional (~50–1000 km), continental (~1000–5000 km) and global (>5000 km) spatial scales). Even a specific BC monitoring site can be largely affected by contributions from local and neighborhood-scale emission sources (e.g., emissions from high traffic congestion, industrial emissions, power plants, and fire events) (Watson and Chow, 2001). Herein, having access to the Tehran air quality and BC monitoring network allows us to closely examine BC profile in specific monitoring sites and determine different spatial scales contributions in each site. Short-duration measurements are analysed to determine and quantify contributions from different larger-scale contributions spanning from local nearby emission sources to a mixture of sources representing exposure from larger areas at city-scales. In this study, we analysed BC measured at various regions in Tehran (e.g., urban area, industrial and traffic hotspots) to investigate the spatial and temporal variation of different spatial-scale contributions of BC in various types of environments in Tehran. Our analysis will clarify how the proximity of different sources and spatial-scale contributions will affect BC concentration in each specific area during a long-term monitoring campaign in Tehran.

2. Materials and methods

2.1. Geographical location

Tehran, the capital of Iran, has a population of about 8.7 million in the capital and approximately 15 million in the greater metropolitan area, which is considered the most densely populated area in Iran and Western Asia (Bayat et al., 2019). The city is divided into 22 municipal districts and administrative centres. Tehran's climate is primarily defined by its geographic location and generally has a cold semi-arid climate characteristic and a typical Mediterranean climate precipitation pattern. Annual precipitation in Tehran usually occurs from late autumn to mid-spring. The highest temperatures are usually experienced during July with mean temperatures ranging from 26 °C to 37 °C while lowest temperatures are during January with average temperatures varying from −5 °C to 10 °C. Detailed information about the meteorological parameters and time series is provided in Fig. S1 in the SI.

2.2. BC monitoring

Tehran air pollution monitoring network consisting of a total of 35 monitoring stations belong to Tehran Air Quality Control Company (AQCC) and the Tehran Department of Environment. BC measurements in Tehran were conducted at various administrative centres and municipal districts in the city. Monitoring stations were selected to cover different types of environments in Tehran and were classified as being residential, traffic hotspots, urban, and industrial. Detailed information about the location of our measurement sites and their specific characteristics could be found in Fig. S2 and Table S1 in the SI. All the stations were equipped with black carbon monitoring equipment (Aethalometer AE33-Magee Scientific) since 2017.

The aethalometer is based on optical measurement, and hence the BC concentrations could be estimated from the light absorption of the particles. AE33 was operated at the flow rate of 5 L min^{-1}, and BC concentrations were estimated by utilizing a mass absorption coefficient (MAC) value. The filter model PN8060 was used in all of our AE33 instruments operated in our monitoring areas. AE33 measured the light attenuation of aerosol particles loaded on the filter at seven wavelengths (370, 470, 520, 590, 660, 880, and 950 nm). The increased filter loading and multiple scattering of the filter will result in the error of BC estimation by Aethalometers (Drinovec et al., 2015). The AE33 instrument is capable to automatically compensate for these errors by utilizing a dual-spot correction to real-time data. The sensitivity parameter (C) of 1.39 and the leakage parameter (Z) of 0.01 were implemented to our BC monitors in Tehran (Drinovec et al., 2017). We also applied a recommended MAC value of 7.77 $m^2 g^{-1}$ at 880 nm for AE 33 instrument (Drinovec et al., 2015).

In this study, we will present the results of a 3-year continuous BC measurement from January 2018 to December 2020 at all of the monitoring sites in Tehran. The measurement sites were classified into four categories, including Residential (RES), Roadway Traffic Hotspot (RTH), Urban (URB), and Industrial and mixed (IND) (Table S1).

2.2.1. Residential area (RES)

RES area is located in the central part of the city with predominantly official land use. Also, there is a highway adjacent to this station. Based on the characteristics of the environment and the area, we consider this station as being a residential and urban station (RES).

2.2.2. Urban area (URB)

URB is located inside the campus of the Sharif University of Technology. The campus is adjacent to Azadi Street in the south, Jenah street in the west, and Sheikh Fazlullah Nouri in the north. Azadi Terminal, which is a gathering place for intercity and BRT buses, is also in the vicinity of this station. The dominant wind direction from the west could also transport the urban pollution to other areas in Tehran. Therefore, we consider this station as an urban station (URB) based on the station's location and the characteristics of the environment, which is the recipient of the urban mixture.

2.2.3. Roadway traffic area (RTH)

RTH was located at the intersection of Sadr and Modares highways and also near the exit of the Niayesh tunnel. Therefore, it is expected that the data measured at this station will be greatly affected by high traffic congestion. As a result, this station is considered a roadway traffic hotspot station (RTH).

2.2.4. Industrial area (IND)

IND was located in the south of Tehran, in the vicinity of several emission sources. Tehran ring road, where heavy HDDV traffic and the BRT terminal near the station. Besides, various dominant industries, including the Tehran refinery complex, are also adjacent to the station. Based on the characteristics, we consider the area as an Industrial and mixed emission (IND).

2.3. Spatial-scale contributions

In this research, we employed a successive moving average subtraction method to resolve different spatial-scale contributions. The method has been successfully applied to resolve spatial-scale contributions of elemental carbon in Mexico city (Watson and Chow, 2001). In the successive moving average subtraction method, the hourly average BC of 60 values surrounding the 1-min interval BC measurements is calculated in each monitoring site in Tehran. The new hourly averaged data is then compared with the original 1-min data. The hourly-averaged data is retained only in case that it is less than the original 1-min BC data. The second step estimates the 30-min interval BC with averaging 30 values surrounding the new database in each site. The 30-min database is again compared with the original 1-min data, and the new averaged data is retained if it is less than the original data. The 15-min average is finally calculated from the new database to obtain short-duration baseline values for all monitoring sites. The local-scale contributions at each site were then calculated by subtracting the measured value at each site by the corresponding baseline values. The city-scale contributions of each site were determined by the baseline at URB. However, the neighborhood-scale contributions at each site were calculated by subtracting the baseline values of each site from the baseline at URB.

In this research, URB data was considered as the reference area. URB was located inside the campus of Sharif University in Tehran and close to some important urban emission sources. We believe that the URB area was a recipient of an urban mixture. On the other hand, RES area can only characterize minor fractions from urban emission clouds and therefore cannot show all the city emission characteristics.

2.4. Polar annulus analysis

Herein, we employed a Polar Annulus analysis to visualize the temporal aspects of each spatial-scale BC concentration by wind direction. These plots can demonstrate significant details concerning potential sources. Visualizing as an annulus prevents the complexity in interpreting values toward the origin of the plot. The analysis is similar to the polar plot analysis described in detail in Grange et al. (2016); however, the Polar annulus displays how the contributions of a specific source would vary against wind direction and time. The time in this analysis could be interpreted in various ways, including monthly, weekly, hourly, and seasonally. Herein we analysed the data by hourly average. In general, plotting polar annulus involves using wind direction as the polar axis and aggregating measurement data into wind direction bins. A smoothed interface is plotted with these intervals using a generalized additive model (GAM) to fit a continuous surface representing polar coordinates. In this study, we used *openair* package (version 2.8-1) within the R environment (version 4.0.2) for the polar annulus analysis (Carslaw and Ropkins, 2012).

3. Results

3.1. Monthly variation of spatial-scale

Fig. 1 shows the monthly variation trends of each spatial-scale contribution of BC from 2018 to 2020 for all of our measurement sites. According to Fig. 1, monthly contributions from city-scale BC ranged from 38% to 81% during the whole study period. In general, we observed higher contributions of city-scale emissions in RES (65.7%) areas. Besides, significantly lower contributions of city-scale emissions were found at IND (53%) and RTH (59%). Furthermore, contributions from neighborhood-scale emissions were larger at IND (26.5%, 1163.4 ng/m^3) compared with RES (15.6%, 641.4 ng/m^3) and RTH area (9%, 448.6 ng/m^3) (Fig. 1).

RTH was a traffic hotspot located at the intersection of main highways in Tehran with high traffic volumes and a large density of BC plumes in the area. Significantly higher contributions from nearby emission sources (i.e., from adjacent highways) were observed compared with larger-duration neighborhood surroundings. We observed few negative cases of neighborhood-scale emitters (i.e., the slightly negative cases were not statistically significant), specifically at the traffic hotspot (RTH) and residential (RES) areas, mainly because the baseline levels at URB were slightly higher than the corresponding RTH and RES levels (Fig. 1).

Local-scale contributions however, was largest at RTH with 1399.1 ng/m^3 (31.7%) followed by IND (25.8%, 1286.5 ng/m^3), and RES stations (18.6%, 728.2 ng/m^3) (Fig. 1). IND and RTH are located close to high emitting plumes from traffic and industrial sources, and therefore, higher contributions from shorter duration emissions (i.e., local-scale emissions) are expected. By contrast, the RES station is located far from the main highways or major traffic emission sources, and therefore, lower contributions from short-duration and nearby pollution plumes (i.e., local-scale contributions) are observed. Monthly variation trends of the city- and local-scale BC emissions at URB are also provided in Fig. S3 in the SI.

Fig. 2 illustrates the variation trends of average monthly contributions of city, neighborhood, and local-scale BC for all of our monitoring sites during the three years study period. We did not observe significant monthly variation patterns for city-scale contributions in Tehran with an average concentration of 2546.31 $\pm$ 459.11 ng/m^3 (62%). The city-scale contribution was relatively larger during autumn and winter and lowered during the spring (Fig. 5). BC's city-scale contributions were largest during December (with approximately 3200 ng/m^3) and January

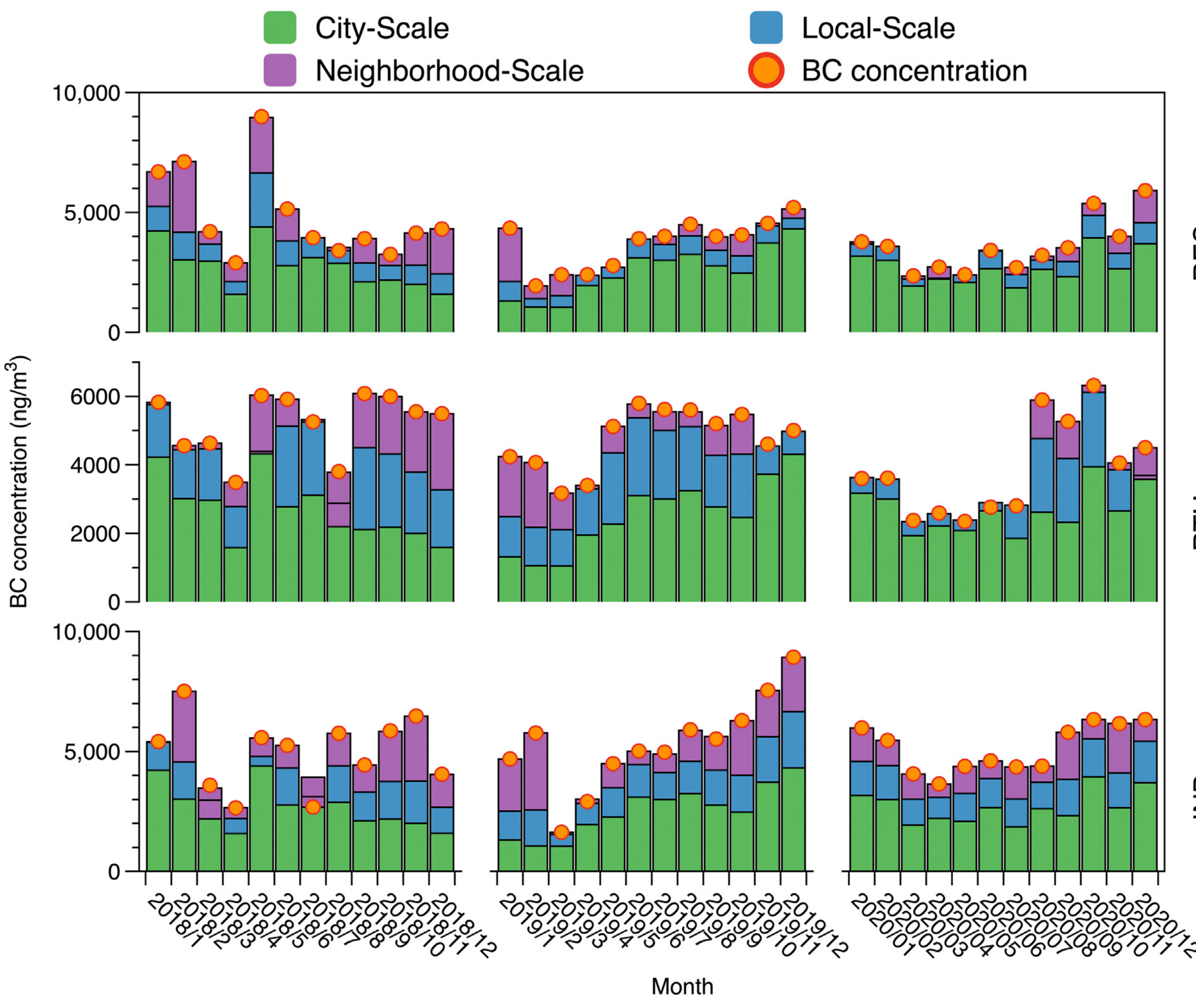

Fig. 1. Monthly-averaged contributions from city-, neighborhood-, and local-scale emitters of black carbon in all of our monitoring areas in Tehran using a 1-hr moving average.

(2900 ng/m^3). The lowest contributions of city-scale emission were observed during July (1800 ng/m^3). Local-scale contributions did not show clear monthly variation profiles. We observed relatively large contributions of BC related to the local-scale emission sources for highly polluted areas in Tehran, averaging at around 1024.15 ng/m^3 (22.7%).

IND and RTH areas contributed approximately 65% of the total local-scale BC profile in Tehran (1286.5 $\pm$ 372.3 (25.8%) ng/m^3 for IND and 1399.1 $\pm$ 407.1 (31.7%) ng/m^3 for RTH). The contributions of local-scale emissions of RES and URB areas were 728.2 $\pm$ 145.2 (18.7%) and 682.8 $\pm$ 131.2 (21%) ng/m^3 for RES and URB (Fig. S3), respectively. IND was located far from the city centre but largely influenced by traffic and industrial sources. Also, the RTH area captures major traffic emissions of two main highways. These nearby emission sources (less than 1 km distant) affect short-duration measurements at each monitoring site that could represent exposures from local-scale sources. However, relatively low contributions of BC were generated from neighborhood-scale emission sources in Tehran with average concentrations of 751.1 $\pm$ 230 ng/m^3 (15.3%). Approximately 50% of the neighborhood-scale BC profile in Tehran was contributed by the IND area (3418.3 $\pm$ 1100.3 ng/m^3) (Fig. 2). Various mixtures of emission sources are located in the vicinity of the IND area, and therefore, we expect significant contributions of BC from neighborhood surroundings.

RTH contributed 266.7 $\pm$ 51 ng/m^3, and RES accounted for 616.6 $\pm$ 159.2 ng/m^3 of neighborhood-scale contributions in Tehran. Lower neighborhood-scale contributions in Tehran illustrate that our monitoring sites could largely reflect BC exposures from surrounding nearby (~1 km) and larger city-scale emissions. Generally, for continuous point-source emissions and high emitting plumes, wider pollution plume expansion (typical of city-scale) could happen due to large-scale transportation and vertical mixing that correspond to more slowly varying BC concentrations. According to Pasquill-Gifford curves, horizontal plume magnitudes of 50–500 m and vertical extents of 20–1000 m at 1 km from the emitter could be expected. Also, horizontal plume dimensions of approximately 500–2000 m and vertical extents of around 50 m to the depth of the mixed layer would happen at a distance of 10 km from the source (Watson and Chow, 2001). Therefore, shaving high emitting plumes from short-duration local-scale sources leave the contributions that correspond to mixtures of emitters located further away (will be discussed more in the following sections).

3.2. Weekly variation

Fig. 3 showed weekly variation patterns of spatial-scale BC contributions in all of our monitoring sites during the three years study period. We observed similar weekly variation profiles with different spatial-scale contributions of BC in Tehran. As described earlier, lower local and neighborhood-scale contributions were observed in weekly profiles in Tehran. The highest contributions of weekly local-scale BC were related to IND (1284.4 ng/m^3) and RTH areas (1425.1 ng/m^3). The lowest local-scale BC, however, was observed at RES with average weekly contributions of 705.4 ng/m^3. Besides, lower BC contributions were observed from neighborhood-scale emissions (567.8 ng/m^3) in Tehran (Fig. 3). Weekly variation patterns of BC emissions from the city and local scales at URB are also provided in Fig. S4 in the SI.

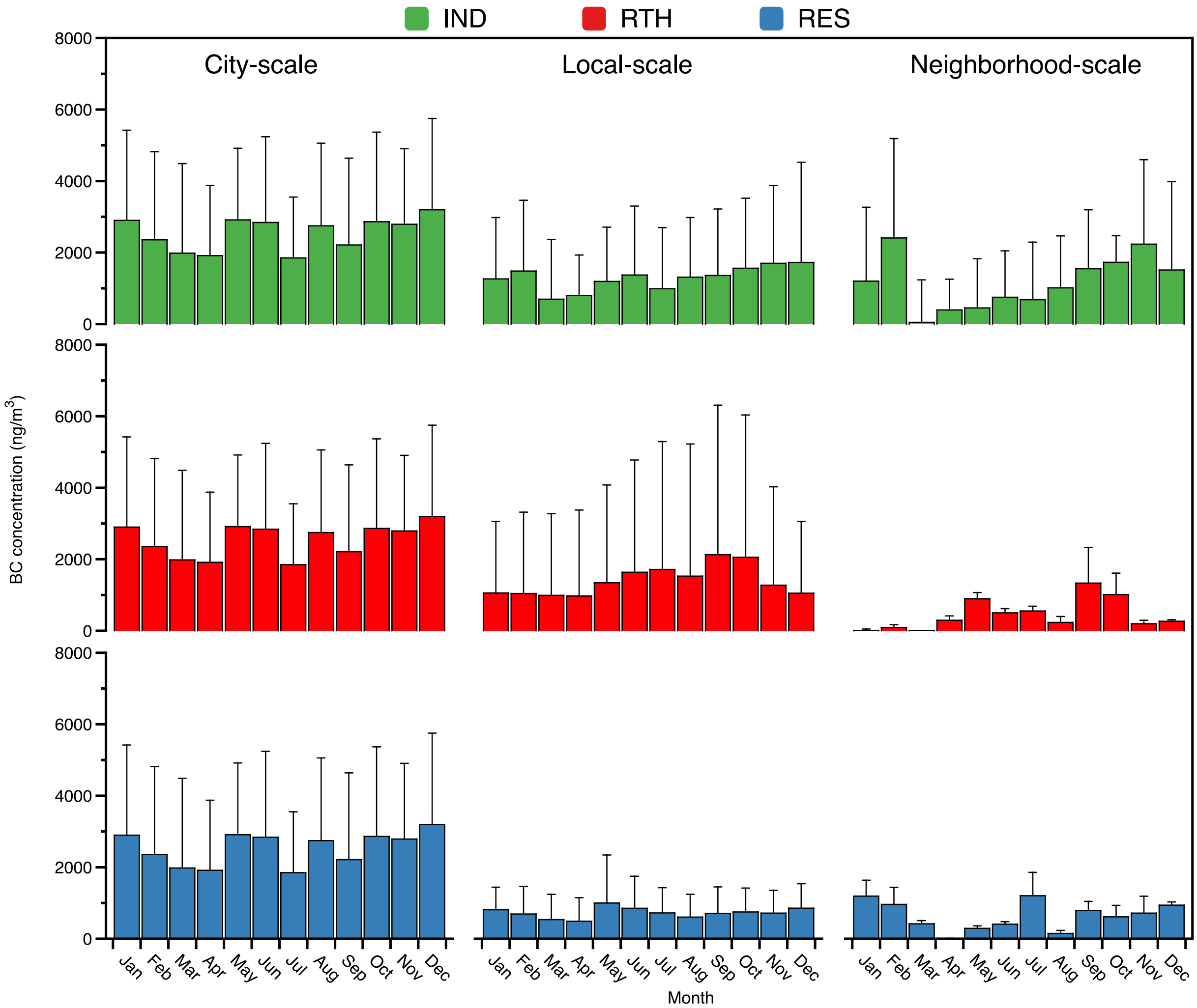

Fig. 2. Monthly-averaged contributions from different spatial-scale emitters of black carbon in all of our monitoring areas in Tehran for the 3 years study period.

We did not observe a significant difference in the city (2671 ng/m^3), neighborhood (631.1 ng/m^3), and local-scale (1082.2 ng/m^3) contributions of BC during weekdays (i.e., Sunday to Thursday); however, contributions of all spatial scales were significantly smaller during Friday and Saturday (2263, 471.5, and 885.5 ng/m^3 for all city, neighborhood and local-scales respectively). This could be due to the reduced background activities and traffic emissions in Tehran during the weekends.

The magnitude of reductions among different spatial-scale BC contributions was the lowest from larger-duration city-scale emissions (approximately 20%), which was lower than those from local (28%) and neighborhood scales (35%) (Fig. 3). This illustrated that the reduced emissions during weekends would be less effective in reducing BC emissions from large-duration plumes and urban mixture (city-scale contributions) in Tehran. However, these reductions would be more beneficial in reducing short-duration exposures from nearby areas (local and neighborhood scales). Similar to Fig. 2, large city-scale contributions of BC were observed for all of our monitoring sites, averaging around 2554.4 ng/m^3 (61.4%). Local and neighborhood-scale emissions contributed to 1025.98 ng/m^3 (24.6%) and 585.50 ng/m^3 (14%) of total BC in Tehran, respectively.

3.3. Diurnal variation

Fig. 4 illustrates the hourly averaged BC contributions from different spatial scales over the 3-years study period. Overall, similar diurnal variation profiles were observed for the city and local-scale emissions in Tehran. However, closely looking at the diurnal variations of each specific monitoring site, different profiles were observed specifically for neighborhood and local-scale emissions of IND and RTH (will be discussed in detail in the following section). Diurnal variation patterns of city and local-scale emissions at the URB area are also provided in Fig. S5 in the SI.

As described earlier, low BC contributions from the neighborhood-scale (570.17 ± 436.61 ng/m^3) were observed. BC contributions from local (1030.2 ± 541 ng/m^3) and city-scale (2569.5 ± 928.6 ng/m^3) emissions, however, were larger compared with neighborhood-scale emission (Fig. 4).

As observed in Fig. 4, a similar general pattern is expected to follow for BC concentrations in Tehran. During the first hours of the day, BC concentrations slightly increased or stayed relatively stable mainly because of the lower Planetary Boundary Layer (PBL) heights and surface cooling and the activities of HDDVs that were admitted into the streets (until 6:00 local time). The magnitude of early hours increase was relatively more pronounced for shorter duration emission than the larger duration city-scale emissions. After the sunrise, Higher PBL heights and the prohibition of the activities of HDDVs resulted in lowering BC concentrations, and their mixing ratios remained low until night. Given the Tehran traffic rules, the diurnal decrease and increase patterns are expected to follow approximately around 6:00 and 22:00 local times.

Fig. 5 illustrates the hourly averaged BC contributions from different spatial scales for every season at a typical industrial area with mixed

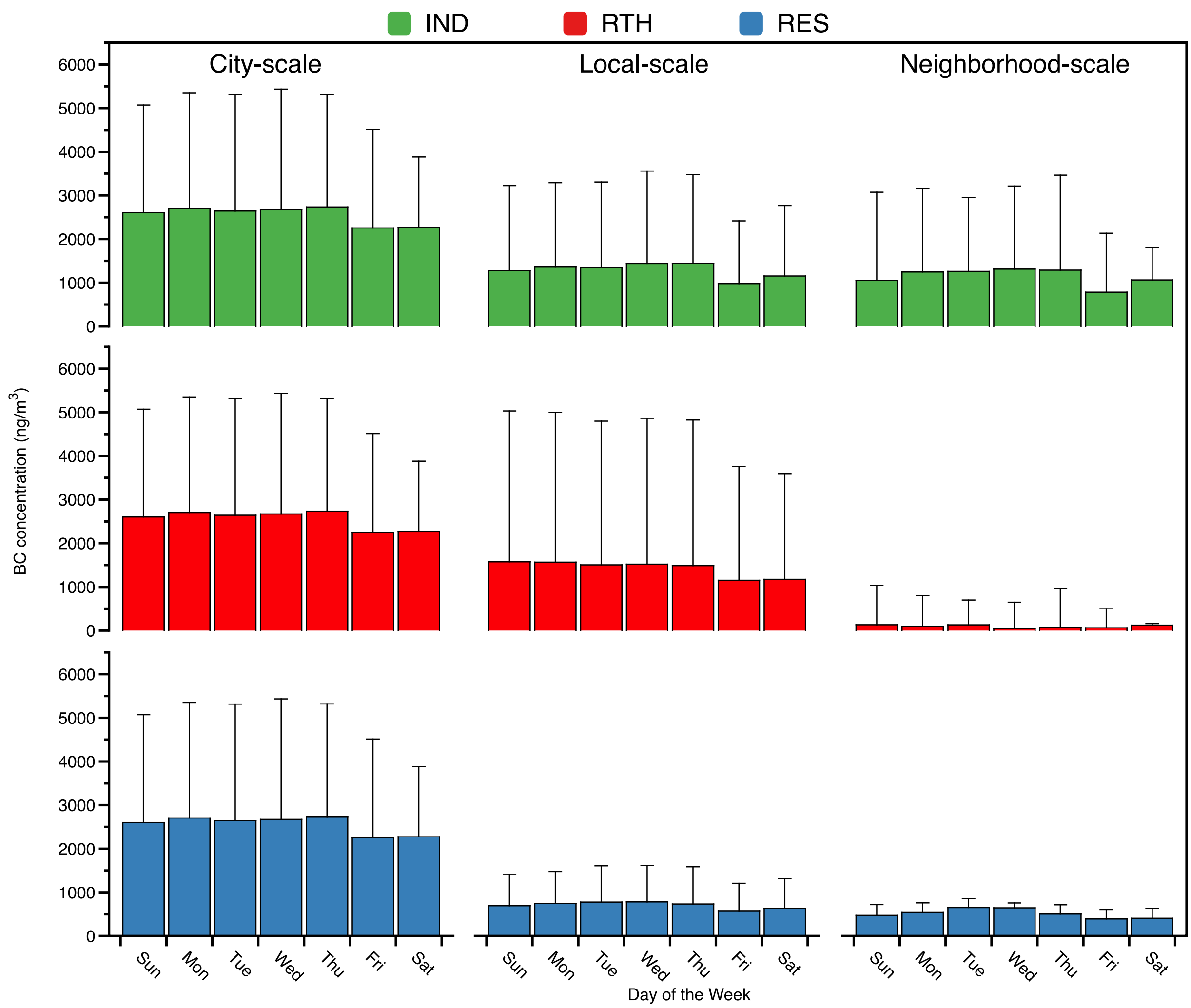

Fig. 3. Weekly-averaged contributions from city-, neighborhood-, and local-scale emitters of black carbon in all of our monitoring areas in Tehran for the 3 years study period.

emission sources (IND) in Tehran. Hourly averaged spatial scale contributions of other areas are also provided in Figs. S6, S7, and S8 in the SI.

We observed relatively higher contributions of city-scale BC emission in Tehran during autumn (2770 ng/m^3) and winter (2800 ng/m^3) compared with spring (2200 ng/m^3) and summer (2700 ng/m^3). For IND, we observed the highest contributions of local-scale emissions during autumn. Besides, significantly larger neighborhood scales at IND were observed in autumn and winter. For RTH again, the highest local and neighborhood-scale emissions were also observed during autumn compared with other seasons. In general, local-scale emission in Tehran was highest during autumn (1243.4 ng/m^3). Neighborhood-scale emissions also experienced larger contributions during autumn (1072.7 ng/m^3) compared with spring (41.4 ng/m^3), summer (35.3 ng/m^3), and winter (851.8 ng/m^3) in Tehran. Relatively similar results were also observed for other monitoring sites in Tehran. Studies show that PBL is the key factor in determining the concentrations of spatial-scale BC in Tehran. PBL tends to be smaller during the colder seasons, and therefore we expect larger concentrations of BC as the result of the pollutants being trapped in a limited area (Taheri et al., 2019).

As described in previous sections, diurnal variation profiles of the neighborhood and local-scale emissions at IND were different from city-scale (Fig. 5). Diurnal variations of city-scale at IND followed the same general BC profile rules of Tehran described previously. However, local-scale BC increased significantly during the first hours of the day and reached their maximum level at 7:00 local time and decreased to their minimum levels during the day. During the afternoon hours, local-scale BC at IND started to increase again at around 17:00 local time and reached their maximum level at 21:00. Neighborhood-scale diurnal profiles of IND were similar to that of local-scales; however, 1-hour time difference was observed in neighborhood-scales of IND area.

3.4. Polar annulus analysis

Polar annulus analysis was carried out to further analyze the temporal aspects of a spatial-scale BC by wind direction (Fig. 6). Polar annulus analysis of Tehran showed that the increase in city-scale BC during the first hours of the day was responsible for a series of winds ranging from north to easterly directions. However, the early hours' increase (1:00–4:00 local time) was more important for northerly winds. In contrast, the increase from (4:00–9:00 local time) was mainly important for southeasterly/southwesterly winds (Fig. 6). City-scale contributions mainly depend on large-scale transport and vertical mixing rather than the immediate narrow plumes that would emerge as spikes. Therefore, urban mixture contributions would usually correspond to a wide range of mixture of emitters located further away from the location. Polar annulus analysis of city-scale BC for all areas also showed similar patterns as that observed in Fig. 5.

Polar annulus analysis of local-scale emissions at the IND area showed that the increase during 7:00 local time was important for southwesterly to southeasterly wind directions. However, relatively higher night-time concentrations in this area (at around 21:00) were

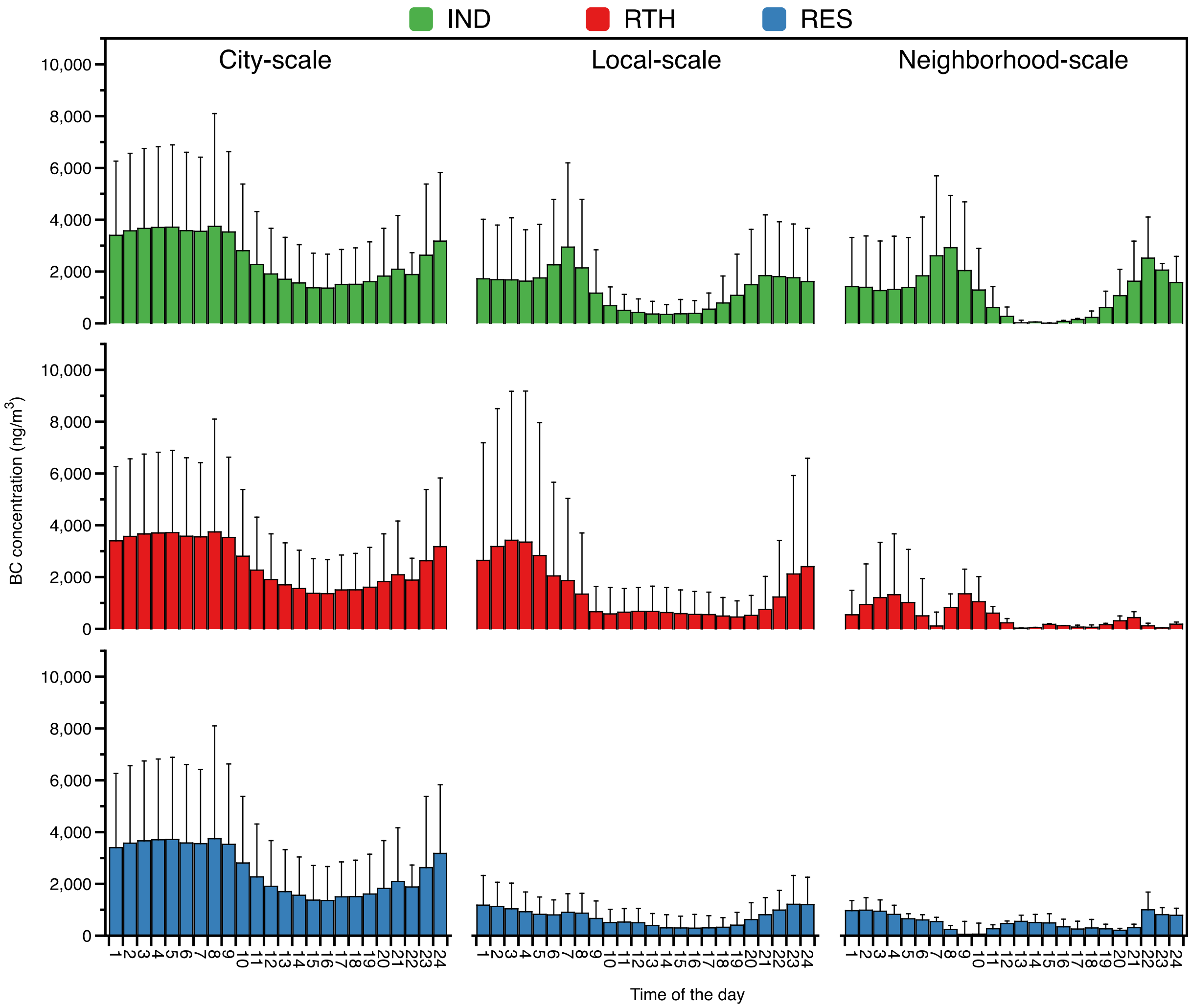

Fig. 4. diurnal variations of contributions from city-, neighborhood-, and local-scale emitters of black carbon in all of our monitoring areas in Tehran for the 3 years study period.

important for northern directions (Fig. 6). However, analysis of neighborhood-scale emissions at IND illustrated that northwesterly wind directions in addition to the southwesterly to southeasterly directions were confirmed as important emissions during the early hour's increase. Northwesterly directions again were confirmed as important hotspot sources for night-time peaks (22:00 local time) (Fig. 6). IND area is close to the Tehran refinery complex which is located at the south of the station, where significant industrial activities are carried out. Also, the area is close to Tehran's ring road, where HDDVs are admitted without limitations (located north of the study area). These emission sources would largely contribute to high local and neighborhood-scale emissions in the IND area. Polar annulus analysis of IND for neighborhood-scale (Fig. 6) also showed north-western directions as hotspot emission sources during both morning and afternoon peaks. We assume that high emitting plumes of traffic from Tehran's ring road located at the north/northwest of the monitoring site would contribute to higher concentrations of local and neighborhood-scale emissions at the IND area during the morning hours (Fig. 6). We also observed larger contributions from southwesterly to southeasterly directions for both local and neighborhood-scale emissions during morning measurements. We assume that high emitting plume from the Tehran refinery complex (located in southern parts of the area) would contribute to larger local and neighborhood-scale emissions at IND.

Similar profiles were observed at RTH, RES, and URB areas (Figs. S9, S10, and S11). For instance, at RTH, local-scale BC during the day increased sharply to the maximum levels at 3:00 local time. Contributions were then decreased again and reached their minimum level at 9:00 local time when concentrations remained relatively stable over the day. Local-scale contributions started to increase again at around 21:00 and reached the maximum at 24:00 local time. Polar annulus analysis of the RTH illustrated that the early morning increase of local-scale emissions was due to the northerly winds (Fig. S9). Night-time peaks of local-scale BC at the RTH area were also important for northerly winds. Polar annulus analysis of neighborhood-scale contributions of the RTH area showed similar sources as that observed for local-scale (Fig. S9). This demonstrates the possibility of return flow of contribution from local-scale emissions at RTH station. RTH was the intersection of two main highways. Therefore, we expect that those nearby high emission sources can contribute to significant local-scale sources. The contributions of neighborhood-scale emissions in this area were significantly smaller compared with local nearby emission sources. This demonstrates that the majority of short-duration sources were from nearby local sources, and neighborhood surroundings only accounted for minor contributions in traffic hotspot areas.

We observed relatively larger contributions of local-scale emission (for both morning and afternoon peaks) for the RES area mainly related to the northern parts of the monitoring site (Fig. S10). The polar annulus of neighborhood-scale sources also illustrates northern directions as important hotspot areas. RES is located in a downtown area in Tehran and is close to one major highway located approximately 700 m away at the north of the site. We expect that the emission plumes from the highway

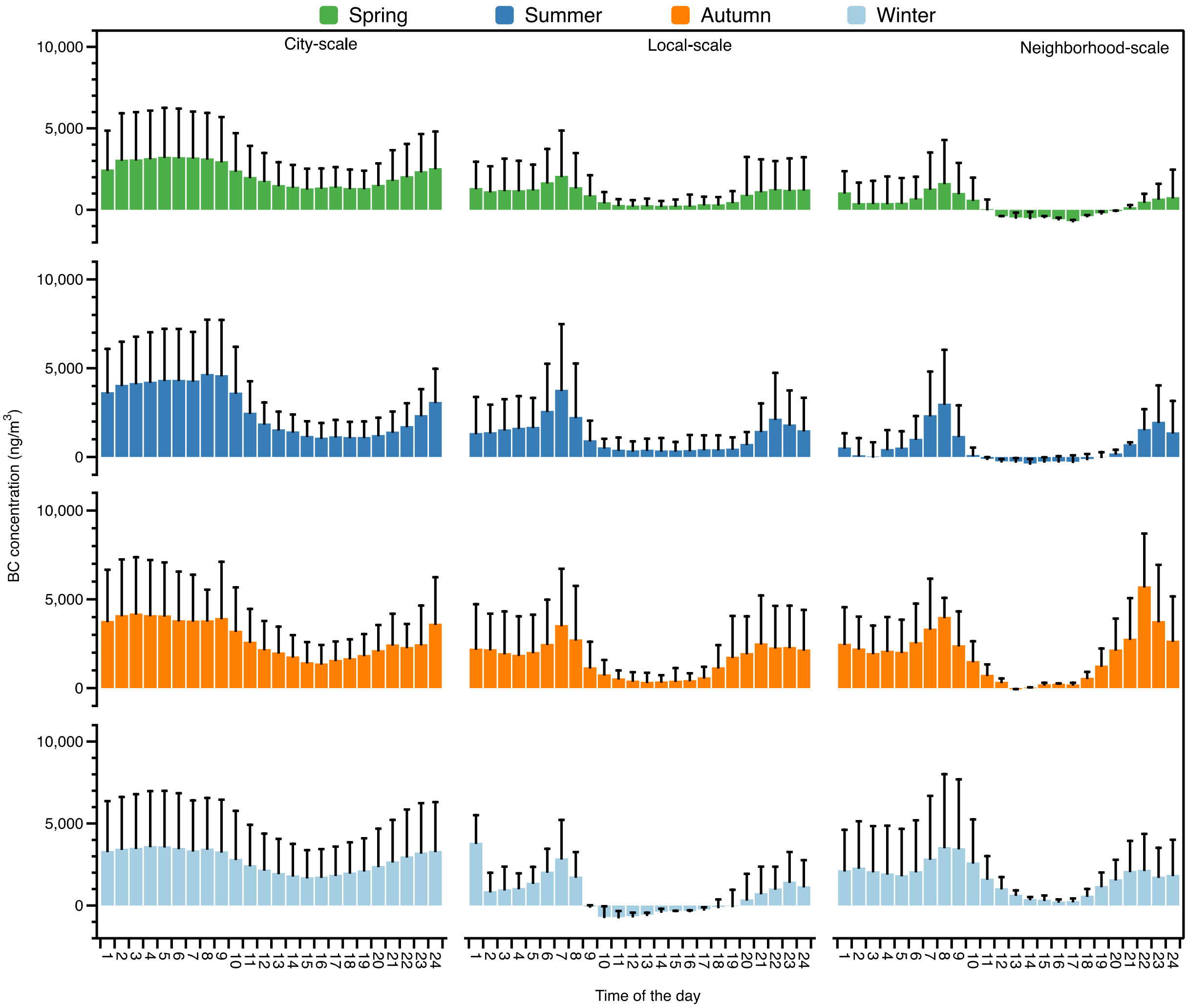

Fig. 5. Diurnal variations of contributions from city-, neighborhood-, and local-scale emitters of black carbon at typical industrial areas (IND) in Tehran for the 3 years study period.

located at the north would contribute to larger local and neighborhood-scale emissions in RES. Polar annulus analysis also confirmed larger contributions from the northern directions as hotspot sources of both local and neighborhood-scale (Fig. S10).

For the URB area, we observed a mixture of emission sources ranging from west to southwesterly directions as hotspot sources for their emissions (Fig. S11). URB is a recipient of the urban mixture and is surrounded from the east, north, and west by three roadways located close to the area. During the rush hours, approximately 6000 vehicles pass through these roadways. Also, Tehran's key bus terminals are located at the west of the area. The dominant wind blows from the west that brings pollutants from adjacent areas at URB. The domestic

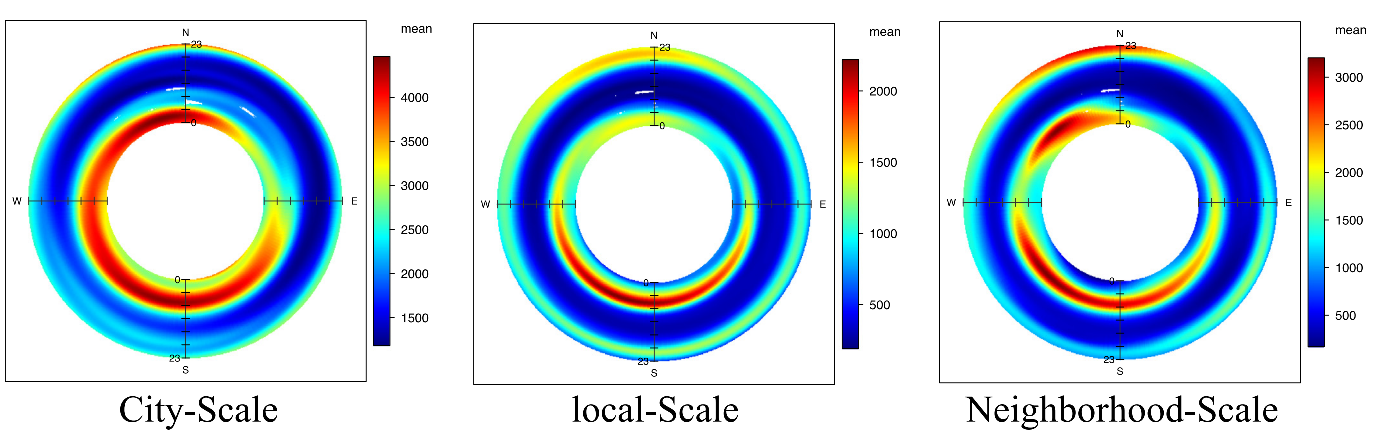

Fig. 6. Polar annulus analysis of diurnal variations of contributions from city-, neighborhood-, and local-scale emitters of black carbon at typical industrial areas (IND) in Tehran for the 3 years study period.

Mehrabad airport is also located southwest of the station. Polar annulus analysis also confirmed that a mixture of emission sources ranging from west to southeast parts of the URB area as important hotspot locations (Fig. S11).

4. Conclusions

In this research, the most comprehensive Black Carbon (BC) study was implemented in the megacity of Tehran. We accessed the data of Tehran's long-term BC monitoring network consisting of aethalometers installed at various types of environments in the city (i.e., residential, traffic hotspots, urban background, and industrial). We illustrated how BC's different spatial-scale contributions in Tehran could be estimated with the successive moving average technique. Also, we demonstrated that how short-duration local-scale emissions of BC can be distinguished from different larger-scale contributions spanning from sources within a kilometer to a mixture of the source at tens of kilometers of the monitoring sites (Watson and Chow, 2001). The method has been applied to monthly, weekly, and hourly averaged periods from January 2018 to December 2020. Our analysis illustrated that 62% of BC in Tehran were from city-scale, 22.7% from local-scale, and only 15.3% from neighborhood-scale emissions. City-scale contributions were significantly smaller in representative industrial sites in Tehran and accounted for approximately 52% of total BC. Neighborhood-scale emission in the industrial areas also accounted for approximately 26.5%, which was significantly larger than the average neighborhood-scale BC in Tehran. Our analysis of the traffic hotspot locations illustrated relatively smaller city-scale (59.2%) and larger local-scales (31.6%) of BC in Tehran. Neighborhood-scale emissions at high traffic areas were significantly smaller than the average (9.1%). This indicates that BC monitoring networks in Tehran can represent significant exposures from large urban mixture and city scales, albeit with various local high emitting plumes (i.e. plumes from traffic exhaust) that could affect the monitoring sites.

Polar annulus analysis in Tehran also confirmed that city-scale BC in Tehran originates from a mixture of emission sources ranging from north to easterly directions as hotspot sources for their emissions. This illustrates that the urban mixture mostly relies on large-scale transport and vertical mixing rather than the emissions from surrounding or nearby emission sources, and therefore city-scale emissions would contribute to wider pollution plume expansions and large-scale transport and vertical mixing related to the mixtures of emitters located further away from the area. Our analysis of local and neighborhood-scale contributions with polar annulus analysis mainly pointed to the local and nearby emission sources surrounded by the monitoring areas.

CRediT authorship contribution statement

Bijan Yeganeh: Writing – original draft, Conceptualization, Methodology, Software, Investigation, Supervision, Writing – review & editing. **Reza Bashiri Khuzestani:** Writing – original draft, Conceptualization, Methodology, Data curation, Software, Visualization, Investigation, Supervision, Writing – review & editing. **Ahmad Taheri:** Data curation, Methodology, Conceptualization, Investigation, Writing – original draft. **James J. Schauer:** Conceptualization, Supervision, Investigation, Writing – review & editing.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.scitotenv.2021.148364.

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