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Long-term comparative study of columnar and surface mass concentration aerosol properties in a background environment

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HIGHLIGHTS

- Long-term surface and columnar aerosol data in a background environment are analysed.
- The best correlation between AOD and PM10 is obtained using yearly means.
- High turbidity events show weak correlation of surface and aerosol properties.

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ABSTRACT

The relationship between columnar and surface aerosol properties is not a straightforward problem. The Aerosol Optical Depth (AOD), Ångström exponent (AE), and ground-level Particulate Matter (PM_X, x = 10 or 2.5 μm) data have been studied from a climatological point of view. Despite the different meanings of AOD and PMx both are key and complementary quantities that quantify aerosol load in the atmosphere and many studies intend to find specific relationships between them. Related parameters such as AE and PM ratio ($PR = PM_{2.5}/PM_{10}$), giving information about the predominant particle size, are included in this study on the relationships between columnar and surface aerosol parameters. This study is based on long measurement records (2003–2014) obtained at two nearby background sites from the AERONET and EMEP networks in the north-central area of Spain. The climatological annual cycle of PMx shows two maxima along the year (one in late-winter/early-spring and another in summer), but this cycle is not followed by the AOD which shows only a summer maximum and a nearly bell shape. However, the annual means of both data sets show strong correlation (R = 0.89) and similar decreasing trends of 40% (PM_{10}) and 38% (AOD) for the 12-year record. PM_{10} and AOD daily data are moderately correlated (R = 0.58), whereas correlation increases for monthly (R = 0.74) and yearly (R = 0.89) means. Scatter plots of AE vs. AOD and PR vs. PM_{10} have been used to characterize aerosols over the region. The PR vs. AE scatterplot of daily data shows no correlation due to the prevalence of intermediate-sized particles. As day-to-day correlation is low (especially for high turbidity events), a binned analysis was also carried out to establish consistent relationships between columnar and surface quantities, which is considered to be an appropriate approach for environmental and climate studies. In this way the link between surface concentrations and columnar remote sensing data is shown to provide useful information for aerosol characterization from a climatological context, despite some limitations.

1. Introduction

A common reference indicator for particulate air quality is the concentration of particulate matter (PM) at ground level, which is given in units of mass per unit volume of air (μg m^{-3}). The PM size fraction represented by PM_{10} and $PM_{2.5}$ are the most available and commonly used metrics. The PM_{10}, often called "inhalable particles" (EMEP, 1996; Brown et al., 2013), refers to particle fraction with aerodynamic diameters less than 10 μm. In the same way, $PM_{2.5}$ or "fine particles" (diameters below 2.5 μm) is another measure of particulate matter. The latter is associated to hazardous effects, having far greater efficiency than "coarse particles"

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(2.5–10 μm) to penetrate the respiratory system and reach the alveolar regions. Consequently, PM_{10} is usually used as a standard for measuring aerosol loading, while $PM_{2.5}$ is linked to health and visibility impacts (Pope III, 2000; Pope III and Dockeri, 2006).

In the last decades national and international institutions have set limits and guide values for the concentration of various PM size fractions with the aim to protect public health and environment (Delucchi et al., 2002; WHO, 2006; EC, 1999, 2008). Although so far this objective has not been universally achieved (Füssel and Jol, 2012), decreasing trends in yearly average have been observed in many European countries (EMEP, 2011,2014; Tørseth et al., 2012; Cusack et al., 2012; Boucher et al., 2013; Querol et al., 2014). These reductions are certainly attributed in a great part to the application of these abatement strategies of air pollution (EMEP, 2014). A significant effort has been dedicated to the implementation of continuous ground-based "in-situ" monitoring networks. The European Monitoring and Evaluation Programme (EMEP) established these networks with the goal of studying Long-Distance Atmospheric Pollution. This network provides to scientific community and governments quantitative information on the transport of air pollutants across national boundaries, associated deposition and concentration levels (Tørseth et al., 2012; EMEP, 2011, 2014). However the EMEP PMx observations are too sparse to resolve the large spatial and temporal aerosol variability and thus other measurement techniques, such as remote sensing at ground-based or satellite platform, may also be used.

Other networks for aerosol studies are based on powerful remote sensing techniques, like AERONET (Aerosol Robotic Network), which was created in the 1990's as a federation of national and regional networks managed by NASA. It is a dense network of ground-based sun photometers providing a continuous database of remotely sensed aerosol measurements at more than 400 sites around the globe (Holben et al., 1998). Such networks constitute a valuable source of information for the establishment of local and regional aerosol characterization and climatology (Holben et al., 2001; Dubovik et al., 2002; Toledano et al., 2007a; Bennouna et al., 2011, 2013; Mateos et al., 2015).

The primary aerosol parameter provided by remote sensing is the Aerosol Optical Depth (AOD), describing the extinction of the electromagnetic radiation in a given atmospheric column attributed to aerosols at a given wavelength. This is the key parameter for measuring the columnar aerosol load. The advantage of this methodology using radiation-particle interaction is the complementary information provided by AOD wavelength dependence, related to the size of particles. The Ångström exponent (AE) derived from AOD wavelength dependence is the parameter supporting this kind of information being the smallest this parameter the largest the particles. However, the AOD is a complex function of the aerosol mass concentration, mass extinction efficiency, relative humidity, and vertical distribution of aerosols, and hence several authors have investigated the relationships between AOD and columnar aerosol volume/mass concentration, surface PMx, mass deposition, or other quantities (Cachorro and Tanré, 1997; Kacenelenbogen et al., 2006; Pelletier et al., 2007; Kokhanovsky et al., 2009; Rohen et al., 2011; Toledano et al., 2012; among others).

The AOD, as **a** parameter representing the extinction over the whole atmospheric column, has a theoretical link with columnar particle volume concentration or columnar mass concentration through the definition of volume/mass efficiency factor (Cachorro and Tanré, 1997; Kokhanovsky et al., 2009; Toledano et al., 2012), but the link of these columnar properties with surface concentration given by PM_{10} (or $PM_{2.5}$) measurements is not a straightforward problem and hence empirical relationships are usually established (e.g., Estellés et al., 2012; references herein; Rohen et al., 2011).

In this context and restricting the study to AOD data given by ground-based observations we are interested in the relationships AOD-PM_{10} including derived quantities such as Ångström exponent (AE) and ratio of PMx fractions ($PM_{2.5}/PM_{10}$), related with particle size, which also need to be involved in the study of these relations. Thus, the objective of this work is to investigate in detail the relations between these four complementary parameters from a climatological point of view relying on 12 years of overlapping AOD and PM_X data (2003–2014) over two background stations of the large region of "Castilla y Leon" in the North-central Iberian Peninsula. This plateau presents a clean continental background aerosol without local pollution and it is adequate for this kind of study. The sites belong to EMEP and AERONET-Europe networks respectively, which certify the quality of the used data. To our knowledge this is the first time that this kind of study is carried out taking an area with these characteristics and lengthy records, emphasizing the climatological aspect.

It is relevant to note here that in the study area the highest levels of PMx are attributed to desert dust intrusions (Rodríguez et al., 2001; Escudero et al., 2005, 2007; Toledano et al., 2007a; Cachorro et al., 2008), because events of high AOD can also be due to external anthropogenic pollution (showing less influence on PMx values). Impact of desert dust aerosols on AOD (Toledano et al., 2007b; Cachorro et al., 2013) and PMx (Querol et al., 2009; Cachorro et al., 2014; Pey et al., 2013) are of particular interest for the Mediterranean Basin because they have a strong influence on the relationships established hereafter which opens new perspective on their potential use in aerosol studies.

The paper begins by introducing the region of study (section 1) and the description of the datasets (section 2). The results are presented in several sections. Section 3.1 gives a brief analysis of the annual cycle, interannual variability and temporal trends. In section 3.2, columnar scatter plots of AOD-AE and surface scatter plots PM_{10}-PR are examined in order to address general findings in terms of general aerosol characterization. Section 3.3 establishes and analyses the relationship PM_{10}-AOD and section 3.4 the PR-AE one. Section 3.5 gives the latter relationships under the analysis of binned data.

2. Measurement sites and data

The locations of the two sites used in this study are presented in

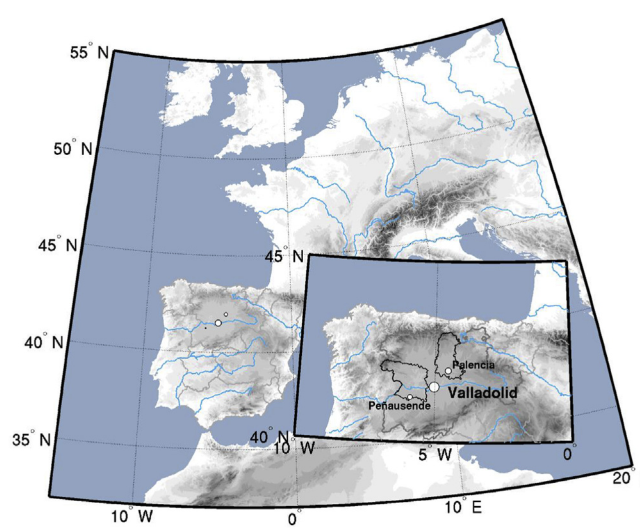

Fig. 1. Map of the area of study showing the location of the EMEP site of Peñausende and the AERONET site of Palencia within the region of "Castilla y Leon" in Spain.