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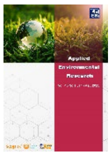

Research Article

Comparative Study of Aerosol Optical Properties along a Megacity

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Abstract

Interest in the study and characterization of aerosols has been growing given its multiple connections with air quality and climate. However, the limited ground-based stations for measurements in many countries are not enough to completely study a big city or a region. Low-cost technology arises as a viable alternative to improve the spatial resolution of measurements. This study aims to demonstrate that the properties of aerosols may change within a megacity and that is advisable to segment measurements according to their characteristics. We measured the optical properties of aerosols in the outskirts of Buenos Aires and compared the data obtained with AERONET data from the city center. This is the first study of its kind in the region. The comparison of concentration, sizes, and types of aerosols shows differences between the sites regardless of the background levels that they share regionally. While the city center has a strong influence from traffic and sea particles and levels of pollution typical of a dense city, the outskirts present several aerosol sources and characteristics of a semi-rural site with a moderate influence of anthropogenic sources.

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Introduction

Atmospheric aerosols are small particles (liquid or solid) that remain from a few hours to several weeks suspended in the air, depending on their size and the conditions of the atmosphere [1]. Its understanding is crucial given its direct (air quality) and indirect (climate) impacts on human health. Some particles can be transported through the upper respiratory tract into the bronchioles and the lungs producing direct health diseases, while the finer particles can penetrate deeper into the respiratory system and get deposited in the lungs increasing airflow resistance [2]. The International Agency for Research on Cancer (IARC) determined in 2013 that particulate matter (PM) is carcinogenic to humans [3], and the European Environmental Agency (EEA) attributed around 400,000 premature deaths in 41 European countries to PM in the air [4]. Aerosols also modify the transmission of both short-wave and long-wave radiation by scattering and absorption processes producing cooling and heating effects. This can alter the cloud microphysics, the hydrological cycle, and atmospheric stability to name a few well-documented consequences [5–6]. However, the effects of aerosols on climate are complex and there is great uncertainty about their behavior and its impacts, given their physical and chemical variability [7].

"Close-to-the-surface" aerosols are mainly characterized as PM mass concentrations using continuous/discontinuous monitoring systems around the world that provides information about local impacts. On the other hand, the aerosol atmospheric load is commonly represented as aerosol optical depth (AOD) measured with optical instrumentation (e.g solar photometers, lidars) deployed on the earth's surface and/or on satellite platforms. However, the necessary equipment for this kind of measurement under regulation is expensive resulting in

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sparse networks and wide regions without cover, especially in low and middle-income countries [8].

AOD is a dimensionless extinction coefficient related to the amount of direct sunlight that is prevented from reaching the ground at a determined wavelength by the presence of aerosols [9]. This extinction coefficient integrates the atmospheric column from the top of the atmosphere to the ground and provides information about the aerosol load present in the whole column, and hence many researchers have used it to characterize regional pollution [2, 5–6]. AOD changes in response to different processes, such as emissions of primary aerosols, transport, and removal from the atmosphere [10]. Likewise, AOD has seasonal and daily trends and changes in response to meteorology. AOD measured in different wavelengths is used to classify aerosols according to the particles' size and their light absorption characteristics. The classification gives us an idea about the main types of aerosols present in the atmosphere and we can trace their source and behavior over time but there are different approaches to do it. Chen et al. [11] compared different classification schemes using ground-based and satellite observations and found that every one of them was consistent with a reference method and each other. The main difference between the classification schemes used in literature seems to be the level of detail in the results.

Many studies have been devoted to classifying aerosols at a given location using a network of surface stations around the world called Aerosol Robotic Network (AERONET) [12] or satellite data. Recently, great importance has been given to inversion products of optical properties to extract information [8, 13–14]. Inversion products are aerosol characteristics such as size distribution, single scattering albedo, phase functions, and the complex index of refraction, and are derived from Sun photometer sky radiance measurements [12]. However, these kinds of products are not available in quantity and quality on many sites. Therefore, researches created other classification schemes using only direct solar radiation products. One of the most widely used is AOD vs Engstrom coefficient. Some studies that have applied this approach are, as example: Otero et al. [15], Otero et al. [16], and Casasola et al. [17] used data from AERONET to characterize aerosols over different locations in Argentina; Rezaei et al. (2019) discriminated aerosol types over Tehran city using MODIS data [18].

It is worth to mention that the scarce number and distribution of operational ground stations impose restrictions to studies using AERONET data. On the other side, studies using satellite data have considerable uncertainties associated with calibration issues [19]. Therefore, it is difficult to analyze the distribution of aerosol optical properties in a region or a large city. In general, results of a study for a specific site are taken as representative of a whole city. This responds to that a ground-based optical properties measurement integrates an atmospheric column, which turns to be similar in a wide area. However, a big city with non-homogeneous characteristics may not have a uniform aerosol distribution. Given this problem, low-cost manual solar photometers are a viable alternative to measure aerosol properties in places without official stations, allowing the possibility of collecting information on aerosols over large geographical areas [20]. These instruments use a light-emitting diode as a wavelength-selective photodetector. They can be easily calibrated with reference instruments and their cost is affordable for many low-income research institutions [20–22].

As a case study, we took the Buenos Aires megacity with about 15 million inhabitants in an area of around 13,200 km^2 [23]. The city center (1.5% of the megacity surface) houses more than 3 million inhabitants and has, as possible pollution sources, a petrochemical industrial facility nearby at the south of Rho de La Plata coastline, port activities, and a high vehicular flow. We selected the northwest area of the megacity as representative of its outskirts, specifically an intersection site and between three major cities in the area 30 km away from the city center (Los Polvorines, San Miguel, and Josй C. Paz, with almost 1 million inhabitants in total in an area equivalent to the city center of Buenos Aires). This area is surrounded by semi-rural zones and an industrial facility at the north and has a big green area in the south (Campo de Mayo military base covering almost 50 km^2). This work presents a comparative study of aerosol optical properties in different locations of the Buenos Aires megacity (city center and outskirts) using data from AERONET and a low-cost solar photometer. We focused on indicators for concentration, sizes, and types of aerosols to determine the effects of sources, activities, and population in the city center and outskirts. The main goal is to find differences that indicate that aerosol optical properties may change along the city.

Materials and methods

Between November 2018 and March 2022, we measured aerosol optical properties in the Buenos Aires megacity outskirts, specifically in the National University of General Sarmiento (UNGS from now on, 34.51888S 58.68986W). For comparison purposes, we also collected data of the same period from the only AERONET station in the whole megacity, the CEILAP BA site (34.5S 58.5W) representative of the city center (BA from now on). Figure 1 shows the location of the study sites.

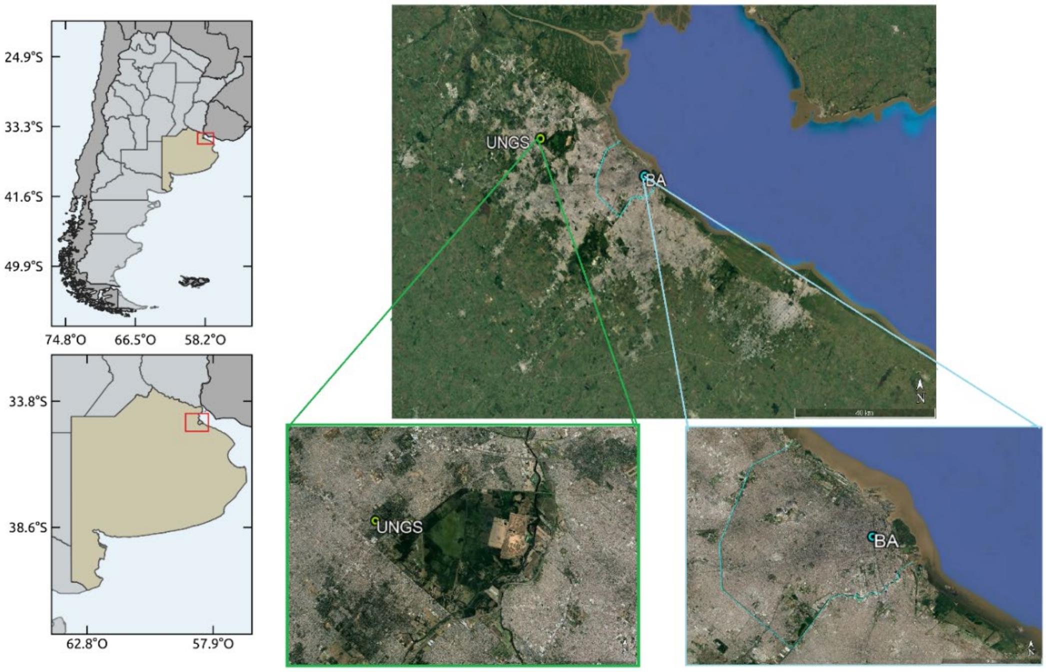

Figure 1 Location of Buenos Aires megacity, the City Center (BA), and the National University of General Sarmiento (UNGS).

The instrument used for the measurements is a low-cost solar photometer CALITOO from TENUM, calibrated with the AERONET network. It is a handheld device that uses a light-emitting diode that experiences electrical voltage at its junctions when receiving light within ranges of wavelengths similar to its emission spectrum. CALITOO photometers were developed within the framework of the GLOVE (Global Learning and Observations to Benefit the Environment) program which aims to study the impact of aerosols on the environment and were used in different studies around the world [24–25]. Every measurement delivers raw light extinction data, and AOD values at three wavelengths, plus date, time, inner temperature, inner atmospheric pressure, altitude, latitude, longitude, and solar elevation angle, all stored in a memory card. As the measurements had to be done manually and only when weather conditions were favorable, there is no fixed sampling frequency.

Each observation in UNGS was made under clear sky conditions and a solar angle higher than 30° to maximize the optical air mass (which limits measurement schedules according to the day of the year), in three different wavelengths: 465 nm (blue channel), 540 nm (green channel), and 619 nm (red channel). We used the blue channel for AOD characterization since it is the closest channel to the 440 nm commonly used [26].

To classify the aerosols, the Engstrom coefficient "α" [27] was computed from the slope of the AOD spectral dependence (between two wavelengths, λ_1 and λ_2) in a logarithm space (see Eq. 1) [28]. Here, the blue and green channels of the photometer were used for the calculations.

$$\alpha = \frac{ln\left(\frac{AOD_1}{AOD_2}\right)}{ln\left(\frac{\lambda_1}{\lambda_2}\right)} \quad \text{(Eq. 1)}$$

This parameter is inversely proportional to the aerosol mean square radius [27], varying between 4 (for small particles) and 0 (for large particles). The relationship between the Engstrom coefficient (from now on) and AOD provides information about aerosol characteristics [26, 28]. In this study, we used the classification scheme developed by Otero et al. [1] following reference literature, since it can be implemented with direct radiation measurements from the CALITOO photometer and has been used for several studies in Argentina [1, 15–17]. This methodology pro-vides some hints about the origin of aerosols present in the atmosphere and categorizes eight different classes: "Clean Continental" (CC), "Average Continental" (AC), "Polluted continental" (PC), "Urban/Industrial" (U/I), "Desert" (D), "Maritime" (M), "Biomass Burning" (BB), and "Antarctic" (A) (Figure 2). It should be considered that the limits for classes proposed by different authors are diffuse in different schemes and change with location, so that some cases near limits may correspond to another class than the one assigned or to none. This is a limitation of this kind of scheme; nevertheless, percentages give valuable information about aerosols in a certain area. For points falling in an overlapping area (more than one cluster), its classification was decided according to the shortest Euclidian distance to the centroids of the clusters.

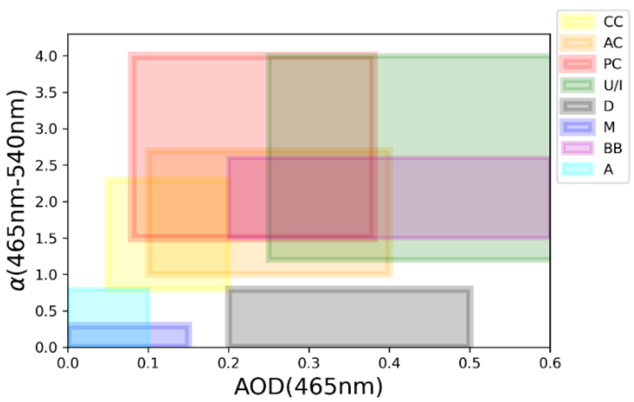

Figure 2 Classification scheme (AOD vs α). The wavelengths shown in the figure are the ones used for calculations with the CALITOO photometer data. The closest available wavelengths were used with AERONET data (AOD at 440nm, and α computed at 440-675nm).

To reduce the effect of outliers and uncertainties in calculations, we used daily means of AOD measurements from CALITOO and AERONET. This approach was used and recommended by other authors [5–7, 11, 14, 18, 24–25]. Days with less than three AOD measurements were discarded, giving a total of 227 days for the comparison.

We analyzed AOD, α, and classes of aerosol distributions, along with seasonal trends to find differences in the background particles between the two sites. In addition, we computed pollution roses (frequency of wind direction temporally correlated with the optical properties of aerosols) to find possible aerosol sources in both sites. We used wind data from the closest stations of the National Meteorological Service to UNGS (Campo de Mayo station) and BA (Aeroparque station).

Results and discussion

The distributions of aerosol optical properties between the two sites are very similar, as Figure 3 shows. This is expected since both sites share the same mesoscale processes. Nevertheless, the slight differences give us the most interesting results. BA site shows a slight shift towards higher AOD values (Figure 3 (a)) and lower values (Figure 3 (b)) than UNGS, meaning a higher concentration of aerosols and larger particles. On one hand, higher concentrations respond to a populated area with high intensity of different activities during the day. On the other hand, the aerosol mass distribution of an urban environment has two distinct modes, the sub-micrometer regime and the coarse-particle regime due to the re-suspension of dust particles from traffic [29]. The second regime seems to dominate the BA site. Otero et al. [1] and Otero et al. [15] found similar results for this site.

Regarding the UNGS site, AOD values are lower than BA as expected for a less populated area. In addition, particles are slightly smaller than in BA but still inside the coarse diameter mode. The values shown in Figure 3 for the UNGS site correspond to a semi-rural site with a moderate influence of anthropogenic sources [29], as was also described in Scagliotti et al. [30]. These results demonstrate that Buenos Aires megacity has different aerosol characteristics in different areas. Mesoscale processes dominate but optical properties measured by CEILAP BA AERONET station should not be taken as representative for the whole city.

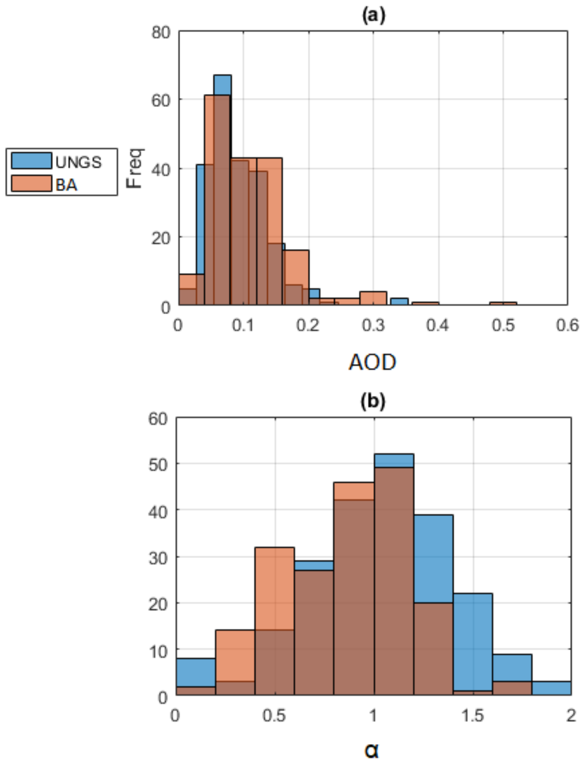

Figure 3 Comparative distribution of aerosol optical properties for Buenos Aires megacity at UNGS site and BA site.

Figure 4 shows a seasonal tendency of AOD in both sites supporting independent results from previous studies [1, 15, 30]. This annual trend can be explained by the solar radiation and the boundary layer height dynamic. During the cold months, the boundary layer has a lower mean height than during the warm months, meaning a shorter atmospheric column with aerosols trapped (and a lower AOD measured) [31]. There was no seasonal tendency found for α.

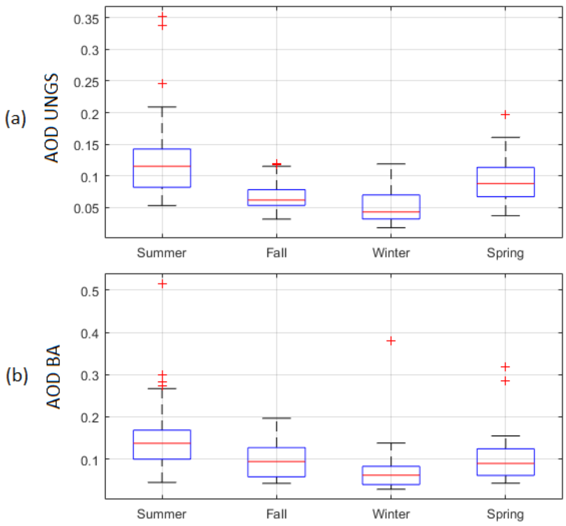

Figure 4 Boxplots with daily AOD values stacked by season for UNGS (a) and BA (b) sites.

Figure 5 shows the frequency of classes of aerosols for UNGS (a) and BA (b) sites, in addition with a bar stack plot (c) for comparison. Again, there is a very close similitude between both sites with the distribution of aerosol categorization present during measurements. These class distributions coincide with previous literature [1, 15, 30]. However, for BA site Otero et al. [1], and Otero et al. [15] discarded "A" and "M" types of aerosols.

There is a higher proportion of "AC" class in BA, and of "CC" in UNGS as expected for a city center compared with outskirts. The closer location of BA to the sea translate in more occurrence of "M" class of aerosols than in UNGS site. The higher proportion of "PC" aerosols in the outskirts than in city center is interesting, and could be explained by industrial activities and the presence of informal dumpsites. However, this finding need additional data to be fully studied.

The values of mean optical properties and class occurrence for aerosols according to wind direction give us information about possible sources in UNGS and BA. The lower values of AOD, coarser particles, and "CC" type of aerosols in UNGS seem to prevail with a wind direction coming from the biggest green area nearby at the southwest (Campo de Mayo military base). While the highest values of AOD, the finer particles, and the "AC/PC" aerosol types coincide with wind from the most populated city (San Miguel) and the closest industries at the west and northwest, as Figure 6 (a, c, e) shows. Regarding the BA site, Figure 6 (b, d, f) shows that there are no clear patterns with the wind direction, meaning the background activities dominate over the punctual aerosol emission.

Some limitations of this study are given by the low-cost solar photometer since it only allows direct solar radiation measurements (no inversion products). In addition, for the time being is not possible to assemble a robust model of aerosols in the whole megacity of Buenos Aires because of the low availability of air quality and meteorological data. More access to data from ground base stations of particulate matter and gases would be a very important addition to this kind of study and would enhance the results and conclusions.

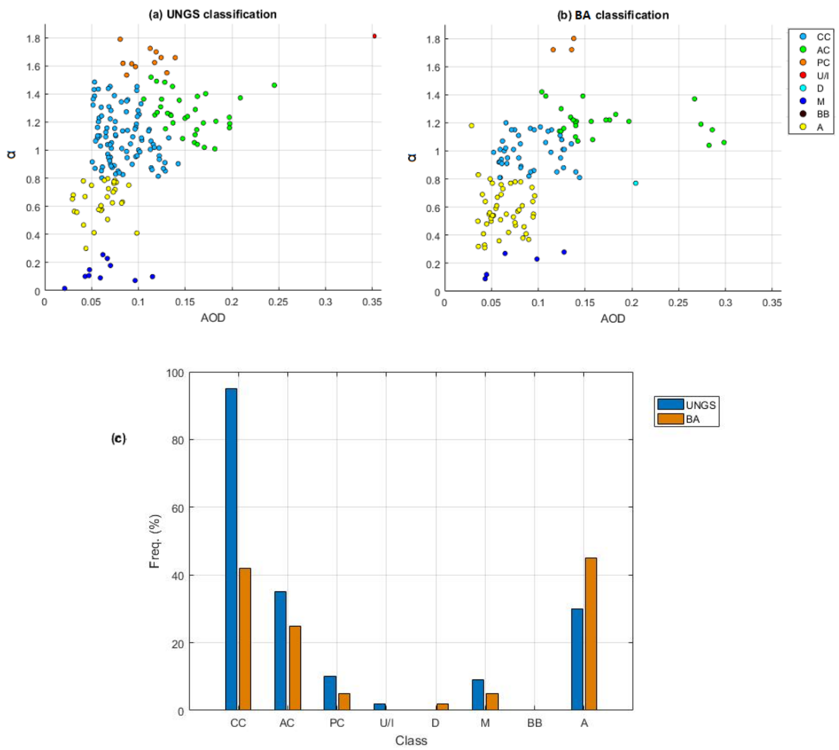

Figure 5 (a) UNGS classification scheme; (b) BA classification scheme; and (c) comparative classes' frequencies between UNGS and BA.

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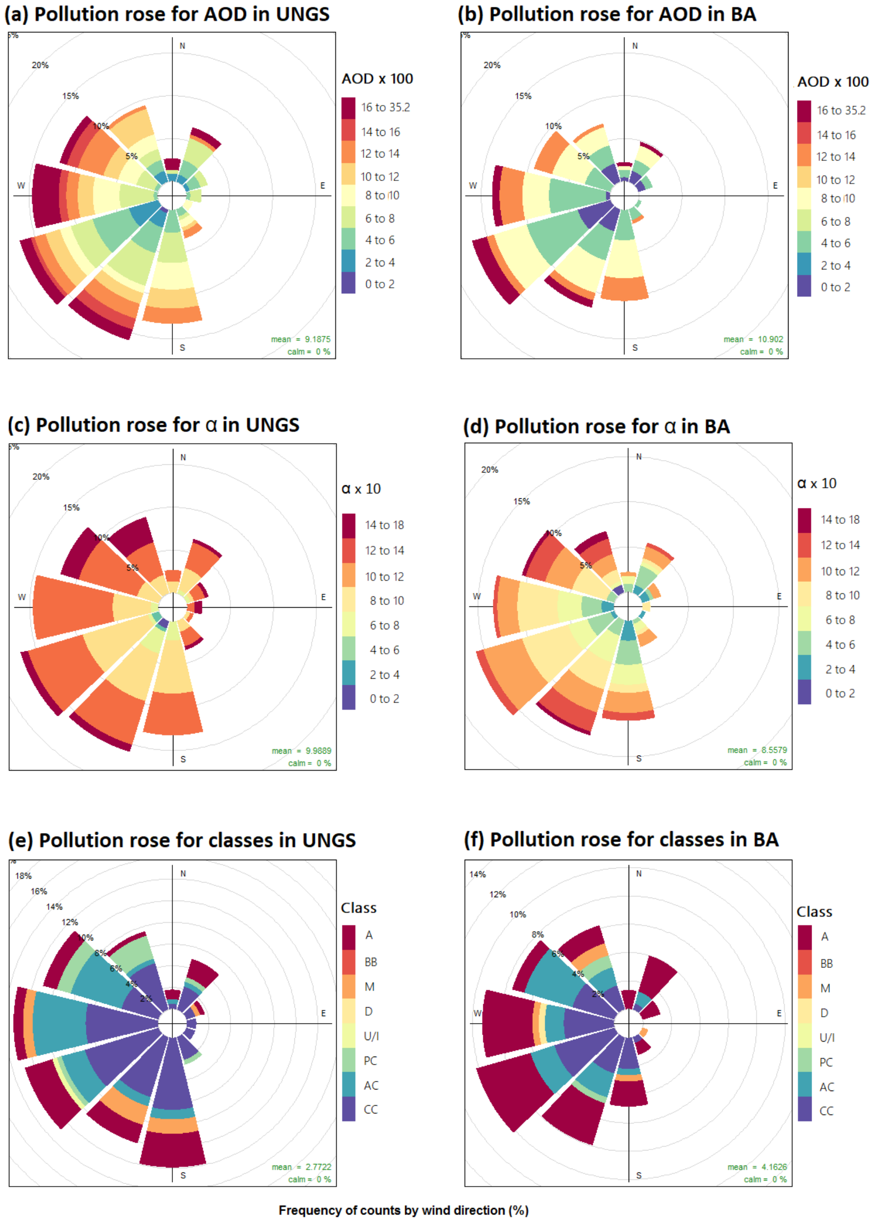

Figure 6 Pollution roses for optical properties and classes of aerosols in UNGS and BA sites.

Conclusions

This study compares optical properties of aerosols in two sites (outskirts and city center) within the megacity of Buenos Aires. The same regional processes, as can be seen in the similarities between the distributions and seasonal trends of AOD, α, and classes of aerosols affect the whole megacity. However, the results of a deeper analysis demonstrate that the two sites have different aerosol concentrations, sizes, and classes according to their characteristics (location, surface use, population, and activities). Moreover, pollution roses helped to detect different possible sources. On one hand, Campo de Mayo green area, San Miguel city, and industries facilities influence UNGS site aerosols. On the other hand, the high intensity of traffic and other activities in BA site make it impossible to detect punctual sources (except for the sea) with the available data since background concentrations are dominant.

This is the first study of its kind carried out in Buenos Aires, and the results help to increase the understanding of aerosol characteristics in the region. We especially highlight the necessity of a segmented study for aerosols in a wide region or a megacity. By design, cities are not uniform in characteristics that affect air quality. It would be of most interest to install several AERONET stations within a city considering aerosol "hot spots" and "sinkholes" on it. Additionally, measurements with low-cost solar photometers in different sites of a region can be a highly valuable source of information.

For future work, we will collect measurements from more sites within the city using the CALITOO photometer, satellite data, and ground-based stations of air quality and optical properties of aerosols. Moreover, meteorological data from reanalysis and air masses trajectories will be considered for a more detailed analysis in the region.

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