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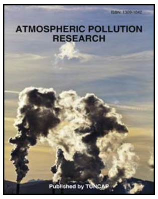

The relation between columnar and surface aerosol optical properties in a background environment

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ABSTRACT

This work presents the results of observation and the numerical simulation relationship between columnar and surface aerosol optical properties. The presented data include sun photometer nephelometer, aethalometer, and ceilometer observation, as well as the Navy Aerosol Analysis and Prediction System (NAAPS) re-analysis obtained between 2013 and 2016. Measurements were made in Strzyzow station (south-eastern part of Poland), which belongs to the AERONET and Poland-AOD network. Observation and simulation data show that the correlation coefficient between aerosol optical depth and surface aerosol scattering coefficient depends on the averaging period. For the monthly mean both parameters are negatively correlated as a result of the seasonal variability of anthropogenic emission in Central Europe and long-range transport of natural aerosol, as well as the change of the meteorological conditions. Reduction of the averaging time leads to an increase in the correlation coefficient, which is almost zero for a 10-day period and 0.4 ± 0.05 when the six-hour data are selected. In addition, the correlation between columnar and surface aerosol optical properties shows significant variation with surface temperature gradient. During convective conditions the correlation coefficient between aerosol optical depth and aerosol scattering coefficient is as much as 0.89 ± 0.03 while during inversion it is approximately 0.48 ± 0.08.

1. Introduction

Aerosol has an important influence on the earth-atmosphere system. It affects climate through three primary mechanisms. Firstly by direct radiative forcing due to its absorbing and scattering properties, and secondly by indirect radiative forcing, which means that it is modifying the cloud properties, e.g. affecting the albedo of clouds (Kaufman et al., 2006). Finally, aerosols have an indirect effect on the atmospheric chemistry by modifying the concentration of climate-influencing constituents (IPCC, 2013; Schwartz et al., 1995). However, large uncertainties in the radiative influences of atmospheric aerosols represent a major constraint on our ability to predict Earth's climate (IPCC, 2013). In addition, particulate matter (PM), or aerosol, is the fraction of air pollution that is most reliably associated with human disease (Anderson et al., 2012). Exposure to aerosol with diameters smaller than 2.5 μm (PM2.5) has serious adverse effects on human health (Krewski et al., 2000). In this case the most important is the concentration of PM10 (particles with diameters smaller than 10 μm) or PM2.5 at the surface, while for estimation of the direct radiation effect at least the columnar aerosol properties are required. Aerosol optical properties are measured from ground-based stations (Holben et al., 1998) and orbiting satellites (Kokhanovsky et al., 2010; Li et al., 2009). In an effort to study PM2.5 and PM10 characteristics with extensive spatial coverage, many techniques have been developed to derive PM concentrations from satellite optical property observations (Wang and Christopher, 2003; Gupta and Christopher, 2009). The widely used method of predicting PM concentration from satellite data is by empirical analysis, where in-situ PM measurements are linearly regressed with the corresponding satellite AOD. In order to reduce the uncertainty of the linear regression models, related parameters such as local meteorological and land use information were also used as an input into PM (Yap and Hashim, 2013). Recently, aerosol extinction profiles retrieved from the CALIOP spaceborn lidar onboard the Cloud-Aerosol Lidar and Infrared Pathfinder Satellite Observations (CALIPSO) satellite observations have also been used to improve the PM2.5–AOD relationship (Ford and Heald, 2013; Toth et al., 2013). Previous studies have reported different statistical significance of relations between AOD and PM10 or PM2.5 measurements (e.g. Hutchison, 2003; Engel-Cox et al., 2006; Al-Saadi et al., 2005; Koelemeijer et al., 2006; Kumar et al., 2007; Liu et al., 2007; Mukai et al., 2008; Yahi et al., 2013; Grgurić et al., 2014; Zeeshan and

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Oanh, 2014; Seo et al., 2015). Significant correlations are found between AOD and PM2.5 in the Eastern and Mid-west U.S. On the other hand, data from the Western U.S. stations show almost no correlation (Wang and Christopher, 2003; Hutchison, 2003; Engel-Cox et al., 2006). Filip and Stefan (2011) report quite small correlation between raw datasets of AOD and PM10 in two suburban sites near Bucharest, Romania and one urban site in Leicester, UK, during the summer–-autumn seasons. However, after filter procedure the linear correlation coefficient is nearly 0.7. Long-term measurements of PMx and AOD at AERONET stations in Spain indicate generally low correlations for daily data, they improve considerably for monthly or yearly means, and give very consistent relationships for binned data. In addition, these relationships depend on the aerosol characteristics of the site, and because of the clean and background conditions of our study area, they present a short range of AOD and PM10 values compared to other more polluted areas (Bennouna et al., 2016). In the northern part of Europe (Helsinki region, Finland) Natunen et al. (2010) reports the time-averaging increased the correlation coefficient as compared with one-hour PM2.5 measurements. Regarding the monthly averages of PM2.5 and AOD, the correlation coefficients were between 0.57 and 0.91. Over the extremely polluted Beijing region the correlation between the PM2.5 and PM10 concentration and MODIS AOD was significant (r^2 was between 0.61 and 0.81) and similar to the correlation between the PM2.5 and PM10 concentration and the ground-observed AOD (Kong et al., 2016). Therefore, MODIS AOD can be used to retrieve the distributions of PM2.5 and PM10 in the Beijing metropolitan region. Kong et al. (2016) reported that the absolute relative errors of the retrieved seasonal PM2.5 mean were lower than 8% and 33% in the urban/suburban region and the clean background, respectively. Song et al. (2009) reported that annual mean values of the PM10 concentrations and AOD show a strong spatial correlation, indicating the consistent presence of aerosol concentrations in China. However, the temporal correlation between the monthly means of the PM10 concentrations and AOD indicates a regional contrast in their seasonality. The correlation coefficients are 0.6 or higher in the southeastern coast region whereas they are -0.6 or lower in north-central China.

Aerosol layers in free troposphere, as well as different local meteorological conditions and variations in aerosol chemical composition play an important role in determining the strengths of PM/AOD relationship (Schaap et al., 2009). Koelemeijer et al. (2006) showed that the correlation between PM and AOD is improved when the AOD is divided by the mixing layer height but also when it is corrected for hygroscopic growth. Therefore, the relationship between AOD and PM should be determined regionally to account for its specific weather and aerosol conditions (Schaap et al., 2009). Observation at the surface of chemical properties as well as the single-scattering properties can help separate the radiative effects of different aerosol species (Aryal et al., 2014). However, vertical variability of chemical and physical properties complicates extrapolation of surface optical observation to columnar aerosol optical properties. Surface measurements by lidar or ceilometer devices can provide some information about vertical variability of the aerosol optical properties, however, due to the overlap problem this device does not measure aerosol optical properties within a few hundred metres which limits using of the standard algorithms (Aryal et al., 2014). Another possibility is to use the space lidar such as the CALIPSO but these kinds of observations are rare. Aryal et al. (2014) reported a correlation coefficient for AOD versus surface scattering coefficient (western North Atlantic Ocean at Bermuda) of about 0.5 (r^2) for all data with little variability among flow patterns ($r^2 = 0.45$–0.65). In addition, the correlation between surface scattering and lidar extinction is largest when the scattering Ångstrom exponent is high and flow is mostly over the land.

The main objective of this paper is to investigate the relation between columnar and surface aerosol optical properties for different time scales and weather conditions at a background station in Central Europe. These studies have examined extensive parameters, which depend on the quantity of aerosol, such as the scattering, extinction coefficient, and AOD, and intensive parameters, which depend on the characteristics of the aerosols, such as the scattering Ångström exponent defined below. Data analysis has been done based on remote sensing and in-situ aerosol equipment as well as model simulations (described in Section 2). Section 3 includes a discussion on the annul cycle of columnar and surface aerosol optical properties from the perspective of the thermodynamic stability of the lower atmosphere and vertical profiles of ceilometer signals. In the next section the variability of the correlation coefficient between both types of aerosol properties is discussed. In Section 5 analysis of the aerosol mixing height on relation between surface optical properties and AOD is provided. Section 6 includes analyses on impact air mass back trajectories on the correlation between observed AOD and aerosol scattering coefficient. Finally, the summary and conclusion are presented in Section 7.

2. Methodology

2.1. Research station SolarAOT

The research station SolarAOT is situated in Southeast Poland (49.88° N, 21.86° E). It lies 25 km to the Southwest of Rzeszow (population about 190,000) and 30 km to the North of Krosno (population about 45,000). The nearest town is called Strzyzow, and the station is situated 5 km from its centre. The population of this town is less than 9000 people, so it is quite a small locality. Measuring instruments are situated at the one of the highest points of Carpathian hill at 443 m a.s.l. Due to its location – far away from local sources of emission of anthropogenic aerosol – the station can be classified as a background station, so observations of aerosols can be representative of a large area and are related to long-distance carriage of aerosols from different geographical regions, e.g. Central Europe, the Middle East, or northern Africa. Measurements from the SolarAOT station include standard weather condition, aerosol optical properties, as well as the radiation budget. The SolarAOT station is part of the AERONET (Holben et al., 1998) and Poland-AOD (www.polandaod.pl) network (Markowicz et al., 2016).

2.2. Instruments and models

In this subsection one can find a short description of the measuring instruments and also the models used in this study. The in-situ observation of aerosol optical properties were done by an inlet mounted about 3 m above ground level.

2.2.1. Sun photometer CIMEL

The sun photometer CIMEL CE318 placed at the SolarAOT station is a multi-channel, automatic scanning radiometer that measures the direct solar irradiance and sky radiance at the Earth's surface. Measurements are taken at several different wavelengths: 340, 380, 440, 500, 675, 870, 936, 1020, and 1064 nm to determine atmospheric transmission and particle optical and microphysical properties. Additionally, the instrument has one filter wheel containing polarisers to measure linear polarisation and retrieved single-scattering optical properties. An automatic system of tracking the sun makes it possible to set the instrument at any angle horizontally and vertically and is responsible for pointing the instrument in the direction of the sun. It takes measurements only during daylight hours and also has a wet sensor to detect the precipitation and stop the measurement and protect the detectors from moisture (Holben et al., 1998). Data collected while making measurements are sent to the network and are then available on the AERONET website as a finished product at three different quality levels: 1.0 – with cloud occurrence, 1.5 – cloud screened data, and 2.0 – manually checked, quality assured data. The columnar aerosol optical properties, measured by CIMEL sunphotometer, used to this analysis are: aerosol optical depth (AOD), absorption aerosol optical depth