Atmospheric Environment 269 (2022) 118849

Contents lists available at ScienceDirect

Atmospheric Environment

journal homepage: www.elsevier.com/locate/atmosenv

Aerosol number fluxes and concentrations over a southern European urban area

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HIGHLIGHTS

- Eddy-covariance method was used to evaluate aerosol number fluxes in an urban area.
- Daily, weekly and seasonal patterns of aerosol flux and concentration were analyzed.
- The studied urban area acted as a net source for atmospheric aerosol particles.
- Urban aerosol flux is primarily driven by traffic emissions.
- Domestic heating and biomass burning significantly impact urban aerosol in winter.

ARTICLE INFO

Keywords:
Particles flux
Eddy covariance
Aerosol number concentrations
Urban air quality

ABSTRACT

Although cities are an important source of aerosol particles, aerosol number flux measurements over urban areas are scarce. These measurements are however important as they can allow us to identify the different sources/sinks of aerosol particles and quantify their emission contributions. Therefore, they can help us to understand the aerosol impacts on human health and climate, and to design effective mitigation strategies through the reduction of urban aerosol emissions. In this work we analyze the aerosol number concentrations and fluxes for particles with diameters larger than 2.5 nm measured by eddy covariance technique at an urban area (Granada city, Spain) from November 2016 to April 2018. This is the first study of particle number flux in an urban area in the Iberian Peninsula and is one of the few current studies that report long-term aerosol number flux measurements. The results suggest that, on average, Granada urban area acted as a net source for atmospheric aerosol particles with median particle number flux of 150×10^6 m^{-2} s^{-1}. Downward negative fluxes were observed in only 12% of the analyzed data, and most of them were observed during high aerosol load conditions. Both aerosol number fluxes and concentrations were maximum in winter and 50% larger than those measured in summer due to the increased emissions from domestic heating, burning of residual agricultural waste in the agricultural area surrounding the site, as well as to the lower aerosol dilution effects during winter. The analysis of the seasonal diurnal variability of the aerosol number concentration revealed the significant impact of traffic emissions on aerosol population over Granada urban area in all seasons. It also shows the impact of domestic heating and agricultural waste burning emissions in winter as well as the influence of new particle formation processes in summer and spring seasons. Closer analysis by wind sector demonstrated that both aerosol concentrations and fluxes from urban sector (where high density of anthropogenic sources is located) were lower than those from rural sector (which includes agricultural area but also the main highway of the city). This evidences the strong impact of aerosol emissions from traffic circulating on the highway on aerosol population over our measurement site.

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https://doi.org/10.1016/j.atmosenv.2021.118849
Received 25 May 2021; Received in revised form 29 October 2021; Accepted 10 November 2021
Available online 14 November 2021

1. Introduction

Urban areas are sources and sinks of a variety of air pollutants that potentially affect the climate and human health. For these reasons, the European Commission has implemented several stringent emission control measures in order to reduce air pollution and improve air quality in urban areas and hence protect human health. As a result of these abatement policies, concentrations of some air pollutants (such as SO_2) have shown a large decrease along the last decades, resulting in general improvement of air quality with respect to these pollutants across the European Union. However, some European cities are still being exposed to high level of some pollutants such as NO_2 and aerosol particles (e.g., Casquero-Vera et al., 2019), and the impact on human health, due to exposure to these pollutants, is still significant (e.g., Jonson et al., 2017; EEA, 2018). In this sense, atmospheric aerosol particles are still one of the main pollution concerns in European urban areas (EEA, 2018).

To develop efficient control strategies and mitigate negative aerosol effects on health and climate through reduction of emissions, better understanding of the spatial and temporal variability of aerosol sources and sinks and their strengths in different urban areas in the world is of critical importance. Although the different sources of aerosol particles in urban areas are well known, their emission strength, spatial and temporal variability, and contribution to particle concentrations is highly uncertain (IPCC, 2013). Aerosol particle flux measurements can provide important quantitative information about the strength and spatial distribution of aerosol sources and sinks in urban areas (e.g., Järvi et al., 2009; Deventer et al., 2015, 2018). The eddy covariance technique (EC) allows a direct and accurate measure of aerosol fluxes, providing important information about the spatial variability of aerosol sources within a city and the net emission due to those sources (Mårtensson et al., 2006). However, although cities are an important source of aerosol particles, EC flux measurements in urban areas are still scarce and most of previous studies have been based on short-term measurement campaigns (e.g., Dorsey et al., 2002; Mårtensson et al., 2006; Schmidt and Klemm, 2008; Martin et al., 2009). Furthermore, most of these studies were conducted in central and northern European cities, with only a handful studies in southern Europe where the anthropogenic activities and the meteorological conditions are different to those in central and north of Europe (Contini et al., 2012; Conte et al., 2018, 2021; Donateo et al., 2019). Since urban particle fluxes largely depend on the characteristics of the site, more urban particle flux data from different cities are needed to fully understand the role of urban sources and their spatial variability.

In this study, we present the results of aerosol number concentrations and fluxes measured by eddy covariance method at the southern European urban area of Granada, Spain, from November 2016 to April 2018. In Granada city, located on mountainous area, the emission characteristics and meteorological conditions are distinct to those in central and north of Europe and thus aerosol number fluxes can vary from those measured in other cities. The main aims of this work are to explore the driving processes of the diurnal, weekly and seasonal changes in the aerosol number concentration and fluxes in Granada, and to identify the different sources/sinks of aerosol particles. The impact of different surface cover types on aerosol number concentration and flux is also investigated.

2. Measurement site

Measurements presented here were carried out in the frame of ACTRIS (Aerosol, Clouds and Trace Gases Research Infrastructure) from November 2016 to April 2018 over an urban site in Granada city (37.16° N, 3.58° W, 680 m a.s.l), southern Spain. Granada is a medium-size city with a population of about 232 208 inhabitants (www.ine.es; 2018). The climate is typically Mediterranean-continental, with cool winters, dry and hot summers and large diurnal temperature variability. Due to its location in a valley surrounded by mountains, Granada has a well-characterized mountain-valley wind regime with up-valley winds from NW and W during day, and down-valley winds from SE and E during night and early morning (Ortiz-Amezcua, 2019). Due to its geographical position, Granada is influenced by anthropogenic pollution from Europe and natural dust from North Africa (Lyamani et al., 2006, 2010; Mandija et al., 2016, 2017; Valenzuela et al., 2012a, b). There are no large industries in the metropolitan area and the main local aerosol source is road traffic (Lyamani et al., 2011; Titos et al., 2015). The meteorological conditions together with the orography of the city, located in a valley surrounded by mountains of high elevation from 1000 to 3398 m a.s.l, favor episodes with high atmospheric stability and heavy pollution, especially in winter (Lyamani et al., 2012).

Aerosol flux measurements were made at a tower in the southern part of the city. The measurement tower is about 500 m away from the principal highway that surrounds the western sector of the city and about the same distance from Camino de Ronda; that is one of the main busy roads of the city (Fig. 1). The closest road with motor traffic is around 50 m away from the tower. The measurement site is surrounded by several surface types, including residential, commercial and road areas in the wind direction between 0° and 225°, and agricultural and suburbs areas as well as the principal highway of the city in the wind direction between 225° and 360° (Fig. 1). The surrounding area is flat within a radius of about 6 km and the surrounding buildings are quite uniform with mean height of 20 m. The flux footprint (source area) accounting for 90% of the measured aerosol flux extends from approximately 650 m–1500 m around the tower, covering the different land cover types in the area of study (Fig. 1). It is important to note that the contours shown in Fig. 1 represent the average relative contributions of different land areas over the entire measurement period and that the measured individual 30-min aerosol fluxes may originate from areas outside these contours. For the calculation of the flux footprint we used the footprint model proposed by Kljun et al. (2004) assuming displacement height and roughness length of 13 m and 2 m, respectively, for wind direction between 0° and 225° and of 3.33 m and 0.5 m, respectively for wind direction between 225° and 360°.

3. Instrumentation

Instruments for the measurement of turbulent fluxes were mounted on the top of a tower at a height of 50 m above ground level. The tower height is 2.5 times the average height of the surrounding buildings (approximately 20 m) and hence eddy covariance setup should be in the inertial sublayer. Therefore, the aerosol flux measurements presented here can be considered to be representative of the local scale (Grimmond and Oke, 1999). In order to minimize any possible disturbances from the tower itself and avoid flow distortion, the measurements were taken at 10 m above the upmost level of the tower. The measurement flux system consists of an ultrasonic anemometer (Model 81000, R. M. Young, USA) to measure the three wind components and sonic temperature, and an ultrafine condensation particle counter (UCPC, model 3776, TSI Incorporated, USA) for measurements of particle number concentrations in the diameter range 2.5 nm - 3 μm. Sonic anemometer was mounted at 50 m height and the sampling inlet for UCPC was situated 25 cm below the sonic anemometer center. The air was sampled through a 10 m main stainless steel tube (4 mm inner diameter) at a flow rate of 16 l/min. The side air flow to UCPC was drawn through a 1.0 m long tube (with 2 mm inner diameter) from the main tube at a flow rate of 1.5 l/min. Both anemometer and UCPC data were acquired at frequency of 10 Hz.

To obtain information on planetary boundary layer dynamic we used vertical wind speed profile measured by Doppler lidar system Stream Line at 100 m from the tower. This instrument is operating in continuous and automatic mode since May 2016 at the Andalusian Institute for Earth System Research (IISTA-CEAMA). The Doppler lidar emits pulsed infrared radiation at 1500 nm and measures the Doppler frequency shift in the backscattered radiation due to the aerosol particles movement with wind. The Doppler lidar data are acquired in Stare mode (only the

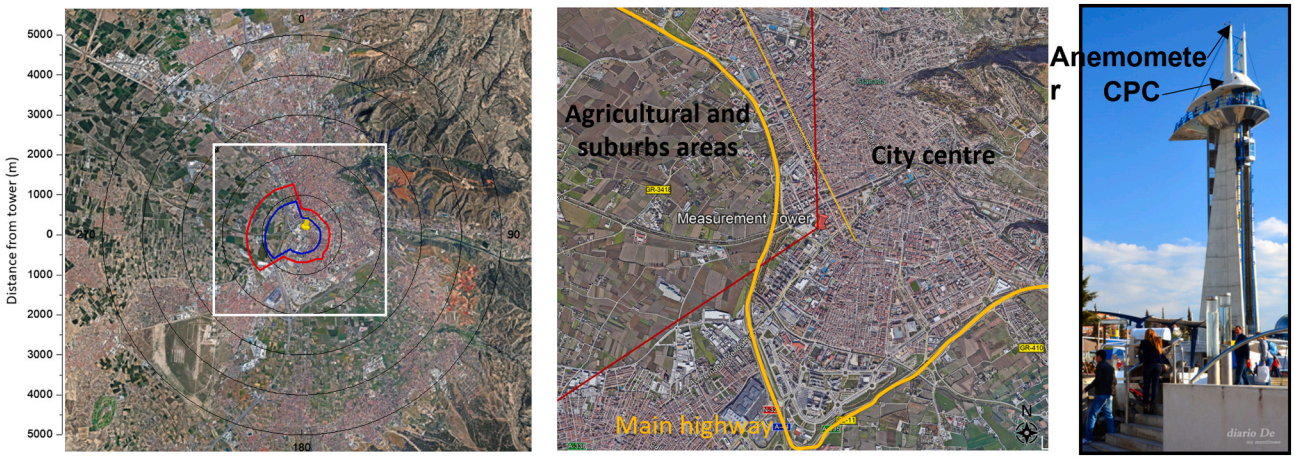

Fig. 1. (Left) Location of the tower in Granada city and its surroundings. Red (blue) contour show the average flux footprint (source area) accounting for 90% (70%) of the measured aerosol flux for the entire measurement period. (Center) Zoom on the white square area that shows the measurement tower location and its surroundings separated in two sectors: urban (0-225°) and rural (225-360°) sectors. The main highway surrounding the urban area and one of the main city streets are marked with yellow lines. The flux tower is marked with a pin. (Right) Measurement tower and location of the instrumentation. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

vertical wind speed is measured) with a time resolution of 2 s. The mixing layer height in stable and unstable cases is estimated from the 1-h variance of the vertical wind speed profile (σ_w^2) using variance threshold method (e.g., Moreira et al., 2018) with variance threshold value of 0.16 m^2/s^2. This threshold value was confirmed with Doppler lidar measurements and mathematical modeling by Large Eddy Simulations (Lenschow et al., 2012). More detailed information about the retrieval method of mixing layer height using vertical wind speed profiles is given by Moreira et al. (2018).

4. Aerosol number flux calculation and data processing

During the whole analyzed period we obtained 26 380 raw data. After excluding bad data due to the occasional instrument malfunction and short-term regular maintenance of the instruments from this raw data set we obtained 19 742 of 30-min aerosol data (76% of raw data). The 30-min particle number fluxes (N_F) were calculated from these raw data as the covariance of instantaneous fluctuations of the vertical wind component and particle number concentrations following Aubinet et al. (2000) guidelines. Data was processed with EddyPro 6.2.0 software (Li-Cor, Inc., USA). Before the calculation of particle number fluxes, vertical wind component and particle number concentrations raw data were, firstly, de-spiked following Vickers and Mahrt (1997) method to remove large spikes in vertical wind speed or aerosol concentration and, secondly, linearly de-trended to eliminate the presence of a possible trend in the 30-min time series. In addition, a two-dimensional coordinate rotation was applied to eliminate errors due to sensor tilt relative to the ground. Then, time lag between the concentration and wind measurements induced by the sampling lines was corrected by maximizing the covariance. The time lags between particle number concentration and vertical wind measurements ranges from 0.1 to 3.6 s with a mean value of 2.1 s. Finally, low frequency attenuation was corrected following Moncrieff et al. (2004) method and high frequency attenuation was corrected with Horst et al. (1997) method using UCPC time constant of $\tau = 0.8$ provided by the manufacturer. This resulted in an average (±standard deviation) frequency attenuation corrections of aerosol flux of 7 ± 5%. In order to ensure the quality of flux data we applied a stationarity test proposed by Mauder and Foken (2004). In this test the 30-min mean flux values should not deviate more than 60% from the average of fluxes from six sub periods of 5-min from the same 30-min period. Removing aerosol flux data by non-stationary test we obtained 16 393 high quality data, which represent 62% of the 26 380 original raw data. It is important to note that this data coverage is relatively high compared to other studies. Seasonal data coverage was also relatively high with data coverage higher than 60% in each season (Table 1). Data coverage during day and night time was also high representing 50% and 85% of the original data, respectively. For the following analysis, we only used these quality assured data.

5. Results

5.1. Meteorological and atmospheric conditions

Table 1 shows a statistical summary of meteorological variables and atmospheric conditions for each season at Granada during the analyzed period (November 2016 to April 2018). In this work, winter corresponds to the months from December to February, spring from March to May, summer from June to August and autumn from September to November. The study period was characterized by large seasonal temperature differences, with cool winters (−2 °C minima) and hot summers (41 °C

Table 1
Statistical summary of maximum mixing layer height, wind speed, temperature, friction velocity u* and stability parameter z_e/L (Obukhov length $L = -\frac{u^{*2}}{k(\frac{g}{T_0})T^*}$, where k is von Kármán constant, g/T_0 is a buoyancy parameter and T* is the friction temperature) registered at Granada during the analyzed period.

	Winter	Spring	Summer	Autumn
Maximum diurnal mixing layer height (m)				
Mean ± SD	1100 ± 400	1800 ± 600	1900 ± 500	1500 ± 600
Median	1100	1700	1800	1500
Minima/Maxima	500/2200	700/3500	1150/3600	680/2700
Wind speed (m s^{-1})				
Mean ± SD	2.0 ± 1.6	3.0 ± 2.0	3.0 ± 1.8	2.2 ± 1.5
Median	1.8	2.4	2.6	1.9
Minima/Maxima	0.03/10.00	0.10/14	0.10/10	0.06/9
Temperature (° C)				
Mean ± SD	10 ± 4	15 ± 6	29 ± 5	19 ± 7
Median	10	14	28	19
Minima/Maxima	−2/20	3/33	16/41	4/34
u* ($m\ s^{-1}$)				
Mean ± SD	0.7 ± 0.1	0.9 ± 0.2	1.0 ± 0.1	0.9 ± 0.2
Median	0.7	0.9	1.0	0.8
z_e/L (−)				
Mean ± SD	0.06 ± 0.09	0.01 ± 0.07	0.01 ± 0.05	0.04 ± 0.07
Median	0.06	0.01	0.01	0.03

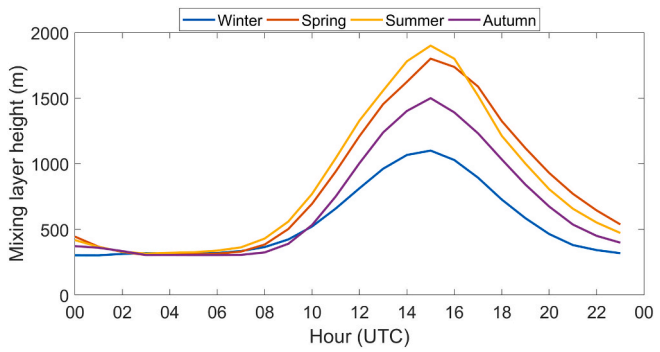

Fig. 2. Seasonal diurnal variations of mixing layer height over Granada averaged over all data available from November 2016 to April 2018.

maxima) with mean temperature values of 10 °C and 29 °C, respectively. Higher temperatures observed during summertime favours that the mixing layer reaches significantly higher altitudes (1900 ± 500 m) than in winter (1100 ± 400 m) with longer periods of mixing layer development (Fig. 2). In addition, the low mixing layer height observed in winter coincides with the predominance of low wind speed values (2.0 ± 1.6 m s^{-1}, compared to those observed during summer of 3.0 ± 1.8 m s^{-1}) and low friction velocity (u*) values (0.7 ± 0.1 m s^{-1}, compared to those observed during summer of 1.0 ± 0.1 m s^{-1}). Concerning the stability conditions, the atmosphere was in general unstable/near-neutral ($z_e/L < 0$, where z_e and L are the effective measurement height and the Obukhov length, respectively; Järvi et al., 2009) during warm period and it was near-neutral/stable ($z_e/L > 0$) during cold period, and it was predominantly stable ($z_e/L > 0.1$) and unstable ($z_e/L < 0.1$) during night- and day-time, respectively. In this sense, the combination of the reduced mixing volume and low wind speed and friction velocity in winter, favor lower vertical and horizontal dilution of pollutants in the atmospheric column.

5.2. Seasonal variation of aerosol number concentrations and fluxes

A statistical summary of aerosol number concentrations and fluxes measured at Granada urban site for each season and whole period between November 2016 and April 2018 is displayed in Table 2. For the analyzed period, the median aerosol number concentration was 7800 cm^{-3}, and the 25th and 75th percentiles were 5000 cm^{-3} and 12 200 cm^{-3}, respectively. The median value of aerosol number flux during the whole period was 150 × 10^6 m^{-2} s^{-1}, and the 1st and 3rd quartiles were 40 × 10^6 m^{-2} s^{-1} and 300 × 10^6 m^{-2} s^{-1}, respectively. Thus, as expected, Granada urban site acted as a net source of particles to the atmosphere during the analyzed period. Fig. 3 shows the monthly variation of aerosol concentrations and fluxes measured above Granada urban site from November 2016 to April 2018. The average monthly aerosol flux is always positive regardless of the month, indicating that the studied urban area is a net particle source also at a monthly scale.

Table 2
Statistical summary of 30 min aerosol number concentrations and fluxes measured at Granada from November 2016 to April 2018. SD is the standard deviation.

	Winter	Spring	Summer	Autumn	All period
Concentrations (10^3 cm^{-3})					
Mean ± SD	12.0 ± 8.0	8.3 ± 5.3	7.5 ± 4.0	9.9 ± 6.5	9.6 ± 6.6
Median	9.6	6.9	6.5	8.1	7.8
1st - 3rd quartiles	6.0–15.5	4.5–10.8	4.5–9.5	5.2–12.7	5.0–12.2
Number of data (data coverage)	5216 (60%)	4775 (63%)	2964 (67%)	3437 (60%)	16393 (62%)
Fluxes (10^6 m^{-2} s^{-1})					
Mean ± SD	230 ± 300	210 ± 240	140 ± 160	160 ± 210	190 ± 240
Median	170	180	115	120	150
1st – 3rd quartiles	45–360	60–325	30–225	20–250	40–300
Number of data (data coverage)	5216 (60%)	4775 (63%)	2964 (67%)	3437 (60%)	16393 (62%)

Despite Granada city acted as a net source of particles during the analyzed period, downward negative fluxes were also observed occasionally (12% of the analyzed data), showing that Granada city behaved during specific periods of time as a net sink of particles. These occasional downward fluxes were observed throughout the analyzed period and most of them were recorded during night-time and early morning. This is consistent with the results of Mårtensson et al. (2006) and Harrison et al. (2012). These studies reported that positive upward aerosol fluxes at Stockholm (Sweden) and London (UK) were the most frequent and that the observed occasional downward fluxes were very small in magnitude and only occur during low aerosol loading conditions. These authors attributed the urban negative fluxes to the stochastic nature of turbulence and uncertainty of the eddy covariance measurements. However, in our case, most of negative fluxes were relatively high in magnitude (median value of −53× 10^6 m^{-2} s^{-1}, with the 1st and 3rd quartiles of −130 × 10^6 m^{-2} s^{-1} and -20 × 10^6 m^{-2} s^{-1}, respectively) and were observed during high aerosol loading conditions (median aerosol number concentration was 9236 cm^{-3}, and the 1st and 3rd quartiles were 6000 cm^{-3} and 14 300 cm^{-3}, respectively). Thus, our results indicate that the downward aerosol fluxes observed over Granada were not caused by stochastic effects in the data. It is important to remember that the observed fluxes are net fluxes, i.e. a combination of both upward and downward transport of particles. Thus, downward particle fluxes can be attributed to updrafts of relatively particle depleted air from surface or to downdrafts of relatively particle enriched air from aloft (entrainment). However, the lack of additional information on aerosol characteristics during these events such as vertical aerosol distribution from ground surface to top of boundary layer and chemical aerosol composition as well as aerosol size distribution make difficult the accurate identification of the origin of these downward fluxes. Thus, no attempt is made here to investigate the origin of these downward fluxes. A detailed analysis of the drivers of the negative fluxes observed in this study will be investigated in the near future.

As can be seen (Table 2 and Fig. 3), both aerosol flux and concentration, showed large seasonal variation presenting their maxima in winter and minima in summer. The median aerosol flux value in winter (170 × 10^6 m^{-2} s^{-1}) was 50% larger than that measured in summer (115 × 10^6 m^{-2} s^{-1}). Meanwhile, the median aerosol number concentration also increased by 50% from 6500 cm^{-3} in summer to 9600 cm^{-3} in winter. Similar seasonal variations in aerosol number fluxes and concentrations have been observed in other urban areas, with highest values in winter and lowest ones in summer (e.g., Järvi et al., 2009; Ripamonti et al., 2013; Donateo et al., 2019). These seasonal variations of both aerosol number concentration and flux are driven by a combination of temporal variability in (1) emissions strength and sources, (2) meteorological conditions and (3) atmospheric dynamics. The summertime mixing layer in Granada reached significantly higher altitudes (1900 ± 500 m) and has longer period of development (Fig. 2) than in winter (1100 ± 400 m), and the summer wind speed values (3.0 ± 1.8 m s^{-1}) were also higher than those registered in winter (2.0 ± 1.6 m s^{-1}), Table 1. Thus, the reduced mixing volume and low wind speed in winter, combined with lower friction velocity (0.7 and 1.0 m s^{-1} in winter and summer, respectively), favor lower vertical and horizontal dilution of the actual aerosol emissions in the atmospheric column and contribute to the increase observed in aerosol concentration in winter months. Additional aerosol sources activated during winter such as domestic heating and burning of residual agricultural waste contribute to the higher concentrations and fluxes observed. A detailed investigation of the potential sources of aerosol in our study area and the possible role of

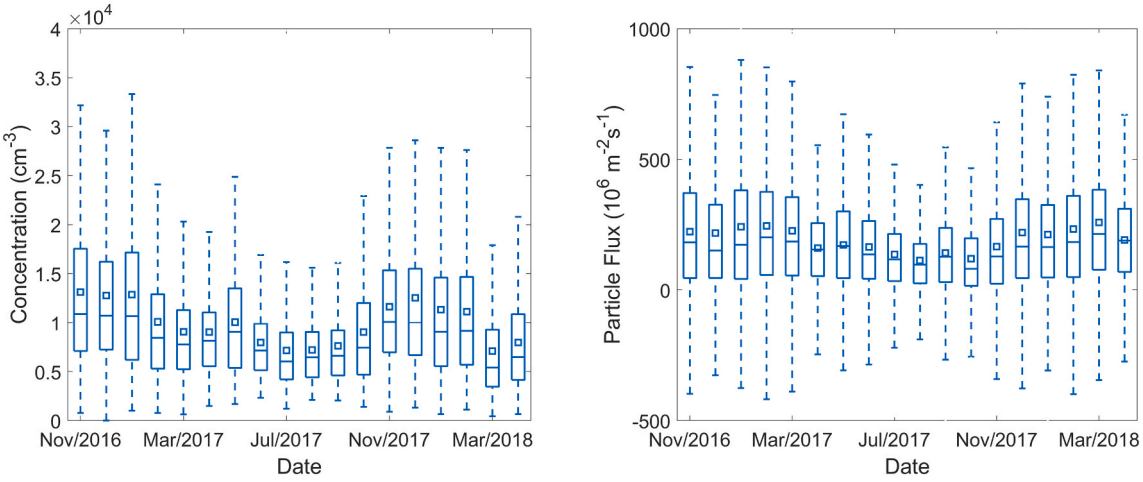

Fig. 3. Monthly aerosol concentration (left) and aerosol flux (right) observed over Granada from November 2016 to April 2018. The center rectangle and line show the mean and median, the lower and upper edges of the box represent the 25th and 75th percentiles of the data, respectively. The length of the whiskers represents the 1.5 × interquartile range, which includes 99.3% of the data.

domestic heating and burning of residual agricultural waste in winter season will be done in the following sections.

It is worth to note that, although aerosol number fluxes and concentrations show almost similar seasonal variation, both metrics show some differences in their behaviors. The median aerosol concentration value in autumn (8100 cm^{-3}) was 15% larger than that observed in spring (6900 cm^{-3}), Table 2. However, the median aerosol flux value in autumn (120 m^{-2} s^{-1}) was 33% lower than that observed in spring (180 m^{-2} s^{-1}), which may indicate that surface emissions are not the main cause of the high aerosol concentration in autumn as compared with spring. Aerosol fluxes are mainly controlled by surface emissions and turbulence strength whereas atmospheric aerosol concentrations do not only depend on the actual surface emissions, but also on atmospheric boundary layer dynamics and meteorological conditions. The boundary layer mixing height in autumn (1500 ± 600 m a.g.l.) was lower than in spring (1800 ± 600 m a.g.l.) and has shorter period of development (Fig. 2), indicating a less volume and time for aerosol vertical dilution in autumn in comparison to spring. Also, wind speeds in autumn (2.2 ± 1.5 m s^{-1}) were lower than in spring (3.0 ± 2.0 m s^{-1}). Thus, since there was no increase in aerosol fluxes from spring to autumn (but there was a decrease), the less vertical and horizontal aerosol dilution effects in autumn as compared to spring can at least partially explain the high aerosol concentrations observed in autumn.

5.3. Aerosol number concentrations and fluxes dependence on wind sector

5.3.1. Wind sector analysis

Atmospheric aerosol number concentrations are largely dependent on emission source/sink spatial distributions and their strengths, synoptic and mesoscale meteorology, boundary layer dynamics and chemical processes, while aerosol number fluxes are mainly driven by local emissions and turbulence strength. On an inhomogeneous surface with different land uses and cover types, such as a city, both aerosol number concentration and flux measurements depend strongly on wind direction due to the non-uniform spatial distribution of surface aerosol sources and sinks. Studies conducted in different urban areas (e.g., Järvi et al., 2009; Martin et al., 2009; Ripamonti et al., 2013; Donateo et al., 2019) revealed that aerosol number concentration and flux measurements are largely controlled by the surface sources/sinks within the footprint area (i.e. the source surface area affecting the site measurements). However, different theoretical studies have shown that the concentration footprints are generally larger than the flux footprints (e.g., Rannik et al., 2012). Therefore, both metrics can be influenced by different aerosol sources and hence can have different behaviors because distant aerosol sources located outside the flux footprint can affect aerosol concentration measurements but not flux levels. Thus, the combined analysis of both aerosol number concentrations and fluxes by wind sector will help us to identify the potential sources and sinks of aerosols affecting our site and provide us insight on the contributions of local and/or distant sources/sinks to both metrics. In order to assess the spatial patterns of emissions and identify the potential sources of aerosols in our study area, we analyzed the dependence of both aerosol number concentrations and fluxes on wind direction. Fig. 4 shows aerosol number concentrations and fluxes as a function of wind direction in bins of 45° for the period between November 2016 and April 2018. The corresponding frequency of occurrence of wind direction in each 45° wind sector is included in Fig. 4.

The predominant direction of the wind (30% of the cases) was the west-northwest direction (270°-315°), while the less frequent (5%) corresponds to the wind sectors 0°-45° (north-northeast) and 135°-180° (south-southeast). For the other wind sectors the frequency of occurrence was almost similar with values in the range 10–14% (Fig. 4). Because of the low number of cases in the wind sectors 0°-45° and 135°-180° (direction from urban area; Fig. 1) the results from these two wind directions will be treated with caution. As can be seen in Fig. 4, the mean and median values of aerosol fluxes are positive in all wind directions, indicating that aerosol emission dominated over the deposition and on average all land type sectors affecting the measurement site acted as sources. Unexpectedly, aerosol flux values were highest with median value of 220 × 10^6 m^{-2} s^{-1} from the SW-W-NW (225-315°) sector including the agricultural area and the principal highway of the city and lowest with median value of 51 × 10^6 m^{-2} s^{-1} from NE-E-SE (45-135°) sector which include the city center, densely built up residential and commercial areas as well as the main traffic roads of the city (Fig. 4). Not only the median value of the aerosol fluxes in the SW-W-NW sector was higher than that obtained in the NE-E-SE sector, but also the mean, third and first quartile values (Fig. 4). The Mann-Whitney test with a significance level of 0.01 confirms that the aerosol fluxes from SW-W-NW sector were statistically significantly higher than those from NE-E-SE sector. Similarly, aerosol number concentrations from SW-W-NW directions were also statistically significantly higher at 0.01 level of confidence than those from NE-E-SE sectors, as confirmed by Mann-Whitney test. Thus, the results suggest the presence of strong local emission sources in SW-W-NW wind sector as compared to the urban sector in NE-E-SE wind directions.

As we commented before, Granada has a well-characterized mountain-valley wind regime with up-valley winds from NW and W during day, and down-valley winds from SE and E during night and early

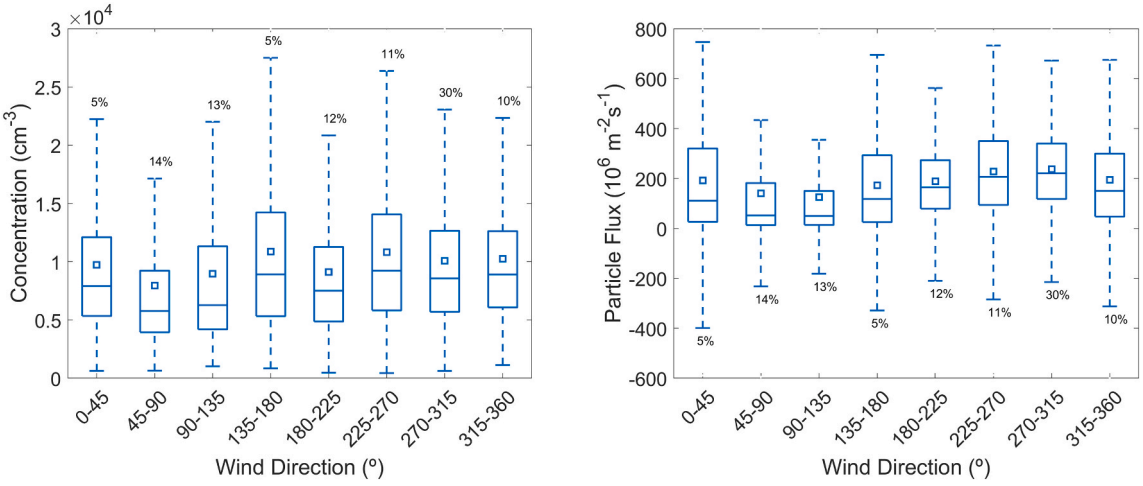

Fig. 4. Aerosol number concentrations (left) and fluxes (right) according to 45° wind sectors for the period between November 2016 and April 2018. The corresponding wind frequencies of occurrence in each 45° wind sector are inserted in the figures.

morning (Ortiz-Amezcua, 2019). In fact, in depth analysis of the data showed that wind from rural sector (225°-360°) occurred more frequently (more than 70%) during day-time, when local anthropogenic emissions are activated. In contrast, the majority of wind (more than 70%) from the urban sector (0° to 225°) occurred during night time, when local anthropogenic activities are reduced, which may explain the low aerosol number concentrations and fluxes recorded for this urban sector in comparison to those observed from the rural area sector. Therefore, to avoid misleading interpretation of the results and obtain reliable information about the emission/sink sources in the studied area, we repeated the same analysis but separating the data for day and night periods. In addition, for a robust statistical analysis of the data, we regrouped the data in two groups: urban sector (corresponding to 0-225° wind sector) and rural sector (corresponding to 225-360° wind sector). The data were separated into these two sectors due to their completely different land-use and because aerosol source distributions and land use types within each sector are very similar. Urban sector (0°-225°) contains the city center, densely built up residential and commercial areas and traffic roads, including one of city streets with more dense traffic of the city that is located just at 500 m from the measurement site (Fig. 1). In contrast, rural sector (225°-360°) contains extensive agricultural areas, poorly built up area and the main highway of the city that crosses this sector from south to north at a distance of 500 m from the measurement site.

The results of this analysis are given in Table 3, showing that both aerosol number concentrations and fluxes from rural sector were statistically significantly higher than those from urban sector (as confirmed by Mann-Whitney test at 0.01 significance level) during both day and night-time, evidencing again the presence of strong local emission sources in rural sector. This result points to the intense traffic on the highway that crosses the rural sector from south to north as a potential source of local emissions. However, the highest aerosol concentrations and fluxes in the rural sector may also be attributed to emissions from agricultural waste burning activities and biomass burning for domestic heating, especially in cold season, when previous studies revealed that the burning of agricultural residues represent an additional important local source of aerosol particles in our study area (e.g., Titos et al., 2017; Patrón et al., 2017).

5.3.2. *Seasonal and sectorial analysis: impact of traffic and biomass burning emissions*

In order to assess and distinguish the possible effects of traffic emissions from the highway and agricultural waste burning emissions, both coming from the rural sector, on aerosol concentrations and fluxes at the urban site, data were further divided into winter (when open waste burning is allowed) and summer (when this practice is forbidden) periods. The results are shown in Table 4.

Table 3
Statistical summary of aerosol concentration and fluxes measured at Granada from November 2016 to April 2018 for urban and rural sectors separated into day and night time periods. SD is the standard deviation.

	Mean ± SD	Median (1st - 3rd quartiles)	Number of data
Concentration (10^3 cm^{-3})			
Urban sector - Day time	9.1 ± 6.7	7.1 (4.8–10.9)	2392
Rural sector - Day time	10.5 ± 6.6	8.9 (5.8–13.3)	5883
Urban sector - Night time	8.9 ± 6.9	6.7 (4.2–11.2)	5518
Rural sector - Night time	9.8 ± 5.8	8.5 (5.7–12.2)	2600
Flux (10^6 m^{-2} s^{-1})			
Urban sector - Day time	250 ± 240	210 (110–350)	2392
Rural sector - Day time	270 ± 210	240 (140–370)	5883
Urban sector - Night time	120 ± 250	50 (10–150)	5518
Rural sector - Night time	140 ± 240	110 (30–230)	2600

In summer season, aerosol number concentrations from rural sector were significantly larger than those from urban sector, especially during night-time periods (Table 4). The mean aerosol number concentrations from rural sector at day and night periods were 30% and 50%, respectively, higher than those observed from urban sector. Aerosol fluxes were also higher for the rural sector at both day and night-time (Table 4), indicating high local aerosol emissions from rural sector in summer. The day-time mean aerosol flux from rural sector was 45% higher than that from urban sector whereas at night-time it was more than twice that from urban sector.

In winter season, however, no statistically significant differences were observed between aerosol number concentrations from rural and urban sectors during both day and night-time (Table 4). In the same way, the differences between aerosol fluxes from rural and urban sectors were small especially in night time, and were significantly lower than the differences between these sectors in summer season, indicating almost similar contributions of aerosol emissions from both sectors to the aerosol population over study area in winter season. Thus, the large differences observed for aerosol number concentration and fluxes between both sectors in summer, together with the small differences in

Table 4
Statistical summary of aerosol number concentration and fluxes measured at Granada from November 2016 to April 2018 from urban and rural sectors in winter and summer and separated into day and night time periods.

	Winter				Summer			
	Urban sector		Rural sector		Urban sector		Rural sector	
	Day	Night	Day	Night	Day	Night	Day	Night
Concentration (10^3 cm^{-3})								
Mean	12.0	11.5	12.5	11.6	6.6	5.4	8.7	8.2
SD	9.0	8.6	7.7	6.5	3.4	2.7	4.5	3.6
Median	9.4	9.0	10.1	10.5	5.9	4.7	7.8	7.6
1st quartile	5.5	5.4	6.8	7.0	4.4	3.6	5.2	5.7
3rd quartile	16.0	14.9	16.0	14.7	7.8	6.5	11.2	9.5
Flux (10^6 m^{-2} s^{-1})								
Mean	340	175	290	180	150	35	220	80
SD	300	300	200	260	130	100	170	160
Median	300	75	270	135	135	25	200	70
1st quartile	160	20	140	30	75	5	120	7
3rd quartile	480	240	400	300	210	65	300	145
Number of data	651	2182	1595	788	517	686	1288	474

winter, indicate that traffic emission from the highway crossing the rural sector is an important source of aerosol in summer at our urban area study.

Another important observation is that the winter aerosol number concentrations and fluxes from rural sector as well as from urban area were significantly higher, both at day and night time, than those observed in these sectors in summer season, reflecting a pronounced increase in local aerosol emissions from both sectors in winter. In the urban sector, the day time aerosol fluxes increased by a factor of 2.2 from median value of 135×10^6 m^{-2} s^{-1} in summer to 300×10^6 m^{-2} s^{-1} in winter, whereas the night time aerosol fluxes increased by a factor of 3 from mean value of 25×10^6 m^{-2} s^{-1} in summer to 75×10^6 m^{-2} s^{-1} in winter. For the rural sector, the day time aerosol fluxes increased by a factor of 1.4 from median value of 200×10^6 m^{-2} s^{-1} in summer to 270×10^6 m^{-2} s^{-1} in winter, while the night time aerosol fluxes increased by a factor of 1.9 from median value of 70×10^6 m^{-2} s^{-1} in summer to 135×10^6 m^{-2} s^{-1} in winter. This finding demonstrates that, in addition to traffic activities, domestic heating and agricultural waste burning are two important emission sources that affect aerosol population and the air quality of the Granada urban area in winter. This information is crucial for the implementation of effective mitigation measures to improve air quality in the city of Granada.

In particular, the local authorities implemented in 2014 a series of control measures with the aim of reducing air pollution, especially with respect to aerosol particles and NO_2. Most of these control measures focused on traffic emissions in the city center but no measures were taken to reduce traffic emissions on the city's main highway. In fact, emission abatement measures led to a decrease in particles and black carbon concentrations in this restricted traffic area, but were not effective outside this area (Titos et al., 2015). Therefore, to improve air quality in Granada, emission reduction measures focusing on traffic emissions on the city's main highway should be implemented. In addition, stringent control measures on emissions from agricultural waste burning activities, and especially on domestic heating emissions, should be implemented to improve air quality and protect human health, particularly in winter season.

5.4. Diurnal and weekly variations in aerosol number concentrations and fluxes

Weekly variations of aerosol concentrations and fluxes can only be explained by local anthropogenic activities, especially traffic, which shows a considerable decrease on the weekend in comparison with working days. Thus, to investigate the role of local emission sources and their influence on aerosol population over our study area, the weekend data were compared to working days. For this, the data were grouped on working days from Monday to Friday and other groups representing Saturdays and Sundays, respectively. In addition, to gain more insight on the role of traffic source in aerosol concentration and flux, weekly variation of both metrics for urban and rural sectors was investigated. It is worth to note that, for this analysis, we only used data collected in daytime hours when traffic activity is high.

Fig. 5 shows weekly variations of particle number concentrations and fluxes in urban and rural sectors. As can be seen, aerosol concentration and particularly aerosol fluxes showed a pronounced increase from weekends to working days in both sectors. In both sectors, aerosol fluxes increased on working days by more than 40% whereas aerosol concentration increased by approximately 33%, pointing out again that traffic was the main important source of aerosol in both sectors. Another interesting finding is that both aerosol concentrations and aerosol fluxes from urban sector were lower than those from rural sector during working days and weekends, as confirmed by Mann-Whitney test at 0.01 significance level. Furthermore, aerosol concentrations and fluxes in all seasons (not shown here) had pronounced reductions during weekends, especially on Sundays, evidencing that weekly cycles were primarily driven by traffic emissions in each season. All these facts, point out again to the strong impact of aerosol emissions from traffic circulating on the highway that crosses rural sector on aerosol population over our measurement site. Therefore, measures focusing on traffic emissions, especially at the main highway of the city, will be an efficient way to improve air quality over Granada.

The study of the diurnal variations can offer further insight into the underlying processes that control the evolution of aerosol concentration and flux in Granada and can help to identify local and regional sources. Fig. 6 shows the seasonal diurnal cycles of aerosol number concentrations and fluxes averaged over all data available from November 2016 to May 2018. As a common feature, the aerosol concentration presented a clear diurnal pattern, in all seasons, with two local maxima in coincidence with the morning and late afternoon traffic rush hours. Similar diurnal cycle was observed for black carbon at ground level, good tracer of traffic emissions, in the same urban area by Lyamani et al. (2011), which was attributed to the daily variation in both mixing layer dynamic and anthropogenic activities. In summer and spring, a third small peak was observed at around 14:00 h local time. This third peak derives from the enhanced new particle formation processes favored by the increased solar radiation intensity and the availability of precursor vapors during summer and spring seasons (del Aguila et al., 2018; Casquero-Vera et al., 2020, 2021). Ground surface measurements of aerosol size distribution obtained during the analyzed period near the site measurements (at approximately 100 m from the tower) corroborate this explanation (not

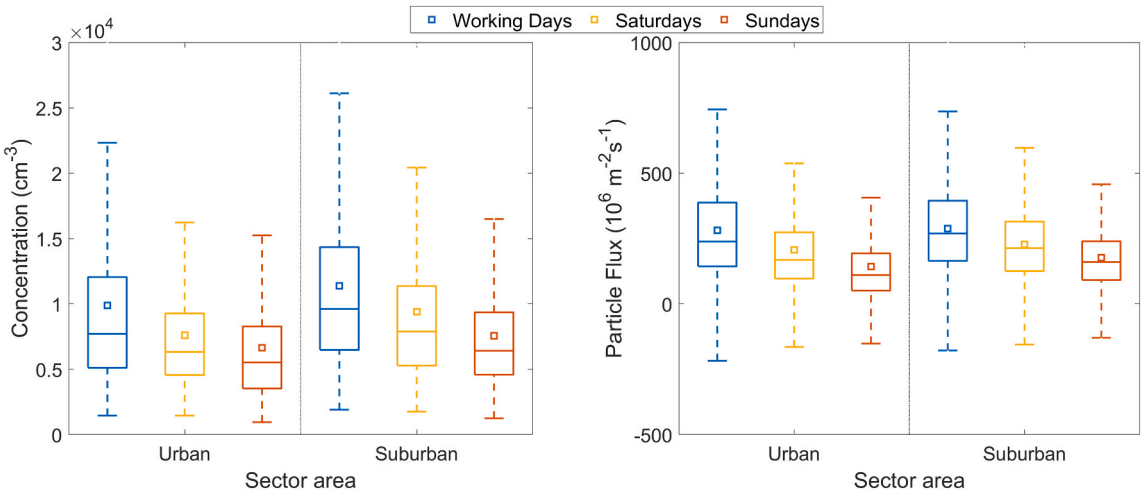

Fig. 5. Weekly variations of particle number concentrations (left) and fluxes (right) in urban and rural sectors averaged over all day-time data available from November 2016 to April 2018.

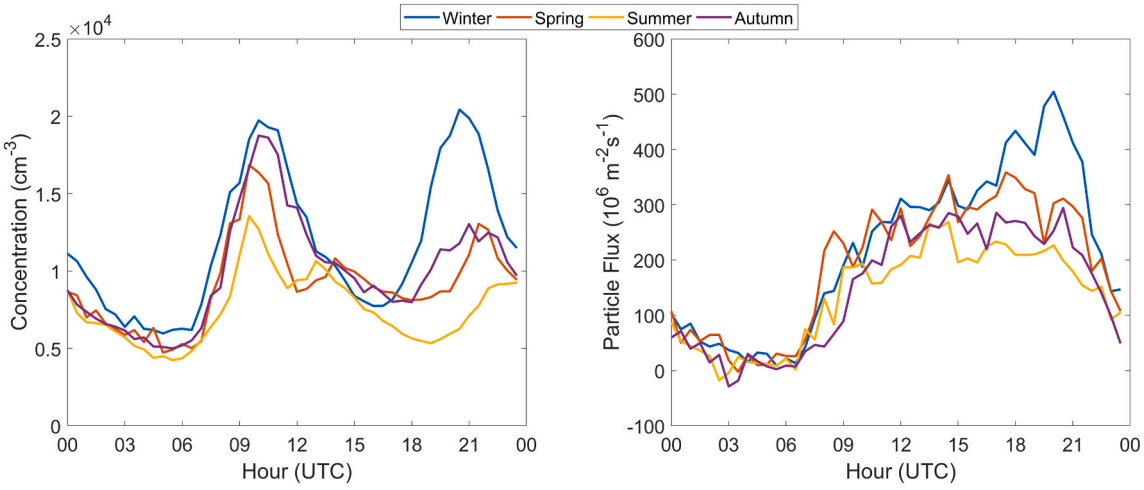

Fig. 6. Seasonal diurnal variations of aerosol number concentration (left) and aerosol flux (right) observed over Granada averaged over all data available from November 2016 to April 2018.

shown here). It is worth to note that the morning concentration peak in winter was almost similar to that observed for the late afternoon peak, whereas, in the other season the second afternoon peak was much lower than the first morning peak. This finding points to the significant impact of domestic heating emissions on aerosol concentration in winter season.

On the other hand, aerosol number flux showed a completely different diurnal behavior. In all seasons, the aerosol flux begins to increase early in the morning around 07:00 h and reaches high values at noon, remaining relatively constant throughout the day until it began to decrease after 19–20 h. However, in all seasons, the aerosol flux did not show a double peak pattern observed in the aerosol concentration. Also, the aerosol flux did not show the large decrease observed in the aerosol concentration around 12:00–14:00 h. The high aerosol fluxes associated with the low aerosol concentration in the middle of the day can be explained by the entrainment of particle depleted air from above the measurement site due to mixing layer growth (see Fig. 2). Similar diurnal aerosol concentration and flux behaviors were observed in other urban areas (e.g., Dorsey et al., 2002; Mårtensson et al., 2006; Järvi et al., 2009; Martin et al., 2009; Contini et al., 2012). It is important to note that aerosol flux in winter showed a large increase in the evening between 18:00–22:00 h which was associated with high aerosol concentration. This behaviour confirms the significant influence of domestic heating emissions on aerosol concentrations and fluxes in winter season.

6. Conclusions

In this study, continuous measurements from November 2016 to April 2018 obtained by eddy covariance method were used to examine the diurnal, weekly, seasonal, and spatial variability of aerosol number flux and concentration at an urban site in Granada, Spain. The majority of aerosol number flux values were positive, and negative fluxes were only observed in 12% of the cases, suggesting that Granada urban site acted as a net source of aerosol particles to the atmosphere. Most of negative fluxes were relatively high in magnitude and were observed during high aerosol load conditions. Aerosol number concentrations and fluxes measured in winter were higher than those observed in summer due to increased emissions from domestic heating and agricultural waste burning and less intense vertical mixing in winter.

The aerosol concentration presents a clear diurnal pattern in all seasons, with two local maxima coinciding with the morning and the evening traffic rush hours, evidencing the large impact of traffic

emissions throughout the year. Beside the two diurnal maxima, a third aerosol concentration peak was also observed around midday during summer and spring, which was associated to enhanced new particle formation processes in these seasons. In all seasons, aerosol flux did not show the two peaks associated with traffic observed in the diurnal cycle of aerosol concentration, which was explained by the entrainment of particle depleted air from above the measurement site due to mixing layer grows throughout the day. Strong weekly cycles were also observed with weekend aerosol number concentrations and fluxes significantly lower than working days, mainly due to decrease in traffic activities on weekends.

Aerosol fluxes were positive in all wind directions, indicating that on average all land type sectors affecting the measurement site acted as sources and that there was no specific sink area of aerosols around the measurement site. Large aerosol number concentrations and fluxes were observed from rural sector which contains an extensive agricultural area and the main highway of the city and low ones were observed for urban sector which includes the city center, densely built up residential and commercial areas and traffic roads. This finding points to the traffic emission from the highway that crosses the rural sector from south to north as a potential source of local emissions that affect our area of study. A detailed seasonal analysis by wind sector demonstrates that, in addition to traffic activities, domestic heating in urban sector and agricultural waste burning in rural sector are two important emission sources that affect aerosol population and the air quality of the Granada urban area in winter. This information is crucial for the implementation of effective mitigation measures to improve air quality in the city of Granada.

CRediT authorship contribution statement

J.A. Casquero-Vera: Conceptualization, Investigation, Methodology, Data curation, Formal analysis, Writing – original draft, Visualization. **H. Lyamani:** Conceptualization, Investigation, Methodology, Data curation, Formal analysis, Visualization, Supervision, Writing – original draft, Writing – review & editing. **G. Titos:** Investigation, Writing – review & editing. **G. de A. Moreira:** Writing – review & editing. **J.A. Benavent-Oltra:** Writing – review & editing. **M. Conte:** Writing – review & editing. **D. Contini:** Writing – review & editing. **L. Järvi:** Writing – review & editing. **F.J. Olmo-Reyes:** Supervision, Project administration, Writing – review & editing, Funding acquisition. **L. Alados-Arboledas:** Supervision, Project administration, Writing – review & editing, Funding acquisition.

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.

Acknowledgments

This work was supported by the Spanish Ministry of Economy and Competitiveness through projects PID2020-120015RB-100, CGL2016-81092-R, and CGL2017-90884-REDT, by the Andalusia Regional Government through project P18-RT-3820 and P20-00136, by the European Union's Horizon 2020 research and innovation program through project ACTRIS-2 (grant agreement No 654109). This research was partially supported by Project RTI2018.101154.A.I00 funded by MCIN/AEI/10.13039/501100011033/FEDER "Una manera de hacer Europa". The authors thank the Parque de la Ciencias for making this research possible. Juan Andrés Casquero-Vera is supported by BES-2017-080015 funded by MCIN/AEI/10.13039/501100011033 and FSE "El FSE invierte en tu futuro". Funding for open access charge: Universidad de Granada / CBUA.

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