

# Retrieval of aerosol mass load (PM<sub>10</sub>) from MERIS/Envisat top of atmosphere spectral reflectance measurements over Germany

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**Abstract.** Results of a new methodology for retrievals of surface particulate matter concentration (PM<sub>10</sub>) from satellite reflectance measurements over Germany are presented in this paper. The retrieval derives effective radii from Ångström- $\alpha$  exponents and benefits from the fitting of a smooth spectral slope from seven MERIS spectrometer channels. Comparisons with ground measurements from the air quality surveillance show standard deviations of 33.9% with –18.9% bias over Hamburg. Over rural sites a standard deviation of 17.9% (bias 12.9%) is reached.

We discuss critically limitations and potential applications of the retrieval. Additionally, we talk about the aspects at comparing of retrieved particulate matter with ground station measurements.

## 1 Introduction

Atmospheric pollution due to natural and anthropogenic emissions of aerosols is a nowadays recognized serious threat to human health due to respiratory and toxic adverse health effects. Studies estimated the increase of total mortality between 0.4 and 1% for each increase of 10  $\mu\text{g}/\text{m}^3$  in PM<sub>10</sub> concentration which mean for instance up to about 24 000 deaths in the USA alone each year (Pope III et al., 2002; Mokdad et al., 2004).

The US Environmental Protection Agency (EPA) introduced regulations and limits for the concentrations of particulate matter with diameters smaller than 10  $\mu\text{g}/\text{m}^3$  (PM<sub>10</sub>) in 1987, and since 1999 also relating to PM<sub>2.5</sub> due to the

more dangerous adverse respiratory health effects of these finer particles. Also since 1999, every country in the European Union has to provide a dense PM<sub>10</sub> measurement system for urban agglomeration with more than 250 000 people (EC, 1997, 2008). PM<sub>10</sub> concentrations shall not exceed 50  $\mu\text{g}/\text{m}^3$  for 35 days per annum in Europe, and 40  $\mu\text{g}/\text{m}^3$  on yearly average. In Germany, 1224 measurement stations, maintained by the federal states and partly by the German Environmental Agency (UBA), measure PM<sub>10</sub> concentrations every thirty minutes. Recently, obligations became more rigid and changed also referring to PM<sub>2.5</sub> (EC, 2008).

Since the early nineties, different airborne and satellite observations are used for the determination of the aerosol optical depths over land (e.g., Herman et al., 1997; King et al., 1999; Kaufman et al., 1997; Deuzé et al., 2001; von Hoyningen-Huene et al., 2003; Lee et al., 2006; Levy et al., 2007a,b) through which information about the aerosol mass load in the atmosphere and near the surface is provided. Aerosol optical depth information can nowadays be validated readily, for instance through the AERONET sun photometer reflectance measurements (Holben et al., 2001). The quality of the retrievals of aerosol optical depths from satellite data is nevertheless variable (Kokhanovsky et al., 2007; Kokhanovsky and de Leeuw, 2009) since the inversion still depends on assumptions and local models. Performing global aerosol retrievals is still a challenge.

Especially for large cities but also in order to fill measurement gaps in the national air quality surveillance systems, more accurately retrieved and validated satellite measurements provide additional information besides the cost-effective ground and sometimes not objective measurement gauging systems, in particular over rural sites where measurements are rare (Al-Saadi et al., 2005). The EPA already started an integrated decision support tool called the Three



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Dimensional Air Quality System (3D-AQS) (Hoff et al., 2006) which is an extension of the Infusing Satellite Data into Environmental Applications (IDEA) inferring aerosol optical depths from the MODerate resolution Imaging Spectroradiometer (MODIS) satellite.

Several approaches are used for a functional correlation between aerosol optical depth and the particulate matter mass concentration (e.g., Chu et al., 2003; Wang and Christopher, 2003; Kacenenbogen et al., 2006; Gupta et al., 2006; van Donkelaar et al., 2010; Koelemeijer et al., 2006; Glantz et al., 2009). This relation is strictly speaking only valid for fine mode particles, e.g., PM<sub>2.5</sub>. For coarse particles, such a relationship cannot be found because of their relatively small spectral impact (O'Neill et al., 2003). Estimations have been found for PM<sub>2.5</sub> (e.g., Engel-Cox et al., 2006, for the eastern USA), and (e.g., Glantz et al., 2009, for Stockholm). To deduce information about the mass load, such retrievals work with additional assumptions about the consistency of the local aerosol or additional information from other ground or space based LIght Detection And Ranging (LIDAR) information about the size distribution of the aerosols, i.e., information about local aerosol model (Engel-Cox et al., 2006) or using synergetic models (Pelletier et al., 2007; Vidot et al., 2007). Although the results are quite promising, those techniques are only successful for the region to which further information from ground measurements or from models are added.

In our methodology, we conclude to the aerosol mass through spectral information (i.e., the Ångström- $\alpha$  coefficient) and an empirical relationship to the effective radius of the aerosol particles. Integration over the distribution function then leads to the aerosol mass.

First theory of this presented methodology and results of PM<sub>10</sub> retrievals have been shown for a MERIS observation (von Hoyningen-Huene et al., 2006) and from retrievals above Moscow from SeaWiFS data (Kokhanovsky et al., 2006) and also over German sites without any corrections of the retrieval with respect to humidity, boundary layer height, temperature and other parameters as listed in Sect. 4. Kokhanovsky et al. (2009) presented first results of the same methodology but without improvements of the BRDF and of the PM<sub>10</sub> retrieval. They used only measurements of one day over Germany. In particular they neither used meteorological data like boundary layer heights – which are shown here to be necessary - nor they have made an adaption of the PM<sub>x</sub> measures by introducing filter functions.

We here present the algorithm description, results and comparisons of the retrieval results of boundary layer PM<sub>10</sub> from the Medium Resolution Imaging Spectrometer (MERIS) over Germany. The methodology is valid in particular for fine particulate matter, and is shown here to work over German sites for PM<sub>10</sub>: furthermore several corrections of the retrieval are presented (BRDF, inclusion of routine meteorological parameter, humidity correction, as well as adaptation of the definition of particulate matter mass).

The paper is structured as following: after introducing the retrieval methodology of AOD and PM<sub>10</sub>, we show comparisons of the aerosol optical depth measurements with those from AERONET and national air quality gauging stations in Germany, followed by a critical discussion about the limits and assumptions of the methodology as well as about the potential usage of the retrieved products for air quality surveillance.

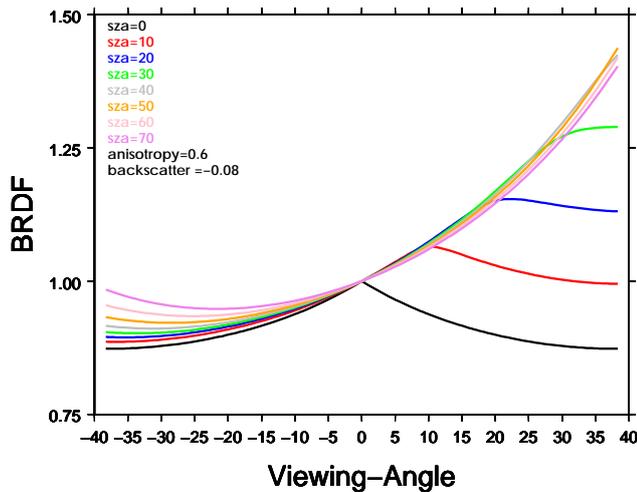
## 2 BRDF corrected MERIS aerosol optical depth retrievals

The basis of the presented PM<sub>10</sub> retrieval is spectral aerosol optical depth as retrieved from MERIS/Envisat data with the Bremen AERosol Algorithm (BAER) (von Hoyningen-Huene et al., 2003). The add-on for the retrieval of particulate matter is hereafter called Particulate Matter Bremen Aerosol Retrieval (PMBAER). This nadir viewing MERIS imager (Baudin et al., 1991) consists of fifteen spectral bands between 390 and 1040 nm. The spatial resolution of MERIS measurements is 1040 × 1200m, over land and at the coasts data with a resolution of 260 × 300m are also available. Seven channels for the retrieval over land are used currently (412.5 nm; 442.4 nm; 489.7 nm; 509.7 nm; 559.6 nm; 619.6 nm; 664.6 nm).

BAER basically subtracts the reflectance caused by Rayleigh scattering and surface reflection from the total reflectance; the result is assumed to be caused by aerosols. The algorithm derives the Rayleigh path reflectance using the radiative transfer model of Nakajima and Tanaka (1988). Surface pressure is obtained through a 30 arc seconds resolute digital elevation model (Row et al., 1995), and temperature is taken from ECMWF models. Experimental scattering parameters have been taken from the Lindenberg Aerosol Characterization Experiment 1998 (LACE-98) (Ansmann et al., 2002), phase functions (Fig. 3) and single-scattering albedo as derived through data from sun and sky radiometers and through the Coupled Inversion Radiative Transfer (CIRATRA) retrieval algorithm (von Hoyningen-Huene and Posse, 1997).

For the separation of the surface reflectance, a weighted mixing of bare soil and green vegetation and a normalized differential vegetation index (NDVI) at 670 and 865 nm is considered which is fitted to a smooth Ångström- $\alpha$  exponent. Spectral reflectance measurements from the Compact Airborne Spectral Imager (CASI) and extensions from the Changes in Arid Mediterranean Ecosystem on the Long term and Earth Observation (CAMELEO) database (Escadafal and Bohbot, 2001) are used for this purpose.

BAER finally uses look-up-tables to deduce the aerosol optical depth  $\tau(\lambda)$  from the top-of-atmosphere reflectance  $R(\lambda)$ , derived by a radiative transfer model of Nakajima and Tanaka (1988).



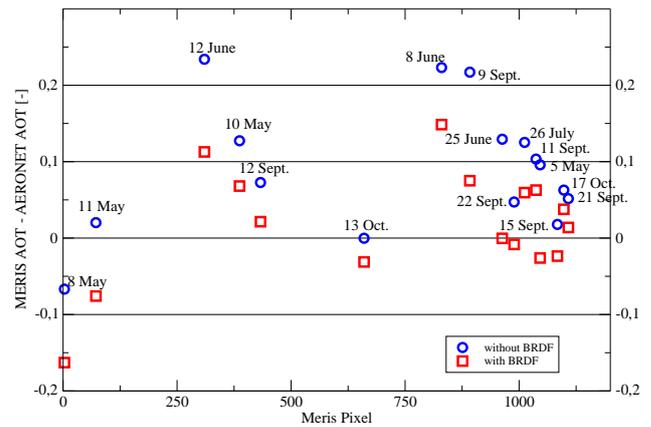
**Fig. 1.** BRDF as used for the retrieval of aerosol optical depth over Germany for different solar zenith angles (Sinyuk et al., 2006).

The agreement of BAER aerosol optical depth as retrieved from data from SeaWiFS, SCIAMACHY and MERIS satellites with AERONET and other satellites has already been shown in several studies (von Hoyningen-Huene et al., 2003; von Hoyningen-Huene et al., 2007; Kokhanovsky et al., 2007; Kokhanovsky and de Leeuw, 2009), and also in applications like observations of Russian forest fires (Lee et al., 2003) or over Korea (Lee et al., 2006). Measurements over water as well over desert ground have also been successfully performed (von Hoyningen-Huene et al., 2006; Dinter et al., 2009).

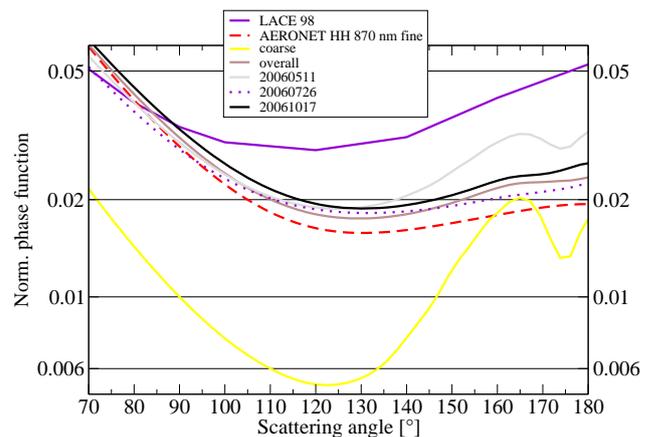
In order to consider the viewing angle range of the MERIS swath, a bidirectional surface reflectance distribution function (BRDF) has been implemented (see Fig. 1 with a fixed azimuth angle of 170°). The BRDF semi-empirical model was taken from Sinyuk et al. (2006), parameters for the anisotropy and for the backscatter have been found empirically through comparisons with AERONET data over Hamburg. This model has already been shown to work for different surfaces (Sinyuk et al., 2006) and was also used for MERIS/PMBAER retrievals over desert surfaces (Dinter et al., 2009).

The usage of a surface BRDF is essential and cannot be omitted; this is demonstrated by a first brief comparison of MERIS aerosol optical depth from a set of sixteen measurements over Germany compared to measurements from AERONET over Hamburg (Fig. 2). Mean bias was improved from 0.091 to 0.017 and standard deviation from 0.0841 to 0.075. Applied MERIS scenes will also be used later for the validation of PM<sub>10</sub>.

For this study, measurements at ten arbitrarily chosen cloud-free days over Hamburg AERONET stations in 2006 have been taken for comparisons of the AOD spectra (Fig. 4). Same days are used later for comparisons of the retrieved



**Fig. 2.** Differences between MERIS (with and without BRDF effects) and AERONET AOD at 440 nm over Hamburg for sixteen collocated measurements at cloud free days in 2006.

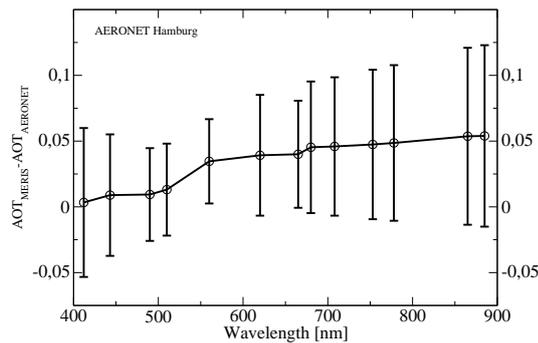


**Fig. 3.** Phase functions as derived from the LACE-98 experiment; AERONET Hamburg 870 nm for fine, coarse and overall aerosols.

PM<sub>10</sub> with measurements of the air quality surveillance stations in Hamburg. The comparisons of AOD reveal a standard deviation between 0.032 and 0.068, depending on the wavelength (see Table 1). For longer wavelengths, an increasing offset is observed.

### 3 PM<sub>10</sub> retrieval methodology

Let  $a$  be the radius of the single aerosol particle which is assumed to be spherical and  $Q_{\text{ext}}(a, \lambda, n)$  the dimensionless extinction efficiency which has been calculated through Mie theory (see also shape of  $Q_{\text{ext}}(a, \lambda, n)$  and the limitation related to the size parameter by Kokhanovsky et al. (2006)).  $n = n(\lambda)$  denotes the refraction index of the particle which



**Fig. 4.** Comparisons of ten collocated MERIS/PMBAER measurements with AERONET over Hamburg. Bars indicate the standard deviation. See Table 1 for exact values.

**Table 1.** Offsets and standard deviations for the comparisons of aerosol optical depth over Hamburg for ten cloud-free days in 2006 (see Fig. 4).

$\lambda$	Bias, Std.-Dev.	$\lambda$	Bias, Std.-dev.
412.5	0.003 $\pm$ 0.056	680	0.045 $\pm$ 0.052
442.4	0.009 $\pm$ 0.045	708	0.050 $\pm$ 0.056
489.7	0.009 $\pm$ 0.034	753	0.047 $\pm$ 0.056
509.7	0.013 $\pm$ 0.032	778	0.048 $\pm$ 0.059
559.6	0.035 $\pm$ 0.045	865	0.056 $\pm$ 0.067
619.6	0.039 $\pm$ 0.040	885	0.054 $\pm$ 0.068
664.6	0.040 $\pm$ 0.050		

is taken from the Optical Properties of Aerosols and Clouds (OPAC), water-soluble (WASO) database (Hess et al., 1998).

The extinction within the objected air mass column has to be integrated over the particle distribution  $\frac{df}{da}$  and the height  $z$  (absorption is neglected in these equations,  $\omega_0 = 1$ ),

$$\tau(\lambda) = N \int_0^{\text{TOA}} \int_0^{\infty} \pi a^2 Q_{\text{ext}}(a, \lambda, n) \frac{df(a, z)}{da} da dz, \quad (1)$$

where  $N$  is the number of particles in the observed air mass. Substitution through

$$\frac{dm}{da} = \frac{4\pi a^3}{3} \rho \frac{df}{da}, \quad (2)$$

where  $\rho$  denotes the humidity corrected density, leads finally to the wanted relationship between  $\tau$  and the mass load,

$$\tau(\lambda) = N \int_0^{\text{TOA}} \int_0^{\infty} \frac{3}{4\rho a} Q_{\text{ext}}(a, \lambda, n) \frac{dm(a, z)}{da} da dz. \quad (3)$$

Under consideration of a vertically homogeneously distributed aerosol concentration  $\frac{dm(a, z)}{da}$ , this relationship can be written as

$$\tau(\lambda) = \frac{MH \langle C_{\text{ext}}(\lambda) \rangle}{\rho \langle V \rangle}. \quad (4)$$

$M$  is the searched aerosol mass concentration,  $H$  denotes the aerosol layer height. In the retrieval, 90% of the aerosol is assumed to be within the boundary layer height. Latter one is routinely provided by the European Center for Medium-Range Weather Forecasts (ECMWF). This estimate is strictly speaking only valid for continental sites.

$$\langle V \rangle = \frac{4\pi}{3} \int_0^{\infty} a^3 f(a) da \quad (5)$$

is the average volume of the particles.

$$\langle C_{\text{ext}} \rangle = \pi \int_0^{\infty} a^2 Q_{\text{ext}} f(a) da \quad (6)$$

is the average extinction cross section and  $f(a)$  is the normalized log-normal distribution function which was chosen to be mono-modal for the retrieval of PM<sub>10</sub> concentration.

The derivation of the aerosol mass  $M$  can therefore be written as

$$M = \gamma(f(a), \lambda) \tau, \quad (7)$$

where

$$\gamma(f(a), \lambda) = \frac{\rho \langle V \rangle}{H \langle C_{\text{ext}} \rangle}. \quad (8)$$

$\gamma$  is not a constant but a function of wavelength  $\lambda$  and size distribution  $f(a)$ . Several groups already used Eq. (8) (e.g., Griggs, 1975; Fraser, 1976; Gasso and Hegg, 1997; Griggs, 1979; Fraser et al., 1984; Kaufman et al., 1990; Gassó and Hegg, 2003; Mishchenko et al., 2002; Levy et al., 2007b). For the presented retrieval of aerosol particles, a lognormal size distribution

$$f(a) = \frac{1}{\sigma a \sqrt{2\pi}} e^{-0.5\sigma^{-2} \ln^2(\frac{a}{\mu})} \quad (9)$$

is assumed, where  $\sigma$  is the half-width and  $\mu$  the mean particle radius which is correlated with the effective radius  $a_{\text{eff}}$  through the division of the volume by the surface integral

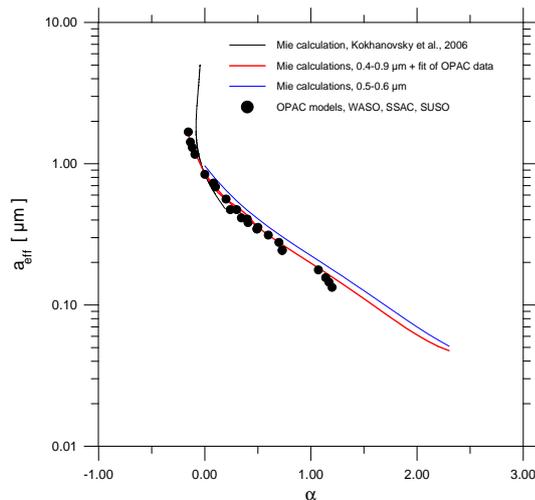
$$a_{\text{eff}} = \frac{\int_0^{\infty} a^3 f(a) da}{\int_0^{\infty} a^2 f(a) da}. \quad (10)$$

Equation 10 can be adequately parameterized by

$$a_{\text{eff}} = \mu \exp(-2.5\sigma^2) \quad (11)$$

where  $\sigma = 0.832$  and  $a$  is limited to less than 20  $\mu\text{m}$  (Kokhanovsky et al., 2006). To find this correlation, Kokhanovsky et al. (2006) assumed also a mono-modal log-normal distribution function with an overall integral of one.

A typical effective radius of 0.282  $\mu\text{m}$ , for instance, corresponds to a mean particle radius of 0.05  $\mu\text{m}$ . To expand or specify the retrieval of different aerosol radii, e.g. PM<sub>2.5</sub>, the size distribution function  $f(a)$  can be modified adequately. For this paper we used PM<sub>10</sub> data due to the fact that those measurements are more available from the national air quality stations in contrast to the PM<sub>2.5</sub>.



**Fig. 5.** Correlation between Ångström- $\alpha$  coefficient and effective radius as derived from Mie calculations and interpolated data from OPAC of water soluble particles.

The effective radius can now be derived through the respective Ångström- $\alpha$  exponent

$$\alpha = \frac{-\ln\left(\frac{\tau(\lambda)}{\tau(\lambda_0)}\right)}{\left(\frac{\lambda}{\lambda_0}\right)}. \quad (12)$$

The top-of-atmosphere reflectance at wavelength  $\lambda$  is derived from the MERIS radiances  $L(\lambda)$  by

$$R_{\lambda}^{\text{TOA}} = \frac{\pi L(\lambda) M_0}{E_0}. \quad (13)$$

$M_0$  is the air mass factor and  $E_0$  the top-of-atmosphere irradiance at wavelength  $\lambda$ . The air mass factor depends mainly on the geometry, but also on scattering which is impacted by meteorological parameter as temperature and pressure (see Sect. 4.2).

Figure 5 shows the correlation which is used to infer the effective radius from the Ångström- $\alpha$  exponent. For the derivation of this correlation, Mie calculations using Optical Properties of Aerosols and Clouds (OPAC) databases (Hess et al., 1998) have been used. The interpolated curve is described by the function

$$a_{\text{eff}} = 0.856 - 2.794\alpha + 9.699\alpha^2 - 18.157\alpha^3 + 11.792\alpha^4. \quad (14)$$

It is very difficult to obtain reliable effective radii if  $\alpha$  is less than 0.16, due to the large slope. In this case, the effective radius is set to 1.7  $\mu\text{m}$  in the retrieval. However, this assumption is also a restriction for the retrieval of coarse aerosol particles and leads to an underestimation of the mass load in general (see also discussion later in Sect. 6).

The multi-channel spectral information from MERIS allows one to infer not only accurate aerosol optical depths as derived in the last section. It also leads to more accurate effective radius than using only a few channels.

## 4 Retrieval corrections

### 4.1 Humidity correction

Due to condensation and evaporation effects, size, density, and shape, as well as the refractive index and the particle size distribution function, are all affected by ambient humidity. This again causes changes of the optical and radiative transfer properties, e.g., the Ångström- $\alpha$  coefficients (Schuster et al., 2006). The particles become larger and more spherical, and their density decreases with humidity. The Hänel (1976) model is commonly used to estimate this correlation between humidity, scattering coefficient, and particle radius. A parameterization of the humidity dependence of radius and scattering coefficient can be given by

$$a(h) = a_{\text{dry}} \cdot (1-h)^{-\epsilon} \quad (15)$$

and

$$\sigma(h) = \sigma_{\text{dry}} \cdot (1-h)^{-\gamma}, \quad (16)$$

where  $h$  is the relative humidity (0..1),  $a_{\text{dry}}$  the radius of the particle in dry state,  $\sigma$  the light scattering coefficient (dry state  $\sigma_{\text{dry}}$ ), and  $\epsilon$  and  $\gamma$  are size growth parameters (see Hänel, 1971) for explicit values). Both equations can be combined to

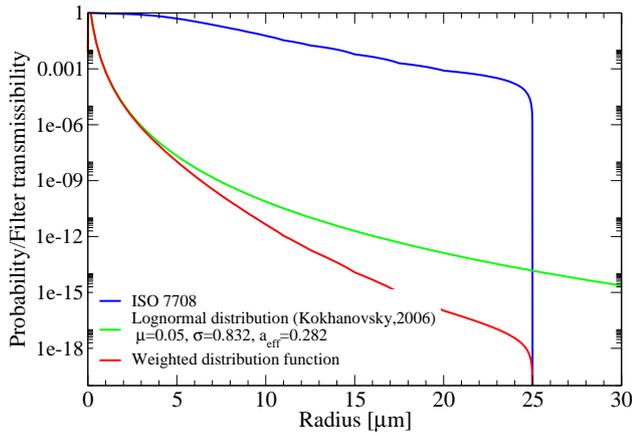
$$\frac{a(h)}{a_{\text{dry}}} = \left(\frac{\sigma(h)}{\sigma_{\text{dry}}}\right)^{\frac{\gamma}{\epsilon}}. \quad (17)$$

Although these correlations are frequently used, they do not consider hysteresis effects; depending on the direction of the changing ambient conditions, the correlation is different. From measurements at cloud-free conditions, a downward motion of the particles from the cooler to the warmer atmospheric layers is expected for MERIS observations during the morning. Thus, the ambient humidity is supposed to change in a decreasing way, i.e., the air becomes drier.

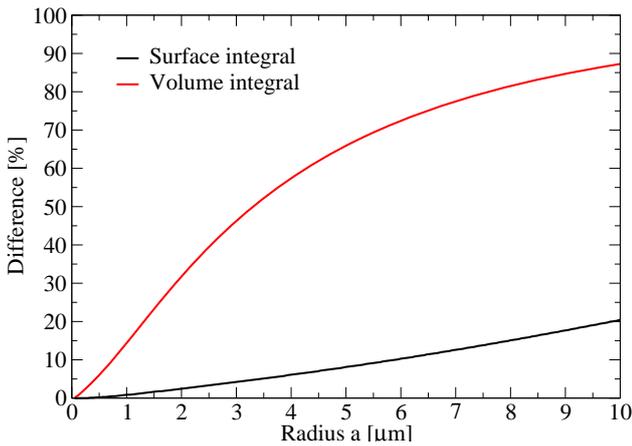
There are many discussions about these hysteresis effects, but, for instance, for increasing and decreasing humidity, a different correlation has been measured for sites over Paris, (Randriamiarisoa et al., 2006). The findings for the measurements with increasing humidity agree with the Hänel model. For decreasing humidity, larger particle sizes have been found. Figure 8 shows the particle growth as derived following the Hänel model for average aerosols ( $\epsilon = 0.25$ ), maritime and dust aerosols ( $\epsilon = 0.18$ ), and for urban aerosols ( $\epsilon = 0.285$ ), measurements from Randriamiarisoa et al. (2006) and corresponding regression curves.

For the humidity range below  $h = 0.4$  and above 0.9, the Hänel model is used for the retrieval with  $\epsilon = 0.25$ . For humidity between 0.4 and 0.9 the parameterization

$$\frac{a(h)}{a_{\text{dry}}} = 2.0138 + 0.94(1-h) - 4.331(1-h)^2 \quad (18)$$



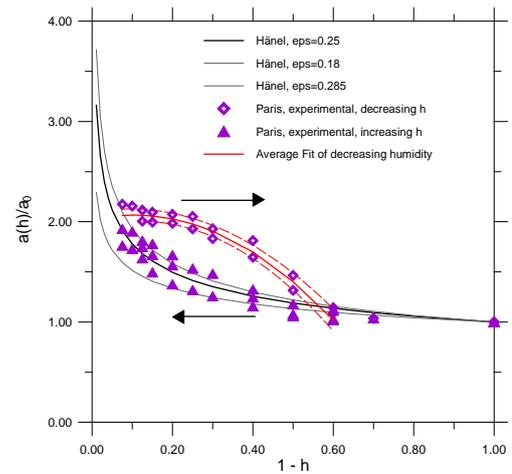
**Fig. 6.** Filter functions for the conversion between the different definitions of particulate matter mass load. The red curve denotes the weighted distribution function which is a folding of the log-normal distribution with the ISO 7708 weighting function.



**Fig. 7.** Effects on the derivation of aerosol mass due to adjustment of different PM<sub>10</sub> definitions; volume integral is equivalent to aerosol mass).

is used. For instance, assuming a variability of humidity  $h$  of 0.3, this would cause a two times larger radius of the particle and therefore an eight times larger mass load if the density is assumed to stay constant.

According to the changing volume of the particle, the particle density was also corrected in the retrieval. The humidity correction is strictly valid only for the particles at the surface. Currently, a homogenous distribution of humidity is assumed for the particulate matter retrieval. Although there are many indications that this is valid within the aerosol layer, the humidity is supposed to decrease with altitude and therefore, an overestimation can be caused due to this simplification.



**Fig. 8.** Change of particle size with humidity as derived by Hänel (1976), and measurements over Paris (see Randriamiarisoa et al. (2006) during times of increasing and decreasing humidity). For the PM<sub>10</sub> retrieval, the parameterization as indicated by the solid red line, is used.

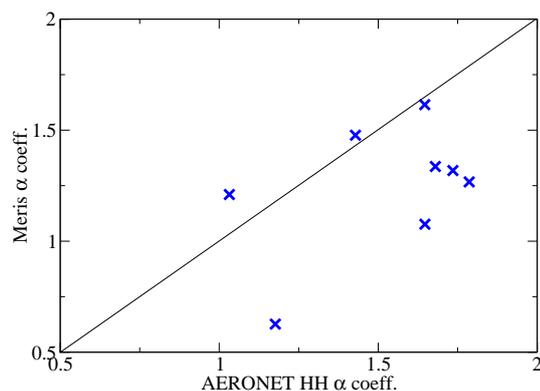
## 4.2 Temperature correction

The temperature affects the Rayleigh correction and the derivation of the air mass factor. If temperature increases, Rayleigh scattering becomes larger; the contribution to the reflectance from the aerosols is therefore smaller, i.e., the aerosol optical depth. In contrast to that, the effect on the air mass factor increases the aerosol optical depth, at least at small aerosol optical depths. At larger aerosol optical depths, the contribution of the aerosols enhances, and with increasing temperatures an increase of aerosol optical depth is also expected. Because of the linear dependence of the air mass factor on these parameters and their relatively small percental changes (say about 30 Kelvin relative to 285 and about 20 mbar relative to 1013), both parameters have a smaller impact on the derivation of the aerosol optical thickness compared to the impact of humidity. Near real time ECMWF model temperatures are now routinely inserted into the retrieval.

## 4.3 Adaptation of PM<sub>10</sub> definition

Due to the different definition of PM<sub>x</sub> by the national air quality measurement devices which follow the ISO 7708 standard, an adjustment towards the physical measure definition of the ground based measurements has been made. The national air quality measurement devices assume not a sharp cutoff but a smooth filter function over the 10 μm border (Fig. 6). In the MERIS retrievals, a cut-off for the integration of the aerosol mass is assumed to be at 20 μm.

The effect of this weighting by the ISO 7708 filter function is negligible for small radii (see Fig. 7), but is large for larger particles (about 65% for particles with a size of 5 μm).



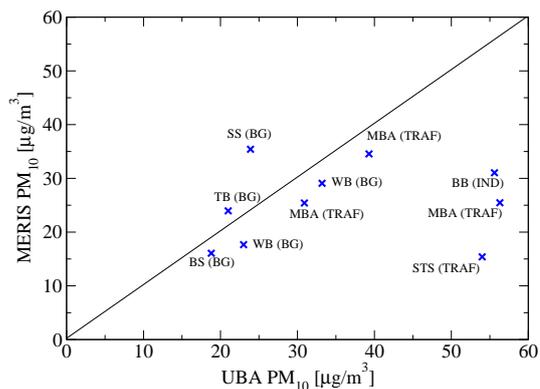
**Fig. 9.** Comparison of Ångström- $\alpha$  coefficient at 440 and 675 nm from MERIS PMBAER and from AERONET L2 in Hamburg.

This means that the PM<sub>10</sub> mass load as retrieved by MERIS/PMBAER is generally larger than those of the ground based measurements which are heated and dried before weighting - by the way. An adequate conversion is also done in PMBAER.

#### 4.4 Routine inclusion of meteorological data

Particulate matter mass concentration retrievals require the inclusion of meteorological parameter (Uhlig and von Hoyningen-Huene, 1993; Koelemeijer et al., 2006). This is reasonable because a change of meteorological parameter like the planetary boundary layer height or the humidity of the air can cause a massive change of the mass concentration whereas aerosol optical depth is affected by these parameters. Also, it is important to know the vertical distribution of the aerosol mass concentration profile when someone aims to infer the PM<sub>x</sub> concentration above the surface.

In the retrieval, 90% of the aerosol layer is assumed to be within the boundary layer height. This correlation was confirmed through CALIPSO LIDAR measurements, even for strong biomass burning injections (Labonne et al., 2007). Labonne et al. (2007) also showed that this assumption cannot be made in areas of strong changing boundary layer height, e.g., at the coast. However, for most of the continental areas, the boundary layer height is linearly correlated with the aerosol optical depth through Eq. (4). For usage in the PM<sub>10</sub> retrieval, the boundary layer height was interpolated to the MERIS local over-flight time of about 10:00 (for the exact Envisat local over-flight times see (e.g., Rohen et al., 2008)). The diurnal change of the boundary layer height increases in the morning up to about 300 m per hour until the maximum is reached about noon (Baars, 2007). Following Eq. (4), such a change would imply a change of the PM<sub>10</sub> by a factor sometimes greater than approximately three. For instance, the boundary layer height varied between 1000 and 2500 m on 12 June 2006 over Germany (see Fig. 12).



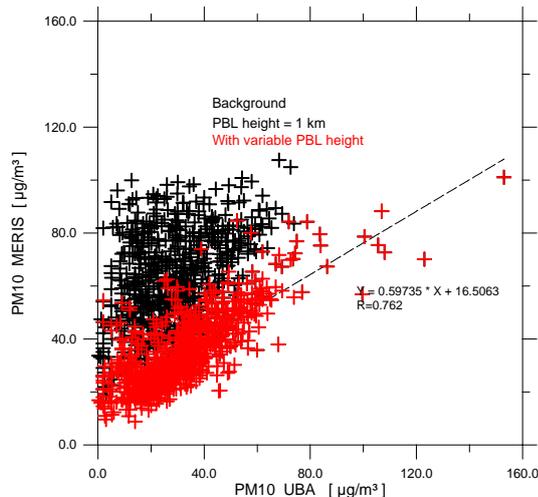
**Fig. 10.** Comparisons of MERIS/PMBAER and national air quality measurements PM<sub>10</sub> in Hamburg. The annotation denotes the gauging station and the site location (BG = background, IND = industry, TRAF = traffic).

## 5 Results

Figure 9 shows the comparison of Ångström- $\alpha$  exponents as retrieved from MERIS/PMBAER and from the collocated AERONET stations in Hamburg for ten cloud-free days in 2005 and 2006. They have been deduced from the aerosol optical depth between 440 and 675 nm. The shown exponents are equivalent to effective radii between 0.03 and 0.2  $\mu\text{m}$  (see Fig. 9). The AERONET Ångström- $\alpha$  coefficients are marginally higher, see also Che et al. (2008) who also found increased values of AERONET.

Figure 10 shows comparisons of PM<sub>10</sub> from MERIS/PMBAER and from the national air quality measurement stations in Hamburg for the same days. The location is indicated – background, traffic, or industry. In general, a good agreement between both measurements can be seen with the exception of three outliers which are reasoned by their specific location at traffic and industry sites. Omitting those three outliers, a correlation coefficient of 0.64 is reached. This corresponds to a bias of  $-2.6\%$ , and a standard deviation of 25.3% w.r.t. UBA measurements and a bias of  $-18.9\%$  and standard deviations of 33.9% over all comparisons. With a spatial resolution of 1200 m, MERIS is not able to look into a street canyon and industry sites. The correlation will be improved by using the full resolution MERIS data. At all background sites, MERIS observations are close to the air quality measurements. Aerosol retrievals over cities are known to be difficult because of the unequal and varying surface and because of different sources and therefore kind of the aerosols; additionally, pollution sources like traffic or industry sites are relatively small and not widely distributed.

The next Fig. 11 shows a comparison of results of PMBAER with air quality measurements in rural sites in Germany on sixteen days in 2005 and 2006. The corresponding correlation coefficient is 0.75, standard deviation is 17.9 with



**Fig. 11.** Comparisons of PM<sub>10</sub> as retrieved from 250 MERIS/PMBAER and national air quality measurements over rural sites in Germany. Results with and without inclusion of ECMWF planetary boundary layer heights (PBL) are shown here.

a bias of 12.9%. Additionally, Fig. 11 shows the effect of the synergy of ECMWF boundary layer height data; without considering this meteorological parameter, no good correlation can be achieved.

Figure 12 shows the aerosol optical depth, effective radii, and inferred PM<sub>10</sub> concentration from MERIS measurements over Germany on 12 June 2006, a sunny and dry day. The aerosol optical depth on that day exhibits reasonable values, and the transition between land and coast is also reasonable, except at the Wadden Sea where the surface model does obviously not work properly. Hot-spots can be observed clearly, for instance the cities Hamburg or Munich or even smaller cities. Cloud patterns in the northeast are easily recognizable although they are hardly visible in the RGB pictures. Artifacts can also be identified at mountains, e.g., at the Alps, where retrieved aerosol optical depth is too large (air there should be cleaner in general); this is due to inaccurate Rayleigh correction and must be investigated in the future.

Comparisons of the particulate matter concentrations will be improved if higher resolution satellite data are used. The comparisons over Hamburg showed that basically good results can be achieved, with the exception of street canyons and singular air pollution, e.g., singular industry sites.

## 6 Critical discussion of limitations of methodology and comparisons

The main uncertainties of the retrieval are

- (a) the errors propagated due to the underlying AOD; the quality of the underlying AOD depends on the wave-

length. Roughly estimated, the precision of the satellite AOD retrievals can be limited to about 20% at best (Kokhanovsky et al., 2007). This estimation includes the possible impreciseness as introduced by large deviations of the phase functions;

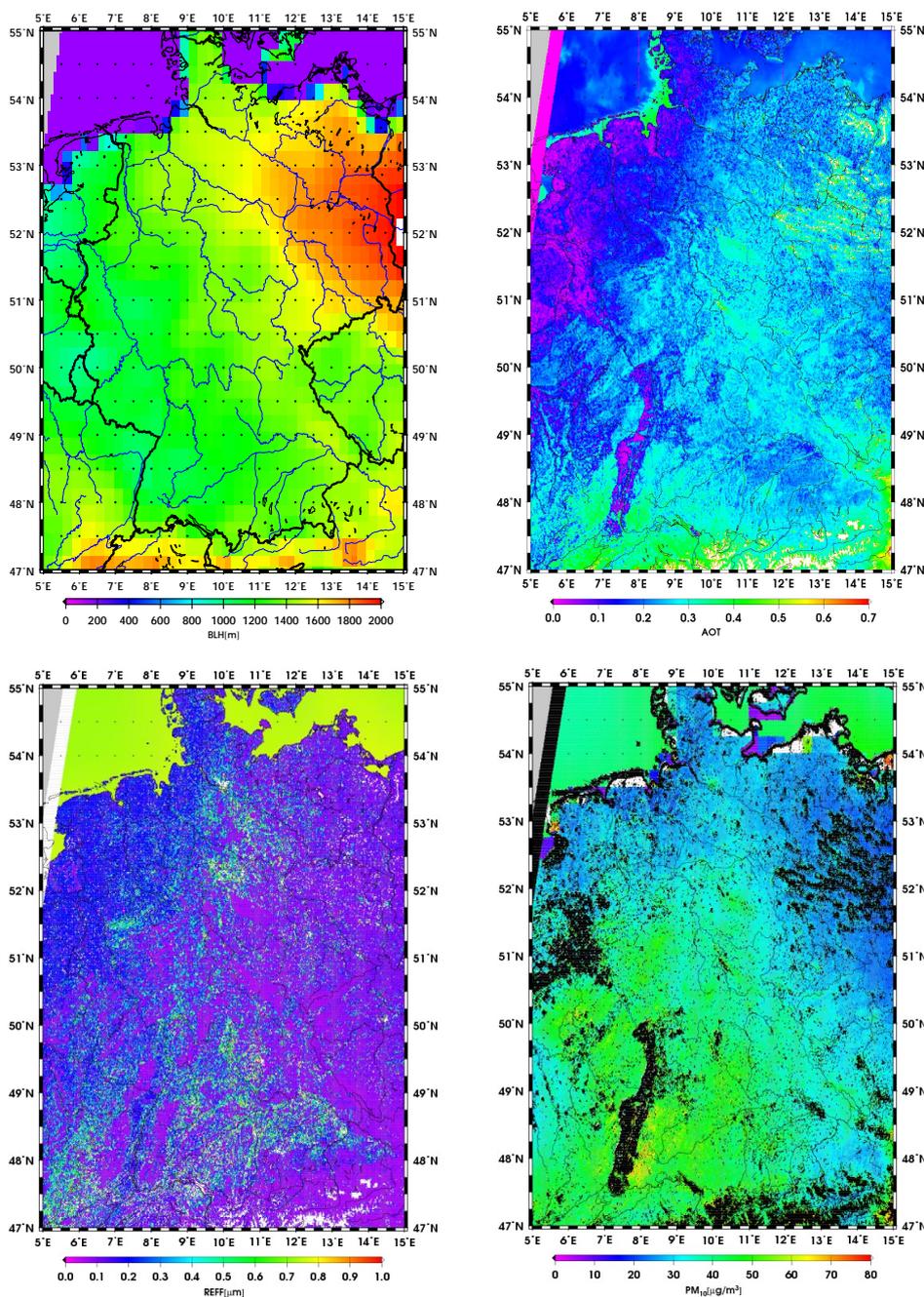
- (b) the sensitive logarithmic relationship between Ångström- $\alpha$  and the effective radius;
- (c) the simplification of the aerosol type – expressed quantitatively by the mono-modal size distribution function;
- (d) the imprecision introduced by meteorological parameters, in particular the boundary layer height;
- (e) the assumption of a vertical profile of homogeneously distributed aerosols.

Additionally, we have to consider also the different type of underlying measurements: retrieval products are based on air-borne measurements of humid and cold particles in a 800 km air column by optical remote sensing measurements whereas the ground devices weight heated, dried, and filtered surface aerosols at a point location. Satellite measurements provide a resolution of about 300 m at best. The distance between the respective gauging stations are of several kilometers and at regions with highly variable aerosol concentrations.

The reason for the worse agreement over Hamburg is firstly the insufficient spatial resolution of the MERIS measurements. Additionally, the aerosol types over cities differ strongly from those over rural sites. A bi-modal size distribution function has to be implemented here at least, or another aerosol classification model.

In view of this, the yielded standard deviations of 17.9% over rural sites as well as of 34% over Hamburg with relatively small biases are reasonable and correspond to about actual retrievals using other methodologies (Hoff and Christopher, 2009): we aimed a standard deviation of 25.31% with a bias of  $-2.59\%$  while Hoff and Christopher (2009) estimated the precision of the most retrievals to about 30%.

The retrieval methodology itself becomes imprecise for particle measurements of particles with radii larger than about  $1.7\mu\text{m}$ . The limitation for coarser particles explains the underestimation of  $-18.9\%$  over rural sites of Germany as seen from Fig. 11. The relatively good agreements are thanks to the fact that most of the particles as measured at the selected scenes are of sizes below  $0.6\mu\text{m}$  (see Fig. 12). At sites with more coarse particles like over deserts, this retrieval is not able to yield good results.



**Fig. 12.** ECMWF@ boundary layer height for 12 June 2006 over Germany, interpolated to 10:00 a.m., aerosol optical depth at 440 nm, effective radii and PM<sub>10</sub> mass concentration.

## 7 Conclusions

In this paper we presented results of a PM<sub>10</sub> retrieval over Germany. Masses have been derived on basis of the effective radius from the Ångström- $\alpha$  exponent. The retrieval benefits from the spectral information of seven MERIS channels which are used to provide a smooth spectral slope.

In spite of the large possible uncertainties, comparisons with measurements from national air quality gauging stations over Germany show agreement with a standard deviation of 17.9% (bias 12.9%) over rural sites of Germany and of 34.0% (bias -18.9%) over the city of Hamburg. Comparisons over Hamburg provide worse agreements – reasoned by the spatial resolution or the insufficient classification of the aerosol

types. Results are improved by the introduced corrections of the AOD by a proper bidirectional surface reflectance distribution function, or corrections of the physical and optical properties by humidity effects. The retrieval is limited by the steep slope of the Ångström- $\alpha$  coefficient for particles larger than 1.7  $\mu\text{m}$ . For sites in Germany, we show that this limitation has no large impact on the preciseness of the retrieval. Main inaccuracies of the methodology are the sensible relationship between Ångström- $\alpha$  and the effective radius as well as the relatively simple mono-modal size distribution function for deriving the mass load around the effective radii.

This shows that the retrieval can be used for PM<sub>10</sub> and in particular for PM<sub>2.5</sub> measurements over similar sites as used in this study for the observation of large exposure events and their movement. For air quality surveillance over cities, the methodology is in general very sensitive to the small particles but suffers on the bad resolution of the measurements and proper aerosol classification.

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