Journal of Aerosol Science 64 (2013) 11–23

Contents lists available at SciVerse ScienceDirect

Journal of Aerosol Science

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

Retrieving the relative contribution of aerosol types from single particle analysis and radiation measurements and calculations: A comparison of two independent approaches

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ARTICLE INFO

Article history:
Received 26 March 2013
Received in revised form
30 May 2013
Accepted 31 May 2013
Available online 18 June 2013

Keywords:
Aerosol types
Single particle analysis
SEM-EDX
SBDART model
UV-B irradiance
Aerosol optical properties

ABSTRACT

The main purpose of this work is to determine the relative contribution of different types of aerosols at an urban site by using two independent approaches: individual particle analysis, and radiative transfer calculations and irradiance measurements. To accomplish that purpose, we used our UV-B irradiance (280–315 nm) data set, the AERONET (AErosol RObotic NETwork) database, the SEM (Scanning Electron Microscopy, LEO 1450VP) analyses of the collected particles and the Santa Barbara DISORT Atmospheric Radiative Transfer (SBDART) model. On one hand, the collected particles were analyzed by SEM-EDX (Energy Dispersive X-Ray, Genesis 2000) in order to determine their chemical composition. Then, by using a developed algorithm they were classified as rural or urban, resulting in a (24 ± 3)% of rural and (76 ± 8)% of urban. On the other hand, aerosols were incorporated into the SBDART model through two of its default profiles (urban or rural) and by using the Aerosol Optical Depth (AOD) provided by AERONET. The aerosol effect on experimental surface UV-B irradiance was reproduced by a linear combination of the irradiances calculated by using these profiles. From this analysis we found that, in average, a mix of aerosols of (30 ± 3)% rural and (70 ± 7)% urban explains the observed reduction in the experimental irradiance. Considering the agreement between the results obtained by using these two independent methodologies, the use of the irradiance as a surrogate variable to retrieve aerosol types is discussed. The methodology presented here is applicable to any site provided irradiance measurements and AOD are available.

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1. Introduction

The importance of aerosols has been increasingly stressed in the recent decades. Many studies have looked at their influence on climate trough their contribution to radiative forcing. As aerosols scatter and absorb solar radiation, changes in their atmospheric concentrations and their chemical and physical properties can alter the transmission of the radiation through the atmosphere. That is why they are considered drivers of the climate change (Intergovernmental Panel on Climate Change (IPCC), 2007). Regardless of the large number of studies dealing with the radiative properties of aerosols, their net effect on global climate is still unknown and represents one of the major uncertainties in the understanding of Earth's climate system (Bond & Bergstrom, 2006). On other hand, aerosols with aerodynamic diameter less than 10 (PM10) and 2.5

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http://dx.doi.org/10.1016/j.jaerosci.2013.05.008

(PM2.5) μm are considered among the main pollutants of concern, especially in urbanized areas, where they have been directly related with the incidence and severity of respiratory diseases (Dockery, 2009).

Particulate matter comprises a huge number of species with heterogeneous morphology and chemical composition, which leads to different optical properties. In order to deal with this complexity different methods, techniques, and approaches are used.

To study the chemical composition of aerosol samples, two approaches can be considered: bulk analysis and individual particles analysis (e.g. Moroni et al., 2012; Rashki et al., 2013). While bulk analysis provides the average composition of the sample, the analysis of the individual particles, using techniques such as Transmission Electron Microscopy (TEM), Scanning Electron Microscopy (SEM) and Electron Probe X-Ray Microanalysis (EPMA) coupled with Energy Dispersive X-Ray Analysis (EDX), provides more detailed information (e.g. Laskin, 2010). These techniques have been used to analyze the composition of particles which are present in different environments (e.g. outdoor air in Buenos Aires City (Bogo et al., 2003), indoor air affected by tobacco smoke (Slezakova et al., 2011), and Mexico City subway system (Mugica-Álvarez et al., 2012)). Once the chemical composition has been obtained, different algorithms (e.g. Ro et al., 2001; Willis et al., 2002) and the previous knowledge of the site (López et al., 2011) can be used to classify the particles according to the abundance of the elements present in the samples.

To retrieve or infer the optical properties of aerosols different procedures, based on the interaction between particles and radiation, are extensively used (e.g. Giles et al., 2012). The two main approaches are experimental measurements and model calculations. Radiation measurements can be made at the surface (Srivastava et al., 2012) or from different platforms such as tethered balloons (de Roode et al., 2001), hang-gliders (Junkermann, 1994), stratospheric balloons (Madronich et al., 1985), fixed-wing aircraft (Palancar et al., 2011), and satellites (Kaskaoutis et al., 2011). Aerosol optical properties retrieved from satellites are usually validated against surface measurements (Estellés et al., 2012; Aaltonen et al., 2012). On the other hand, radiative transfer models allow including the aerosols properties, measured or supposed for a hypothetical situation, in order to study their effects on radiation. For example, Sinha et al. (2013) have used the Santa Barbara DISORT Atmospheric Radiative Transfer (SBDART) model to calculate the aerosol radiative forcing. Andrada et al. (2008) used the Tropospheric Ultraviolet and Visible (TUV) model to simulate the UV-B irradiance in the presence of aerosols and compare it with measurements at the surface.

Although there are several works that have studied the aerosols chemical composition and others that have analyzed their optical properties, it would be interesting to have a way to relate these two pieces of information. Some authors have already made one step towards this objective. For example, Andreae et al. (2008) related the results obtained for single scattering albedo with chemical composition, Che et al. (2009) studied the spatial and temporal characteristics of aerosol optical depth and Ångström wavelength exponent and their relationship with aerosol chemical composition and morphology, Dumka et al. (2011) studied the altitude profile of spectral aerosol optical depths, mass concentration of aerosol black carbon, and total concentration of composite aerosols measured at several altitude levels, and Titos et al. (2012) have investigated the optical properties, the mass concentration, and the chemical composition of the aerosols at an urban site in Spain.

In Córdoba there are only a few studies regarding to aerosol characterization. In one of these studies, in an air quality campaign carried out by the City government in the period 1995–2001, PM10 concentration was measured and it was the only pollutant that has exceeded several times the 24 h national standard of 150 $\mu g\, m^{-3}$ (Olcese and Toselli, 1997). Unfortunately, the chemical composition of these samples was not determined. This is aggravated by the fact that no additional air quality monitoring is currently underway by any governmental agency. Recently, López et al. (2011) have carried out 24 h samplings of PM10 and PM2.5 during the period July 2009–April 2010 at an urban and at a semi-urban site of Córdoba City. The bulk chemical composition of aerosol particles was determined by Synchrotron Radiation X-Ray Fluorescence. Multivariate analysis of the elemental data resolved a number of components (sources) which, based on their chemical tracers, were assigned physical meanings. According to this study, mobile sources exhaust and pollution caused by traffic (urban resuspended dust) were responsible for 30% and 55% of the sources apportionment to PM2.5, respectively. Considering the scarcity of data in Córdoba, the present work has been performed in order to advance toward the knowledge of the aerosol characteristics and their radiative effects in the region.

The main purpose of this study was to determine the relative contribution of different types of aerosols at an urban site by using two independent approaches: individual particle analysis, and radiative transfer calculations and irradiance measurements. On one hand, SEM-EDX techniques were used to determine the chemical composition of the aerosol particles. A developed algorithm was then used to classify these particles as urban or rural types based on the chemical composition. On the other hand, the SBDART model was used to simulate the UV-B irradiance for aerosol loaded days. Aerosols were incorporated into the model by using two of its default aerosol profiles, urban or rural, and the optical depth provided by AERONET. The calculated irradiance resulting from a linear combination of these profiles was compared against experimental UV-B measurements in order to determine the contribution of each aerosol type. At the end, considerations to further correlate these two independent results are discussed.

2. Materials and methods

2.1. Study area and sampling site

The study was performed in Córdoba, the second largest city in Argentina with approximately 1.3 million inhabitants. It is located at latitude 31° 24′S and longitude 64°11′W, about 470 m a.s.l. The climate is sub-humid with a mean annual

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