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Impact of the mixing boundary layer on the relationship between PM2.5 and aerosol optical thickness

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ABSTRACT

The purpose of this paper is to study the relationship between columnar aerosol optical thickness and ground-level aerosol mass. A set of Sun photometer, elastic backscattering lidar and TEOM measurements were acquired during April 2007 in Lille, France. The PM2.5 in the mixed boundary layer is estimated using the lidar signal, aerosol optical thickness, or columnar integrated Sun photometer size distribution and compared to the ground-level station measurements. The lidar signal recorded in the lowest level (240 m) is well correlated to the PM2.5 ($R^2 = 0.84$). We also show that the correlation between AOT-derived and measured PM2.5 is significantly improved when considering the mixed boundary layer height derived from the lidar. The use of the Sun photometer aerosol fine fraction volume does not improve the correlation.

1. Introduction

Most of the pollution aerosols emitted in the atmosphere are released in the atmospheric boundary layer and then become gradually dispersed and mixed through convection and turbulence. In addition to boundary layer features (e.g. depth, turbulent flux) that are key to understanding of the impact of aerosol on air quality, aerosol mass concentration measurements by air quality monitoring networks help to understand the dispersion of aerosols confined within the boundary layer. However, the aerosol vertical distribution and its temporal evolution are also of primary importance to understanding of changes in the aerosol mass concentrations at ground level, and to better characterize the distribution between local pollution events and large scale transport. In addition to ground-level observations, lidar vertical soundings provide a detailed description of scattering aerosols in the atmosphere. Primary parameters derived from elastic backscattering lidar profiles are the vertical distribution of aerosol backscattering and extinction coefficients. The vertical structure of the atmosphere can be inferred from a change in the backscattering vertical profile. Because the mixed layer has in general a higher aerosol backscattering coefficient than the free troposphere, the lidar can also detect the boundary between the two layers (Menut et al., 1999).

The relationship between aerosol mass and optical properties depends on the chemical composition, size and shape of the particles. Many studies (Chu et al., 2003; Gupta et al., 2006; Kacenelenbogen et al., 2006; Liu et al., 2004; Pelletier et al., 2007; Schaap et al., 2008; Wang and Christopher, 2003) have been devoted to finding the relationship between the columnar aerosol optical thickness (AOT) and the mass fraction PM2.5 or PM10. The PM data can be derived from AOT measurements using a simple linear model (Chu et al., 2003; Kacenelenbogen et al., 2006; Wang and Christopher, 2003). However, the relationship depends on the season and on the site location. There are auxiliary parameters such as meteorological variables or the characteristics of the mixing layer that need to be accounted for (Pelletier et al., 2007). Liu et al. (2004) and Van Donkelaar et al. (2006) improved the capability of the multiangles imaging spectroradiometer-derived AOT in estimating surface level PM2.5 by using aerosol vertical profiles simulated by a global atmospheric chemistry model. This result suggests that the use of vertical information, namely the altitude of the mixed layer or the aerosol extinction profile can improve the determination of PM from AOT measurements. Gupta et al. (2006)

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highlighted the impact of the mixing height on the relationship between AOT and PM2.5. From their data set over Texas, they found that the best correlation between PM2.5 and AOT is seen when the mixing height is between 100 and 200 m and when the relative humidity is less than 50%. However Schaap et al. (2008) did not find a significant improvement in the correlation between AOT and PM when including the lidar-derived mixing layer height in their study in the Netherlands. However they found that the PM2.5–AOT correlation increased when the comparison time slot was centred around and on noon, which suggests that the aerosols were well mixed in the boundary layer. The relative humidity also had an impact on the AOT via an increase in the size of the particles and a change in the refractive index (Hänel, 1976). Shinozuka et al. (2007) found that the fraction of ambient AOT due to water uptake was 37% ± 15% during their field campaign in North America. The change in aerosol scattering or extinction as a function of relative humidity can be parameterized (Kotchenruther and Hobbs, 1998; Raut and Chazette, 2007) but in most cases the relative humidity vertical profile and the aerosol hygroscopic properties remain unknown.

In this paper, we present observations performed at an urban site in the North of France. The experimental site is located on the outskirt of the city of Lille, France. Lille (50.61°N, 3.14°E) is a conurbation of 1.2 million inhabitants and in the vicinity of many urban and industrial aerosol pollution sources. We present the study of a pollution event that occurred during the month of April 2007. In March and April 2007, daily PM10 concentrations often exceeded 50 $\mu g\,m^{-3}$ corresponding to the European-24 h limit that must not to be exceeded on more than 35 days per year. The pollution events were also observed by Schaap et al. (2008) at Cabauw, The Netherlands. This period was chosen according to the availability of data for all of the instruments that were used in this study. We analyze the evolution of the aerosol mass at the ground in conjunction with lidar soundings and Sun photometer measurements. The objective is to analyze the built-up and removal of the aerosol load during the pollution event and to assess the variation in the relationship between aerosol mass at the ground and aerosol optical thickness.

2. Data and methods

2.1. Ground-level measurement of the particulate mass concentration

During the last decades a number of epidemiological studies have shown a link between pollution by airborne particulate matter (PM) and respiratory and cardiovascular diseases either for short-term or long-term exposure (Dockery et al., 1993; Künzly et al., 2000; Pope et al., 1995). The particle mass concentration measured at ground level is a way to evaluate the impact of aerosols on air quality. PM*X* means the mass concentration of particles with an aerodynamic diameter lower than *X*. In the present study we are using PM2.5 and PM10 data collected by a Tapering Element Oscillation Microbalance TEOM (Patashnick and Rupprecht, 1991) operated by the regional air quality network ATMO Nord-Pas de Calais. The measurement site is located downtown Lille (Faidherbe street) at less than 3 km from the lidar site at Université des Sciences et Technologies de Lille. Comparisons of TEOM to gravimetric measurements (Allen and Ress, 1997; Van Dingenen et al., 2004) show that routine TEOMs can underestimate PM10 by up to 35%. As this TEOM is not equipped with a Filter dynamics measurement system (FDMS), we have to apply a so-called correction factor on our PM10 and PM2.5 measurements. This factor is provided by air quality network ATMO Nord-Pas de Calais, and used for PM10. The factor is derived from a systematic comparison with data acquired by two other TEOM-FDMS located in the administrative area Nord-Pas de Calais (Calais and Tourcoing). During the experimental period, the PM2.5 was not measured with the TEOM-FDMS, so the correction factor for PM2.5 remains unknown. Since May 2008, the PM2.5 is also monitored by a TEOM-FDMS. We have compared the correction factor used for PM10 and for PM2.5 for the last ten days of May 2008, corresponding to a similar meteorological situation for our observation period. Both correction factors are well correlated ($R = 0.95$) and the PM2.5 correction factor can be derived from the PM10 one by using a linear relationship:

$$\mathrm{PM2.5_{corrected}} = \mathrm{PM2.5} \times \left(\frac{\mathrm{PM10_{corrected}}}{\mathrm{PM10}} - 0.1\right) \times 1.25 \qquad (1)$$

In this regression, we have only considered PM10 higher than 10 $\mu g\,m^{-3}$.

2.2. Columnar integrated aerosol optical properties using Sun photometer

We have used the data collected by a sky-scanning ground-based automated Sun photometer (referred in the AERONET data base as Lille) belonging to the Aerosol Robotic Network (Holben et al., 1998). A full description of the instrument and the retrieval procedure can be found in Holben et al. (1998) and Dubovik et al. (2000). The primary parameter that can be derived from the Sun photometer is the aerosol optical thickness (AOT) at four wavelengths (440, 670, 870, 1020 nm) and with an absolute uncertainty of ~0.010 to 0.021 (Holben et al., 2001). To be coherent with the lidar wavelength, we interpolate the AOT at 532 nm according to the Angström law and using the channels at 440 and 670 nm.

The columnar integrated volume size distribution $dV/d\ln r$ (in $\mu m^3/\mu m^2$) in range of radii between 0.05 and 15 μm is also derived from sky brightness measurements (Dubovik and King, 2000). The retrieval of particle volume size distribution was demonstrated to be adequate in practically all situations (Dubovik et al., 2002). The error in the retrieved volume density changes as a non-linear function of particle size, aerosol type and actual values of size distribution. In particular, for the intermediate size particle size range 0.1 and 7.0 μm, the retrieval errors do not exceed 10% in the maximum of size distribution and may increase up to 35% for the points corresponding to the minimum values of size in this size range. The retrieved size distribution volume is not independent in the sense that the retrieval technique insures only the fact that the retrieved combination of all of the parameters would accurately reproduce the measured radiation field in the scope of chosen radiative transfer model.

2.3. Vertical profile of aerosol observed by lidar

We have used an aerosol micropulse Lidar manufactured by CIMEL (Pelon et al., 2008). It uses a Q-switched frequency-doubled ND:YAG laser with an expanded beam (14 μJ with a 200 mm exit-lens diameter) and a pulse repetition frequency of 4.7 kHz. The wavelength is 532 nm. During a 10 min data acquisition sequence, 10 individual profiles are acquired and averaged. Then the system waits for 20 min before starting another acquisition sequence. The duration of a pulse is 100 ns leading to a vertical resolution of 15 m. The profiles are averaged to reduce the influence of background noise. During the day time the background noise is dominated by direct or scattered sunlight causing a sharp decrease in the signal-to-noise ratio. The background noise is estimated by taking the average of the backscatter signal between 22 and 30 km, then subtracting it before evaluating the signal. The data processing