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# Optical Measurements of Atmospheric Aerosols in Air Quality Monitoring

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## 1. Introduction

Human activities are changing the composition of Earth's atmosphere, and thus influence the air quality. In that respect, one of the most important elements of the atmosphere are aerosols. They play a fundamental role in physical and chemical processes affecting both air quality and regional and global climate. Aerosols affect the Earth's radiative balance both in the cloud-free and the cloudy atmosphere. These so-called direct and indirect aerosol effects which depend on the chemical and physical aerosol properties still represent an uncertain factor in estimates about climate change.

The direct effect of aerosols is related with scattering and absorption of solar radiation, and as a consequence, reduction of the amount of radiation reaching the surface (e.g. IPCC 2001). In case of indirect effects, we distinguish the first and the second one. Aerosols act as cloud condensation nuclei (CCN), modifying the optical and radiative properties of clouds, e.g. albedo. This is known as the first indirect effect (Twomey, 1977). The second indirect effect is related to the shift in the cloud droplet spectrum resulting from the nucleation. It is seen by a decrease in precipitation and an increase in cloud lifetime (e.g. Charlson et al., 1992).

The best parameters that quantify the direct aerosol effect are the aerosol extinction, linked also to atmospheric visibility, and the aerosol optical thickness. The aerosol extinction reveals how aerosols attenuate the solar radiation in relation to a given distance (e.g. per km); the aerosol optical thickness is the extinction integrated over a whole column of atmosphere (usually in the vertical from the surface to the top of the atmosphere). Aerosol extinction can be derived from in-situ measurements (aerosol counters, impactors) or from path-integrated measurements (transmissometer); aerosol optical thickness is usually obtained from sun photometer measurements.

Nowadays, in the era of AERONET, information about the aerosol optical thickness (AOT) is available online for nearly 400 sites in the world; only in Europe the number of AERONET station is more than 80. Hence, more and more researchers try to find a relationship between the aerosol optical thickness (AOT) and  $PM_{10}$  (particulate matter with an aerodynamic diameter of less than 10  $\mu m$ ) in order to use it as an indicator of air quality. It should be emphasized that the aerosol optical thickness values represent vertical column-integrated properties whereas the  $PM_{10}$  data are the "surface" data. This kind of comparison is justified in the well-mixed boundary layer. Therefore not always both parameters are well correlated. Nevertheless, such a relationship between (AOT) and  $PM_{10}$  would be useful in air quality monitoring.

The variation of the extinction coefficient with wavelength can be presented as a power law function with a constant (related to the power factor) known as the Ångström coefficient.

When the particle size distribution is dominated by small particles, usually associated with pollution, the Ångström coefficients are high; in clear conditions they are usually low. Long residence time of air masses over land and in particular the passage over large urban areas cause high concentrations of fine particles and thus high values of the Ångström coefficients. The opposite effect can be observed over water. The longer the time that the air masses spent over water the more evident is a change in the aerosol size distribution caused by the deposition of continental aerosols. As a result of this process the measured Ångström coefficient values become much smaller. Therefore this parameter is a good tracer for the concentration of aerosols originated over land e.g. black carbon (Kuśmierczyk-Michulec et al., 2007; Kuśmierczyk-Michulec & Van Eijk, 2007). This chapter will give an overview of various efforts tending toward finding a relationship between AOT and PM<sub>10</sub>. Moreover, it will discuss possibility of using the Ångström coefficient in air quality estimation. The aerosol optical thickness and the Ångström coefficient are routinely monitored by AERONET sun photometers, or can be estimated using satellite data, therefore their application in air quality monitoring would be of great value.

## 2. Methodology

This section presents definitions of the main parameters usually used by various authors in the AOT-PM formulas.

### 2.1 Aerosol size distribution and related optical parameters: extinction, aerosol optical thickness and Ångström coefficient

The aerosol size distribution can be represented by the number size distribution  $N(r)$ , the volume size distribution  $V(r)$ , or by the mass size distribution  $m(r)$ . In each case the lognormal function is used and details are presented e.g. by Seinfeld and Pandis (1998).

The aerosol number size distribution for a given aerosol type can be presented by the following equation, where  $r$  is the particle radius,  $r_n$  is the median radius,  $\sigma$  is the standard deviation and  $N_n$  is particle concentration:

$$\frac{dN(r)}{d\ln r} = \frac{N_n}{\sigma\sqrt{2\pi}} \exp\left\{-\frac{(\ln r - \ln r_n)^2}{2\sigma^2}\right\} \quad (1)$$

From the number size distribution can be derived directly an effective radius  $R_{eff}$  (in  $\mu\text{m}$ ):

$$R_{eff} = \frac{\int r^3 \frac{dN(r)}{d\ln r} d\ln r}{\int r^2 \frac{dN(r)}{d\ln r} d\ln r} \quad (2)$$

The effective radius is a very useful parameter in characterization of the aerosol mixtures. Assuming that particles are spherical the following relation can be used to calculate the mass concentration PM at the surface in ( $\mu\text{g}/\text{m}^3$ ):

$$PM = \frac{4}{3}\pi\rho \int_{r_{\min}}^{r_{\max}} r^3 \frac{dN(r)}{d\ln r} d\ln r \quad (3)$$

where  $\rho$  is the aerosol mass density in ( $\text{g}/\text{cm}^3$ ).

The spectral optical coefficient, called the extinction coefficient  $ext(\lambda)$  (in  $\text{km}^{-1}$ ) can be calculated from the number size distribution:

$$ext(\lambda) = \pi \int_{r_{\min}}^{r_{\max}} r^2 Q_{ext} \frac{dN(r)}{d \ln r} d \ln r \quad (4)$$

where  $\lambda$  is wavelength,  $r$  is radius and  $Q_{ex}$  is the extinction efficiency factor, being a function of the complex index of refraction (Mie, 1908). The coefficient  $Q_{ex}$  can be calculated according to algorithm published by Bohren and Huffman (1983). The size-distribution integrated extinction efficiency  $\langle Q_{ext} \rangle$  is defined as (Hansen & Travis, 1974):

$$\langle Q_{ext} \rangle = \frac{\int r^2 Q_{ext} \frac{dN(r)}{d \ln r} d \ln r}{\int r^2 \frac{dN(r)}{d \ln r} d \ln r} \quad (5)$$

The extinction coefficient integrated over the whole column of atmosphere is a dimensionless parameter and it is called the aerosol optical thickness (AOT):

$$AOT(\lambda) = \int_{H_{\min}}^{H_{\max}} ext(\lambda, h) dh \approx \int_{H_{\min}}^{H_{\max}} ext(\lambda) f(h) dh \quad (6)$$

where  $f(h)$  represents the vertical distribution of aerosols,  $h$  is the altitude in km,  $H_{\min}$  and  $H_{\max}$  are the lower and the upper altitude, respectively, at which a given aerosol type can be found. Variation of the extinction coefficient with the wavelength can be presented in the form of a power law function (Ångström, 1929):

$$ext(\lambda) = \gamma_c \lambda^{-\alpha} \quad (7)$$

The same type of relation is also valid for the aerosol optical thickness,

$$AOT(\lambda) = \gamma_\tau \lambda^{-\alpha} \quad (8)$$

where  $\gamma_c$  and  $\gamma_\tau$  are constant and  $\alpha$  is the Ångström coefficient (also known as Ångström exponent or Ångström parameter). Usually, this parameter is determined in the spectral range from 440 nm to 870 nm.

From the set of equations 2-6 the following relation between  $PM$  and  $AOT$  can be derived:

$$PM = \xi \times AOT(\lambda) \quad (9)$$

where coefficient  $\xi$  is defined as:

$$\xi = \frac{4\rho R_{eff}}{3 \langle Q_{ext} \rangle H} \quad (10)$$

and the vertical distribution  $H$  is given by:

$$H = \int_{H_{\min}}^{H_{\max}} f(h)dh \quad (11)$$

## 2.2 Visual range (VR) and air pollution index (API)

Some authors looking for the best correlation between the particulate matter (PM) and optical parameters take into account not only the standard parameters like AOT or aerosol extinction but also other parameters like visual range (e.g. Bäumer et al., 2008; Jung et al., 2009) or air pollution index (e.g. Xia et al., 2006; Lasserre et al., 2008). Because of that reason it is worth to present definitions of the above parameters.

According to the empirical formula by Koschmieder (1925) the total atmospheric extinction coefficient at 550 nm is inversely proportional to horizontal visibility, known also as visual range (VR) in (km):

$$VR = \frac{-\ln(0.02)}{ext(550)} \quad (12)$$

Air Pollution Index (API) is a number used by government agencies to characterize the quality of the air at a given location. For example in China, the estimates of API are provided by the State Environmental Protection Agency (SEPA), ([http://www.sepa.gov.cn/english/air\\_s.php3](http://www.sepa.gov.cn/english/air_s.php3)). The API level in China is based on the level of 5 atmospheric pollutants, namely sulfur dioxide (SO<sub>2</sub>), nitrogen dioxide (NO<sub>2</sub>), suspended particulates (PM<sub>10</sub>), carbon monoxide (CO), and ozone (O<sub>3</sub>) measured at the monitoring stations throughout each city. An API daily value gives information about the most concentrated pollutant of the day. Some authors (e.g. Lasserre et al. 2008) used API index to determine the PM<sub>10</sub> concentration. However in such a case the exact chemical composition is not known.

## 3. Experimental

The overview of the main experimental techniques used to measure the particulate matter (PM) and optical properties like AOT and aerosol extinction are shortly presented in the subsequent subsections. Since the aim of this paper is to give an overview of different approaches leading to a relationships between PM and AOT, it is worth to mention as well various experimental techniques. None of them is perfect. In consequence the method of measurements can have an effect on such a relationship.

### 3.1 Aerosol optical thickness (AOT) measurements

#### 3.1.1 Sun photometer measurements

The spectral aerosol optical thickness and derived aerosol properties are routinely available from AERONET network (Holben et al., 2001). The measurements are made with CIMEL sun photometers. The instruments measure the direct solar radiation, from which the spectral values of the aerosol optical thickness can be derived. Most of the instruments in this network have the following channels: 440 nm, 670 nm, 870 nm and 1020 nm; some of them have more channels and thus can give more detailed spectral information. In addition, the sky radiance in aerosol channels in the azimuth plane (the almucantar technique) and in the principal plane is measured. The technical details of the instrument are described in the

Cimel Sun Photometer Manual. These data are used in the AERONET standard procedures to retrieve information on columnar aerosol characteristics such as the aerosol optical thickness, Ångström coefficient and size information. Data processing, cloud-screening algorithm, and inversion techniques are described by Holben et al. (1998, 2001), Eck et al. (1999), Smirnov et al. (2000), Dubovik and King (2000), and Dubovik et al. (2000).

### 3.1.2 Multi-Filter Shadow-band Radiometer (MFSR) measurements

The spectral aerosol optical thickness data can be also obtained from the measurements using the multi-filter shadow-band radiometer. The instrument measures simultaneously the total sky irradiance and the diffuse sky irradiance. These measurements allow estimation of the direct solar beam irradiance at the central wavelengths in each channel, and then, the aerosol optical thickness spectral values (e.g. Harrison and Michalsky, 1994; Olszewski et al., 1995). Similarly to sun-photometer measurements, the high quality MFSR measurements can be performed only for cloud-free days. This method of measurements of AOT was reported for example by Vaughan et al. (2001) or Alexandrov et al. (2005).

### 3.1.3 Satellite retrievals of the aerosol optical thickness

Satellites are best suited to determine the spatial distribution of aerosols over large areas during extended time periods, needed to evaluate their effects. Satellite observations since more than 25 years provide the aerosol index (AI) from the Total Ozone Mapping Spectrometer (TOMS) (Herman et al., 1997) and, over the oceans, the aerosol optical thickness (AOT) from the Advanced Very High Resolution Radiometer (AVHRR). Instruments become more and more sophisticated, reaching the compromise between spatial and spectral resolution, i.e., the Moderate Resolution Imaging Spectrometer (MODIS) (Kaufman and Tanré, 1996).

The accuracy of the aerosol optical thickness derived from the satellite data depends on the quality of data (e.g. radiometric calibration) but also on the algorithm used for conversion of satellite data to the AOT data. Since the retrieving process of AOT is performed only during daytime and for cloud-free pixels, the important step is the proper cloud mask. The smaller is a pixel size the more efficient is the process of removing cloudy pixels. Usually, instruments used for aerosol retrieval are designed for this purpose, such as Along Track Scanning Radiometer 2 (ATSR-2) or MODIS, and have small pixel sizes. Use of instruments with high spectral resolution but with a large pixel size, like for a example Global Ozone Monitoring Experiment (GOME) or Scanning Absorption Spectrometer for Atmospheric Chartography (SCIAMACHY) results in a very low probability of clear sky pixels which limits the usefulness of such instruments for aerosol retrieval (e.g. Kuśmierczyk-Michulec and De Leeuw, 2005).

It is worth to notice that the AOT retrieved from satellite data is the end product of a very complex data processing which involves many assumptions and may be a source of many uncertainties. The total radiance received by a sensor at the top of the atmosphere (TOA) contains contributions from aerosols, molecules, and the surface. Instead of radiance, it is often more convenient to use reflectance, i.e. a dimensionless function, defining the ratio of the measured radiance to the solar flux at the top of the atmosphere. In both representations the molecular contribution is assumed to be well known.

The main issue is the separation of the aerosol and surface contributions to the total reflectance. The high underestimation in the surface reflectance (e.g. over bright surface),

results in a large overestimation of the retrieved aerosol optical thickness. Similarly, the overestimation in the surface reflectance values may lead to underestimation of the AOT values, producing even the negative AOT values.

Thus, the proper surface correction is crucial. Over land the surface contribution to the TOA reflectance in the spectral range between 380 nm and 670 nm is not higher than 60% (except in the case of snow) (Kuśmierczyk-Michulec and De Leeuw, 2005). This contribution increases with wavelength reaching a value of around 80% at 770 nm. These numbers are based on the analysis of the surface reflectances taken from the GOME surface reflectance database (Koelemeijer et al., 2003). Over water, the proper surface correction is also important, especially in the UV and the visible part of spectrum. In that range the contribution of the water-leaving reflectance to the TOA reflectance is the most significant, around 60%, depending on the type of water and atmosphere. This contribution decreases with the wavelength, reaching at 670 nm around 1–8%. For Case I waters (Morel, 1988), i.e., mainly oceanic waters which optical properties are determined by phytoplankton and their immediate derivatives, the contribution at 670 nm will be almost negligible, because the water-leaving reflectance is close to zero. For Case II waters (Morel, 1988), i.e., all coastal waters which optical properties are also determined by the presence of sediments or dissolved yellow substances, this contribution can be much higher.

To account for the surface contribution different approaches, depending on instruments, can be used. For example, in case of MODIS instrument, the surface reflectivity at visible wavelengths is obtained by assuming a constant ratio between surface reflectivity at 2100 nm and that at 470 and 660 nm (eg. Chu et al., 2003; Remer et al., 2005). For MERIS data Retalis & Sifakis (2010) suggested using the differential textural analysis (DTA) algorithm which quantifies the contrast reduction as local “textural degradation” by comparing “a pollution image” and a “reference image”, representing a day with the lowest level of aerosol concentration. For the aerosol retrieval with GOME data (Kuśmierczyk-Michulec and De Leeuw, 2005), the surface reflectance databases can be used (Koelemeijer et al., 2003) or combined approach using two instruments: GOME and ATSR-2 (Holzer-Popp et al., 2002).

The accuracy of the retrieved aerosol optical thickness values is tested by comparison with collocated sun photometer measurements e.g. available from the AERONET stations. In this way a bias between retrieved and measured AOT values can be found. It is especially important in case both AOT from ground measurements and satellite retrievals are used as a one dataset.

### 3.2 Aerosol extinction measurements

To measure the aerosol extinction, several methods can be used. Wang et al. (2010) reported the aerosol extinction measurements carried out by a visibility meter. According to the empirical formula by Koschmieder (1925) the total atmospheric extinction coefficient at 550 nm is inversely proportional to horizontal visibility. Hence, the aerosol extinction coefficient at 550 nm can be derived after removal the molecular contribution from the total atmospheric extinction coefficient.

Jung et al. (2009) used the transmissometer system, consisting of a transmitter and a receiver, to measure the atmospheric transmission over a long-path at a given wavelength. Using the exponential relationship between the transmittance and extinction, and removing the molecular contribution, the aerosol extinction can be determined (e.g. Kuśmierczyk-Michulec et al., 2008).

### 3.3 PM<sub>10</sub> measurements

The most frequently used techniques to measure PM<sub>10</sub> (particulate matter with an aerodynamic diameter of less than 10 µm) data are presented below. Some of these methods were used to determine PM<sub>2.5</sub> (particulate matter with an aerodynamic diameter of less than 2.5 µm) or even PM<sub>1.0</sub> (particulate matter with an aerodynamic diameter of less than 1 µm).

#### 3.3.1 Beta – absorption technique

The beta-absorption technique is used for example in The Netherlands (e.g. Kuśmierczyk-Michulec et al., 2007). The PM<sub>10</sub> (particulate matter with an aerodynamic diameter of less than 10 µm) and black carbon concentrations are measured by the Dutch National Air Quality Network (LML) operated by RIVM (<http://www.rivm.nl/milieukwaliteit/lucht/>). Black carbon (particles mostly smaller than 2.5 µm) is available as daily values and PM<sub>10</sub> is available on an hourly basis.

RIVM's National Air Quality Monitoring Network in the Netherlands performs continuous PM measurements i.e. PM<sub>10</sub> and PM<sub>2.5</sub>, using an FAG-type β-dust monitor (Van Elzakker, 2000). The sampling air is heated (to 50 °C) in order to remove water from aerosol particles (Buringh and Opperhuizen, 2002). The drawback of the heating is a removal of semi-volatile compounds eg. ammonium nitrate which leads to the losses in PM measurements. The correction methods for a systematic underestimation by the sampling equipment are described by Hammingh (2001) and discussed in detail by Buringh and Opperhuizen (2002). The amount of black carbon is estimated based on the PM measurements using the so-called light-reflectance method (Buringh and Opperhuizen, 2002).

#### 3.3.2 Tapered element oscillating microbalance (TEOM) technique

The tapered oscillating microbalance technique was used for example in Beijing (e.g. Xia et al., 2006; Wang et al., 2010), in Amazonia (Artaxo et al., 2002), in the South-Eastern France (e.g. Péré et al., 2009), in the North France and Belgium (Pelletier et al., 2007) or in Italy (Barnaba et al., 2010). Similarly to beta-absorption technique, the sampled air is heated before collecting the aerosols on a filter, to avoid humidification of the filter. In consequence of heating, a loss of semi-volatile compounds can occur. Using this method of measurements, an underestimation can exist on PM<sub>10</sub> values due to the volatilization of ammonium nitrate or semi-volatile organic species during the conditioning procedure at 50 °C (e.g. Allen et al., 1997). The uncertainty of PM<sub>10</sub> measurements using TEOM, reported by authors, is ± 10%.

#### 3.3.3 Aerosol spectrometer measurements

The aerosol spectrometer determines the concentration of aerosols by quantifying the scattering caused by the passage of particles of various sizes through a light beam produced by a laser diode. This technique was used for example in Beijing (e.g. Jung et al., 2009) to measure PM<sub>10</sub>, PM<sub>2.5</sub> and PM<sub>1.0</sub> mass concentrations. The PM concentrations were measured under dry condition (RH<40%) by diluting the sample air with dry clean air.

## 4. Influence of relative humidity (RH) on aerosol optical properties and particulate matter (PM)

When the ambient relative humidity is changing the hygroscopic atmospheric aerosols undergo processes of phase transformation, droplet growth, and evaporation. The phase

transformation from a solid particle to a saline droplet usually occurs spontaneously when the ambient relative humidity reaches a level called the deliquescence humidity. Its value is specific to the chemical composition of the aerosol particle (e.g. Orr et al., 1958; Tang, 1976).

The relative humidity influences the size of hygroscopic particles, the effective radius of an aerosol mixture, their density and finally the aerosol scattering properties. In consequence, values of the particulate matter (PM), the aerosol extinction, the aerosol optical thickness (AOT) and the Ångström coefficient are modified. Similar effect can be observed when the aerosol composition changes (e.g. Kuśmierczyk-Michulec, 2009).

The main aerosol components affected by a change in the relative humidity are sea-salts (SSA, i.e. NaCl) and anthropogenic salts e.g. ammonium nitrate ( $\text{NH}_4\text{NO}_3$ ) and ammonium sulphate ( $(\text{NH}_4)_2\text{SO}_4$ ). It should be mentioned that another aerosol component on which the relative humidity could have an influence is organic carbon (OC). In contrast to black carbon (BC) which is considered to be insoluble (e.g. Kanakidou et al., 2005), about 40% of organic carbon (OC) is soluble (Rulleau, 2000, personal communication; IPCC, 2007). However, there is still a lack of detailed description of the interactions among organic and inorganic aerosol compounds and water (e.g. Kanakidou et al., 2005). The interest in the hygroscopic properties of OC relevant to atmospheric applications started within the last few years only and the data are still rather scarce. Hence, considering all associated uncertainties, the common assumption is that OC is not hygroscopic.

The relative humidity modify the optical properties not only the hygroscopic aerosol mixtures but also mixtures containing some contribution of non-hygroscopic aerosols like for example organic carbon or black carbon (e.g. Kuśmierczyk-Michulec, 2009). Due to the wetting process the hygroscopic particles grow, modifying the effective radius of an aerosol mixture and finally the values of aerosol extinction or the aerosol optical thickness. This effect is substantial especially when  $\text{RH} > 90\%$ ; at  $\text{RH} = 98\%$  an increase in AOT value may be up to 6 times its value at  $\text{RH} = 80\%$  (Kuśmierczyk-Michulec, 2009). This effect is observed at different wavelengths, but for higher RH, the increase in AOT values is more evident at 412 nm than at 865 nm. As a consequence of such a non-uniform increase, the Ångström coefficient also becomes a function of RH.

The observed variations in the Ångström coefficient values can be explained by changes in the effective radius of a mixture. These changes can be caused by relative humidity, as well as, by aerosol composition. For example, an increased contribution of small aerosol particles causes that the effective radius decreases and the Ångström coefficient increases. However, increase or decrease in relative humidity does not exert such a change in the Ångström coefficient like the one caused by modification in the composition of an aerosol mixture (Kuśmierczyk-Michulec, 2009).

A change from dry to wet radius, and from dry to wet density depends on the aerosol component, and more precisely on the type of salt. The general formulas proposed by some authors like for example Gong et al. (1997) are practical but not so accurate like formulas for different salts based on the laboratory work and suggested by Tang (1996) and Tang and Munkelwitz (1994). In case of known aerosol composition it is better to use the formulas suggested by the latter authors.

## 5. Relationships between optical parameters and particular matter

### 5.1 Linear and non-linear models: overview

In the past years a number of studies have explored the possibility of using optical parameters for evaluation air quality and estimation the particular matter (PM)

concentration. Most of these studies focused on finding a linear relationship between AOT data obtained from ground-based sun photometers and  $PM_{10}$  measurements (e.g. Chu et al., 2003; Xia et al., 2006; Kuśmierczyk-Michulec et al., 2007; Péré et al., 2009; Wang et al., 2010; Barnaba et al., 2010) or  $PM_{2.5}$  (e.g. Jung et al., 2009; Wang et al., 2010). To improve the AOT-PM relationship some authors have investigated the possibility of using non-linear models (e.g. Pelletier et al., 2007; Liu et al., 2005, Vidot et al., 2007). Jung et al. (2009) have found that non-linear models between visual range and PM concentrations ( $PM_{10}$ ,  $PM_{2.5}$ ,  $PM_{1.0}$ ) give very high correlation coefficients (cf. Table 1).

Similarly to the ground-based AOT, also a possibility of using satellite-derived AOT has been explored. For that purpose mostly MODIS data has been used (e.g. Wang & Christopher, 2003; Engle-Cox et al., 2004; Li et al., 2005a, 2005b; Koelemijer et al., 2006; Barnaba et al., 2010), but also MERIS data (e.g. Retalis & Sifakis, 2010), SeaWiFS data (e.g. Vidot et al., 2007; Fischer et al., 2009) and MISR data (e.g. Liu et al., 2005). Since the aerosol retrieval is quite complex thus the AOTs derived from satellite are less accurate than ground-based AOT, and hence a weaker correlation between AOT-PM (c.f. Table 1).

AOT represents the column integrated value while PM is measured at ground level, thus the correlation between them may depend on the vertical distribution of aerosols. In case a boundary layer is well-mixed, the column integrated value should be a good indicator of a surface observation and vice versa. Otherwise, the vertical distribution of aerosols should be taken into account. Another factor having an influence on the AOT-PM relationship is the relative humidity. While PM represents dry aerosols, the AOT measurements are taken at ambient conditions. In case the hygroscopic aerosols are present, an increase or decrease in RH causes changes in their sizes, volume and density, and eventually in the aerosol optical thickness values. Changes in RH may also explain sometimes observed seasonal changes in the AOT values (e.g. Kuśmierczyk-Michulec & Rozwadowska, 1999) which may lead to observed seasonal variations between AOT-PM relationships (e.g. Xia et al. 2006). In winter the air is much drier than in summer, so in winter RH should have lower impact. On the other hand, the altitude of the mixing layer is lower in winter; thus for a given AOT the PM value is expected to be larger in winter than in summer.

Thus, estimation of PM concentrations from AOT may lead to significant errors. To reduce the uncertainties, some authors (e.g. Koelemijer et al., 2006; Li et al., 2005a) have suggested scaling of the AOT values by dividing them by the boundary layer height and the hygroscopic growing factor describing the increase of the aerosol extinction cross-section with relative humidity. The positive result of this approach was reported by e.g. Koelemijer et al. (2006). Similar improvements were noted by Wang et al. (2010). To improve the correlation between the aerosol optical thickness and the particulate matter, the authors applied the vertical-and-RH correcting method to the measurements in Beijing. The authors found that the correlation coefficients between AOT and  $PM_{10}$  and between AOT and  $PM_{2.5}$  have increased from 0.52 to 0.65 and from 0.48 to 0.62 respectively. Analogous effect was achieved by Barnaba et al. (2010). To get information about aerosol vertical profiles, the authors used the lidar measurements to calculate the monthly AOT weighing functions as the ratio between the lidar-derived monthly median aerosol extinction profiles and the associated AOT. These monthly vertical scaling factors allowed for conversion the AOT values into the surface aerosol extinction values. Next, after applying the RH-correction, the dry aerosol extinction values were compared with the dry PM values leading to the significant increase correlation coefficient (cf. Table 1). The potential of this method was demonstrated for the data from the Po Valley (Italy).

Source	Study area	Model description	Optical parameters	R <sup>2</sup>
Kuśmierczyk-Michulec et al., (2002)	The Baltic Sea	Non-linear models: $\alpha$ & BC/TPM $\alpha$ & OC/TPM $\alpha$ & SSA/TPM	Angstrom coefficient ( $\alpha$ )	0.84 0.88 0.91
Chu et al. (2003)	Ispra (Italy)	Linear model between AOT & PM10: PM10=55 x AOT +8	Ground based AOT	0.82
Wang & Christopher (2003)	Alabama (USA)	Linear model between AOT & PM2.5	AOT retrieved from MODIS 10 km	0.49
Engle-Cox et al. (2004)	USA	Linear model between AOT & PM2.5	AOT retrieved from MODIS 10 km	0.40
Li et al. (2005a)	Beijing (China)	Linear model between AOT & PM10 (API)	AOT retrieved from MODIS (corrected) 10 km	0.29
Li et al. (2005b)	Hong Kong (China)	Linear model between AOT & PM10	AOT retrieved from MODIS 1km	0.50
Liu et al. (2005)	Eastern USA	Non-linear model between AOT & PM2.5	AOT retrieved from MISR	0.48
Xia et al. (2006)	Beijing (China)	Linear model between AOT & PM10: summer/autumn/ winter/spring	Ground based AOT at 440 nm	0.77 0.70 0.61 0.37
Koelmijer et al. (2006)	Europe	Linear models between AOT & PM10 AOT & PM2.5	AOT at 550 nm retrieved from MODIS (corrected) 10 km	0.46 0.59
Vidot et al. (2007)	Western Europe	Non linear models: AOT & PM10 AOT & PM2.5	AOT retrieved from SeaWiFS	0.43 0.61
Pelletier et al. (2007)	Lille (France)	Non linear model between AOT & PM10; Semi parametric approach (Principal Component Analysis)	Ground based AOT at 440, 670 & 870 nm	0.76
Kuśmierczyk-Michulec et al., (2007)	The Hague (The Netherlands)	Linear model between AOT & PM10 Non-linear models $\alpha$ & BC/PM10: 1) PM<50 $\mu$ g/m <sup>3</sup> 2) 50 $\mu$ g/m <sup>3</sup> ≤PM<200 g/m <sup>3</sup>	Ground based AOT at 555 nm Angstrom coefficient ( $\alpha$ )	0.59 0.83 0.84
Péré et al. (2009)	South-Eastern France	Linear model between AOT & PM10: PM10=54 x AOT +13	Ground based AOT at 440 nm	0.69
Jung et al. (2009)	Beijing (China)	Non-linear models: VR & PM1 VR & PM2.5 VR & PM10 Linear models between AOT & PM2.5	Visual range (VR) based on aerosol extinction at 550 nm Ground based AOT at 550 nm	0.93 0.93 0.81 0.70 0.44

Source	Study area	Model description	Optical parameters	R <sup>2</sup>
Fischer et al. (2009)	The Taklaman & Gobi Deserts/ The U.S. Pacific Northwest	Linear models between AOT & PM <sub>2.5</sub> AOT & PM <sub>10</sub>	AOT at 550nm retrieved from SeaWiFS, selected for $\alpha \leq 0.7$	0.48 0.50
Barnaba et al. (2010)	The Po Valley (Italy)	Linear models between AOT & PM <sub>10</sub> AOT (corrected) & PM <sub>10</sub> AOT & PM <sub>10</sub> AOT (corrected) & PM <sub>10</sub>	Ground based AOT at 500 nm  AOT at 550nm retrieved from MODIS, 50 km	0.66 0.94  0.32 0.35
Wang et al. (2010)	Beijing (China)	Linear models between AOT & PM <sub>10</sub> $PM_{10} = 96.96 \times AOT + 22.67$ ext & PM <sub>10</sub> AOT & PM <sub>2.5</sub> $PM_{2.5} = 19.58 \times AOT + 8.52$ ext & PM <sub>2.5</sub>	Ground based AOT at 550 nm  Aerosol extinction (ext) at 550 nm	0.52 0.77  0.48 0.66
Retalis & Sifakis (2010)	Athens (Greece)	Linear model between AOT & PM <sub>10</sub> : $PM_{10} = 195.7 \times AOT + 14.5$	AOT at 560 nm retrieved from MERIS	0.83

R<sup>2</sup>- squared correlation coefficient, TPM- total particulate matter, SSA - sea salts

Table 1. Models associating optical parameters with particular matter concentrations in some of the previous studies.

## 5.2 Use of the Ångström coefficient in air quality estimation

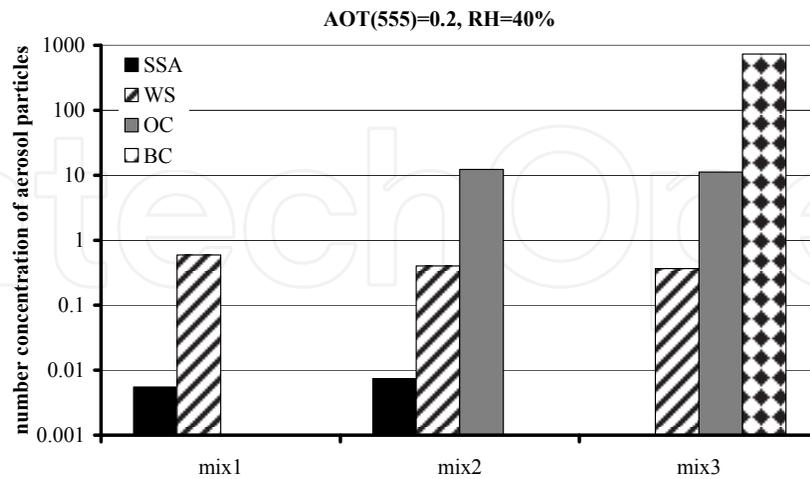
### 5.2.1 Importance of the spectral measurements

Measurements at one single wavelength will not supply accurate information about the aerosol mixture. Figure 1 shows that one single measurement of the aerosol optical thickness at one wavelength, e.g.  $\lambda = 555$  nm, will not provide unique information about an aerosol type. The condition that  $AOT(555) = 0.2$  at RH=40% was satisfied by three different mixtures (Figure 1A). Mixture 1 (mix1) consists of 40% of WS ( $NH_4HSO_4$ ,  $r_n = 0.112$ ,  $\sigma = 0.58$ ), and 60% of SSA (NaCl,  $r_n = 0.5$ ,  $\sigma = 0.56$ ). Mixture 2 (mix2) consists of 20% of WS, 20% of OC (Organic Carbon,  $r_n = 0.06$ ,  $\sigma = 0.3$ ) and 60% of SSA. Mixture 3 (mix3) consists of 40% of WS, 40% of OC and 20% of BC (Black Carbon,  $r_n = 0.0118$ ,  $\sigma = 0.3$ ). In consequence, depending on the type of aerosol mixture, different PM values can be obtained. For example at RH=40%, a mixture 2 would lead to approximately 1.4 times higher mass than mixture 1. Accordingly, a mixture 3 would produce approximately 0.6 times lower mass than mixture 1. The reason is that mixture 3 contains a large amount of fine particles of BC, which results in much smaller total mass.

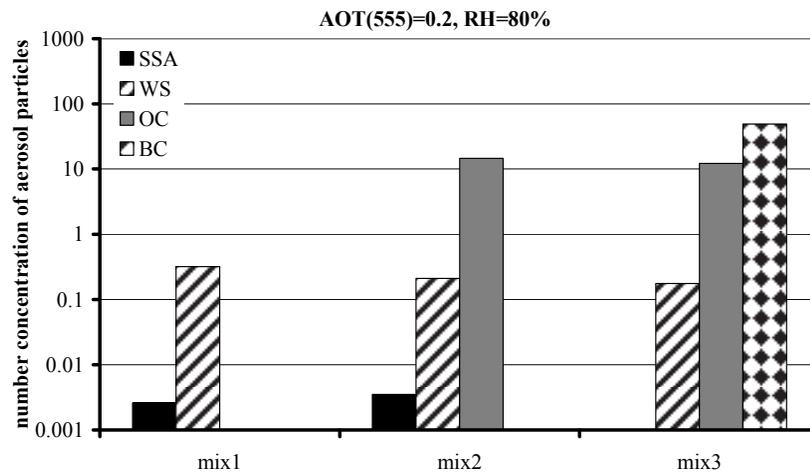
The similar situation is demonstrated in Figure 1B, but for RH=80%. Three different mixtures satisfy the condition that  $AOT(555) = 0.2$  at RH=80%. Figure 1C shows that a distinction among mixtures is possible using the Ångström coefficient as an additional parameter. In each case the same composition of aerosol mixtures were used. The effect of

humidity was taken into account by using the formulas proposed by Tang (1976, 1996) and Tang & Munkelwitz (1994).

A)



B)



C)

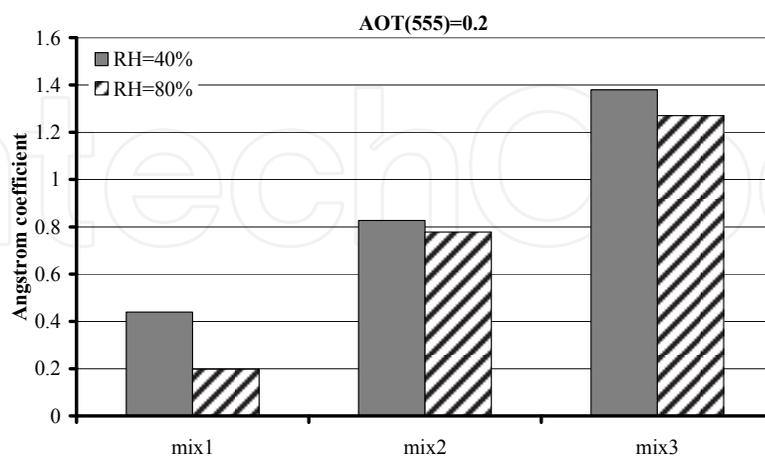


Fig. 1. Three different aerosol mixtures giving as a final result AOT(555)=0.2. The composition of mixtures is as follows: mix1: 40% of WS, 60% of SSA; mix2: 20% of WS, 20% of OC, 60% of SSA; mix3: 40% of WS, 40% of OC, 20% of BC. A) & B) Number concentration of aerosol particles in each mixtures estimated for RH=40% and RH=80%, respectively. C) The Ångström coefficient as a function of aerosol mixture and relative humidity.

### 5.2.2 Ambiguity of the proposed formulas

Table 1 gives an overview of various linear and non-linear formulas, developed for different geographical locations and based on measurements taken under different ambient conditions. Some formulas forecast the amount of PM on the basis of dry AOT values; other use AOT values defined at ambient conditions. The AOT is a function of wavelength. Table 1 demonstrates that usually a wavelength for which AOT was determined depends on the type of instrument used for measurements. Hence, the variety of spectral bands: 440 nm (eg. Péré et al. 2009), 550 nm (e.g. Jung et al. 2009), 555 nm (Kuśmierczyk-Michulec et. al. 2007), 500 nm (Barnaba et al., 2010) or 560 nm (Retalis & Sifakis, 2010). Because of that reason a direct comparison is often not straightforward. In addition, most of the formulas presented in Table 1 do not inform about chemical composition. However, it should be emphasized that the AOT-PM relationship depends on the chemical composition of the atmospheric aerosol mixture.

In general transition from AOT at one wavelength to AOT at another wavelength can be done using the Ångström coefficient (see eq. 8). In case the chemical composition is not known and the spectral measurements are not available, the Ångström coefficient can not be determined. In such a case, a direct comparison between amounts of PM in various locations may lead to ambiguity.

Let's assume that AOT at 550 nm is 0.2. Depending on the aerosol type, the Ångström coefficients will vary, from very low, even negative ones corresponding to e.g. desert dust, to very high values, typical for the industrial aerosol type. The higher is the Ångström coefficient the more significant is the contribution of fine particles usually related to the presence of black carbon or anthropogenic salts (e.g. Kuśmierczyk-Michulec, 2009). To compare PM values based on AOT at 550 nm with PM values determined using AOT at 440 nm, application of equation (8) is necessary. To illustrate this problem the formula of Péré et al. (2009) was selected as one of those which use AOT at 440 nm. Figure 2 shows possible changes in PM values caused only by the fact that the Ångström coefficient is not known. Figure 2 demonstrates that lack of information about the Ångström coefficient value may lead to large uncertainties in PM estimation. A difference between the extreme cases (i.e.  $\alpha = -0.5$  and  $\alpha = 2$ ) is about  $8 \mu\text{g}/\text{m}^3$ .

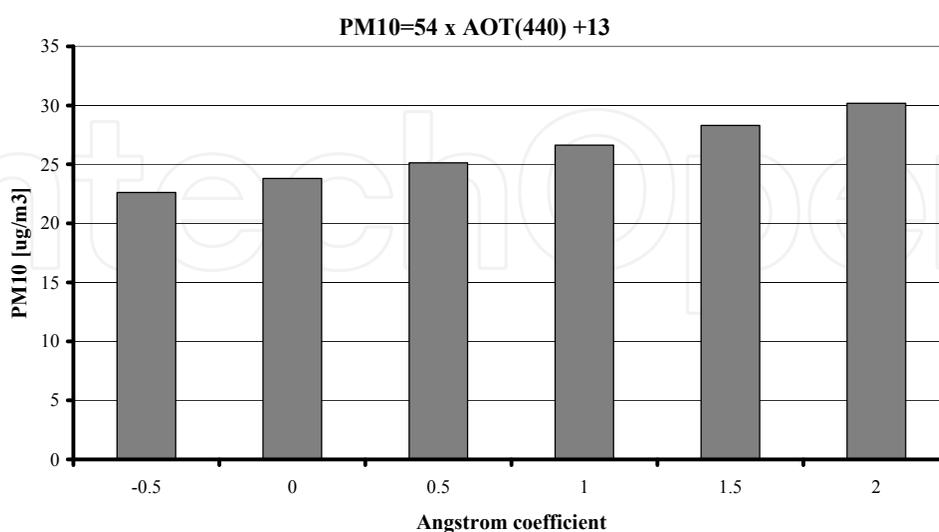


Fig. 2. Influence of the aerosol type, defined by the different Ångström coefficient values on the estimation of  $\text{PM}_{10}$  values. The simulations are based on the formula proposed by Péré et al. (2009) for the South-Eastern France.

Another aspect that should be mentioned is a seasonal character of a data sample which is used to derive AOT-PM relationship. In winter time the amount of PM is usually much higher than in summer time, hence possible variations between various formulas. Because of that reason one formula derived for summer data will give different results than a formulae based on the winter data. Figure 3 illustrates such a situation. The amount of PM<sub>10</sub> for the Netherlands depends on season, or more precisely whether a formula was obtained for winter data or other seasons. It is interesting to notice that PM<sub>10</sub> for France, estimated on the basis of formula obtained for summer data, gives comparable values as the “summer” formula for the Netherlands.

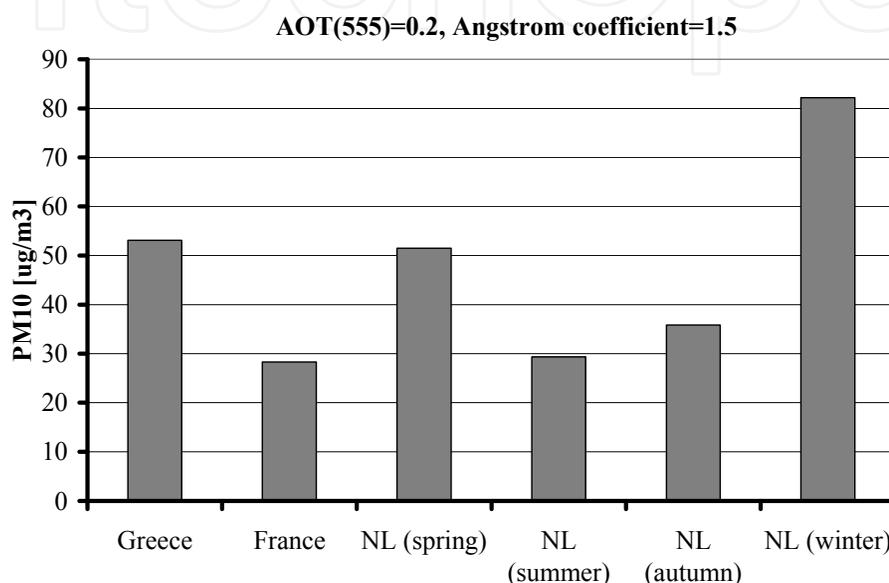


Fig. 3. Estimation of PM<sub>10</sub> for three locations in Europe, for the same AOT. The simulations are based on the following formulas: Greece (Retalis & Sifakis, 2010), France (Péré et al., 2009) and The Netherlands (Kuśmierczyk-Michulec et al., 2007).

### 5.2.3 Application of the Ångström coefficient

When aerosol measurements are made at a land, it can be expected that they represent a mixture which composition depends on the aerosol sources and the air mass history. The large influence will have the type of aerosol emitted by nearby sources, e.g. factories. In general, the main aerosol components of natural atmospheric mixtures include sea-salts (SSA), black carbon (BC), organic carbon (OC), dust-like particles (DL), and anthropogenic salts like ammonium hydrogen sulphate (NH<sub>4</sub>HSO<sub>4</sub>), ammonium nitrate (NH<sub>4</sub>NO<sub>3</sub>) and ammonium sulphate ((NH<sub>4</sub>)<sub>2</sub>SO<sub>4</sub>), which belong to a large group of water-soluble particles (WS). Each of these aerosol components is characterized by its own optical and physical properties like for example refractive index or aerosol size distribution (e.g. McClatchey et al., 1984). The optical properties of the aerosol mixture can be represented by the aerosol extinction or by the aerosol optical thickness. Both parameters can be modelled using the external mixing approach. The resultant aerosol optical thickness (or aerosol extinction) is obtained as the corresponding weighted averages of the extinction coefficients using the volume percentages (e.g. McClatchey et al., 1984; Hess et al., 1998). The influence of aerosol composition on aerosol optical thickness and the Ångström coefficient at various relative humidity conditions is discussed by Kuśmierczyk-Michulec (2009).

Since the aerosol optical thickness represents the integrated effect, measurements of AOT at one single wavelength will not supply accurate information about the aerosol mixture (see sections 5.2.1 & 5.2.2). If spectral measurements are available they provide an additional parameter, the Ångström coefficient. This parameter can help to indicate the probable aerosol type which can be useful information (Kuśmierczyk-Michulec, 2009). The Ångström coefficient is far less sensitive to variation in RH than AOT. Moreover, the variations in aerosol vertical distributions have less impact on this parameter.

The Ångström coefficient values include information about proportion between the fine and coarse modes. The fine mode usually can be attributed to the continental/industrial aerosols while the coarse mode represents sea-salts or dust particles (if there is evidence that dust particles can be present). In view of the dependence of the aerosol optical properties on the size distribution and the chemical composition, and the chemical speciation as function of particle size, a relation is expected between the Ångström coefficient and the chemical composition. Kuśmierczyk-Michulec et al. (2002) demonstrated that empirical relations between aerosol mass concentrations and Ångström parameters can be derived. The authors showed that the relations apply to mass concentrations of sea salts, black carbon and organic carbon, expressed in terms of their ratio to the total particulate matter.

The high correlation coefficients between  $BC/PM_{10}$  and the Ångström coefficients (Kuśmierczyk-Michulec et al., 2002; 2007) prove the usefulness of the latter parameter. Therefore it is recommended to use it as an additional parameter in air quality estimation.

## 6. Conclusions

The increasing number of AERONET stations and thus easy access to ground-based AOT data explains a growing interest in using optical measurements of aerosols in air quality estimation. Moreover, the continuous effort in making improvements of the aerosol retrieval from the satellite data indicates that optical measurements will be one of the standard future methods of estimation the level of pollution.

This chapter gives an overview of various efforts tending toward finding a relationship between AOT and  $PM_{10}$ . It discusses advantages and disadvantages of using optical measurements of atmospheric aerosols in estimation of the particulate matter. One of the aspects discussed is that the AOT-PM relationship depends on the chemical composition of the atmospheric aerosol mixture. Because of that reason, to avoid ambiguity in comparing various formulas proposed by different authors for different locations, it would be valuable to use the spectral measurements, and include the information about the Ångström coefficient.

Since the AOT-PM relationship is a simple parametric equation, having a very local character, we should accept that we will not obtain one general equation valid for all locations. However, it would be advantageous to use a similar methodology in obtaining such a relationship. It would make the comparisons between locations and formulas much easier.

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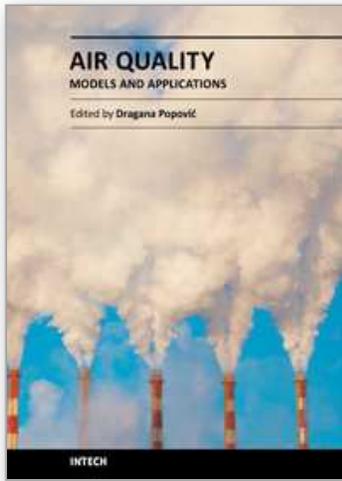
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Air pollution has been a major transboundary problem and a matter of global concern for decades. High concentrations of different air pollutants are particularly harmful to large cities residents, where numerous anthropogenic activities strongly influence the quality of air. Although there are many books on the subject, the one in front of you will hopefully fulfill some of the gaps in the area of air quality monitoring and modeling, and be of help to graduate students, professionals and researchers. The book is divided in five sections, dealing with mathematical models and computing techniques used in air pollution monitoring and forecasting; air pollution models and application; measuring methodologies in air pollution monitoring and control; experimental data on urban air pollution in China, Egypt, Northeastern U.S, Brazil and Romania; and finally, the health effects due to exposure to benzene, and on the influence of air pollutants on the acute respiratory diseases in children in Mexico.

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