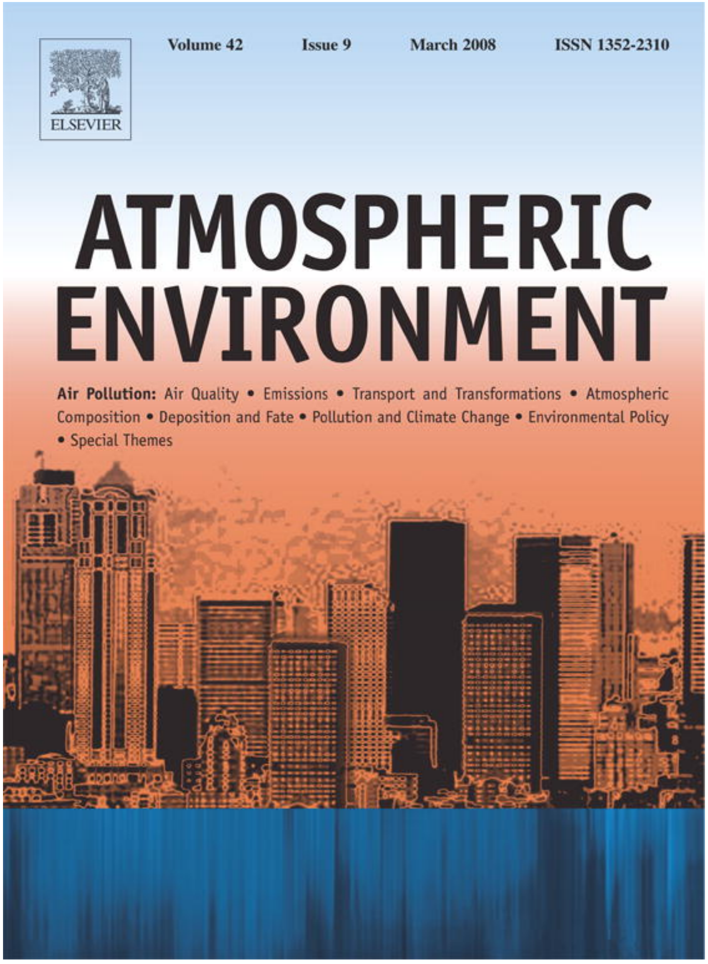
Volume 42
Issue 9
March 2008
ISSN 1352-2310
ELSEVIER
ATMOSPHERIC ENVIRONMENT
Air Pollution: Air Quality • Emissions • Transport and Transformations • Atmospheric Composition • Deposition and Fate • Pollution and Climate Change • Environmental Policy • Special Themes

Available online at www.sciencedirect.com
ScienceDirect
ELSEVIER
Atmospheric Environment 42 (2008) 2187–2197
ATMOSPHERIC ENVIRONMENT
www.elsevier.com/locate/atmosenv

Evaluation of MODIS aerosol optical thickness over Europe using sun photometer observations

M. Schaap[a,*], R.M.A. Timmermans[a], R.B.A. Koelemeijer[b], G. de Leeuw[a], P.J.H. Builtjes[a]

[a]*TNO, Business Unit Environment, Health and Safety, P.O. Box 342, 7300 AH Apeldoorn, The Netherlands*
[b]*The Netherlands Environmental Agency (MNP), P.O. Box 303, 3720 AH Bilthoven, The Netherlands*

Received 20 March 2007; received in revised form 23 November 2007; accepted 27 November 2007

Abstract

Satellite retrieved aerosol optical thickness (AOT) may be useful to improve the insight in PM distributions in Europe in combination with models and ground-based measurements. To use AOT in mapping or assimilation experiments, it requires well-validated satellite data. We have compared the AOT retrieved by MODIS (collection 4) to sun photometer data from the AERONET network in Europe and found a good temporal correlation between MODIS and AERONET. However, we also found a large positive bias of about 50% in the MODIS AOT data, which is in accordance with earlier findings. We highlight the strong seasonal signature in the overestimation of AOT by MODIS with a maximum during summer. After correction for the bias, the accuracy of MODIS AOT retrievals agrees with reported uncertainties and the residuals show a normal distribution. We have introduced a simple method for the evaluation of the possible extent of cloud contamination and hypothesise that on average, up to one-third of the MODIS retrievals may be cloud contaminated. For some stations in central Europe, this percentage was found to be larger than $>50\%$. The consequences of a bias between satellite and in situ data for their use in the mapping of aerosol levels are discussed.

Keywords: Aerosol optical thickness; Europe; Remote sensing; Verification

1. Introduction

Exposure to particles ($PM_{2.5}$) has been associated with adverse health effects and particles are believed to be the most important air pollutant responsible for loss of human health. Short-term exposure to PM_x has frequently been associated with increased human morbidity and mortality (e.g., Brunekreef and Holgate, 2002). Effects of long-term exposure to PM are much more uncertain than the short-term effects, but are believed to have a much greater effect on health loss (Dockery et al., 1993; Pope et al., 1995). Furthermore, fine particulate matter or aerosols play a key role in changing the earth's radiation budget (Charlson et al., 1992; Hansen and Sato, 2001). The combined direct and indirect aerosol effect has masked the real climate sensitivity towards an increase in greenhouse gases to an unknown extent (Anderson et al., 2003). For the assessment of climate forcing by anthropogenic

*Corresponding author. Tel.: +31 55 5493385; fax: +31 55 5493252.
E-mail address: martijn.schaap@tno.nl (M. Schaap).

1352-2310/$ - see front matter © 2007 Elsevier Ltd. All rights reserved.
doi:10.1016/j.atmosenv.2007.11.044

aerosols, as well as for the assessment of the exposure to $PM_{2.5}$, the determination of the aerosol mass and its composition is mandatory. Traditionally, in situ observations are used to derive information on the large-scale features of air pollutants. In case of particulate matter, this approach is hampered by the difficulties in the sampling techniques and the highly variable concentrations in space and time. Satellite remote sensing may be a cost-effective method to monitor the highly variable aerosol fields on regional scales.

Satellite measurements provide full spatial coverage and are—in principle—consistent for the whole European region. However, they are less precise than in situ observations. This suggests that satellite measurements may be useful to improve the insight in PM distributions in Europe in combination with models and ground-based measurements. Various studies have reported good correlations between satellite derived aerosol optical thickness (AOT) and $PM_{2.5}$ surface concentration measurements in parts of the USA (Wang and Christopher, 2003; Hutchison, 2003). In general, promising correlations are found between 1-month time-series of AOT and $PM_{2.5}$ for many stations in the Eastern and Midwest United State. Other stations, however, particularly in the Western United State, show hardly any correlation (Engel-Cox et al., 2004). Variations in local meteorological conditions, occurrence of multiple aerosol layers, and variations in aerosol chemical composition likely play an important role in determining the strengths of such correlations. Koelemeijer et al. (2006b) showed that the correlation between PM and AOT is improved when the AOT is divided by the boundary layer height and, to a lesser extent, when it is corrected for growth of aerosols with relative humidity. Using these improvements, the average correlations are 0.5 (PM_{10}) and 0.6 ($PM_{2.5}$), averaged over rural and (sub) urban background stations in Europe. For Europe, a PM_{10} distribution has been derived using geostatistical techniques combining in situ observations, modelled distributions and MODerate-resolution Imaging Spectroradiometer (MODIS) satellite data (van de Kassteele et al., 2006). The authors showed that the use of both model and remote sensed data improved the quality of the estimated PM_{10} distribution. Furthermore, the advent of satellite observations has also led to a development of data assimilation schemes that assimilate AOT directly into models (e.g., Builtjes et al., 2001; Collins et al., 2001; Koelemeijer et al., 2006a).

The above-mentioned applications depend critically on the quality of the remote sensing data. At present, MODIS satellite data (Kaufman et al., 1997; Remer et al., 2005) appear to be best suited for these studies as they are readily accessible and have a very good coverage. However, some indications for an overestimation of retrieved AOT over land have been reported (Ichoku et al., 2002; Remer et al., 2005). A possible bias in the data has to be recognised for applications in mapping or assimilation experiments. In contrast to previous studies, we present a dedicated validation of the MODIS satellite data for Europe for use in PM mapping/assimilation experiments. The validated MODIS data have been used in an assimilation experiment, which is described by Koelemeijer et al. (2006a).

2. Data sources

2.1. Aerosol optical thickness from MODIS

MODIS was launched onboard the EOS-Terra satellite in December 1999. In May 2002, a second MODIS instrument was launched onboard EOS-Aqua. The MODIS instruments measure sunlight reflected by the Earth's atmosphere and surface as well as emitted thermal radiation at 36 wavelengths. At least two observations of any place in Europe are obtained per day during daylight hours because the Terra and Aqua satellites cross Europe near 10:30 and 13:30 h local solar time, respectively. However, retrieval of AOT is not possible under cloudy conditions. The AOT algorithms for application over land and sea surfaces are mutually independent. This is because the radiative properties of water and land are very different. The retrieval is more accurate over ocean than over land because the reflection by water is relatively low outside the region of direct sun glint, algae blooms and suspended matter, and can be computed accurately from the sea surface wind field.

The MODIS retrieval (v4.2) of the AOT over land employs primarily three spectral channels centred at 0.47, 0.66, and 2.1 μm. AOT is derived at 0.47 and 0.66 μm, and interpolated to 0.55 μm. The AOT is only retrieved for cloud-free pixels in a 20×20 pixel area at 500 m resolution and reported at $10 \times 10\,km^2$ resolution. Only when >12 pixels are classified as cloud free is a retrieval attempted. The AOT is retrieved over surfaces that are not highly reflective (hence snow or ice covered surfaces and deserts are excluded). The algorithms are described in Kaufman and Tanré (1998), and updates since then are described in Remer et al. (2005). The AOT data obtained from these algorithms have been used in numerous studies directed to (among others) the direct aerosol forcing (see Yu et al., 2006 for a review), aerosol–cloud interactions (e.g., Koren et al., 2005; Wen et al., 2007), source characterisation (Dubovik et al., 2004; Koelemeijer et al., 2006a), and the assessment of Air Quality (e.g., Al-Saadi et al., 2005; van Donkelaar et al., 2006).

We have used the operational data (v4.2) for 2003 obtained from the MODIS data centre in October 2005. The collection 4 data have previously been validated on a global scale against AOT measurements from the ground-based Aerosol Robotic Network (AERONET). Validation showed that the retrieved AOT is generally within the pre-specified accuracy of $0.05 \pm 0.20 \times$ AOT over land and $0.03 \pm 0.05 \times$ AOT over ocean (1σ level, for individual retrievals). Exceptions are found in situations with possible cloud contamination, over surfaces with sub pixel surface water such as coastal areas and over surfaces with sub pixel snow cover (Chu et al., 2002; Ichoku et al., 2005).

2.2. AERONET data selection

For the validation of the MODIS data, we have used the data from the AERONET (Holben et al., 1998). AERONET consists of CIMEL automatic sun-sky scanning spectral radiometers. The network provides globally distributed observations of AOT. Here, we have used the 36 stations within the European domain that were operational in 2003. The stations used in this study are listed in Table 1 and shown in Fig. 1.

We have determined the MODIS AOT values at the locations of the AERONET sites. We have used the (average of the) AERONET observations in between the Aqua and Terra overpasses (between 11:00 and 13:00 h) for the validation. Tests using larger windows showed only a small sensitivity to the exact window size and confirmed our findings reported below. The latter can also be deduced from the fact that the Aqua (afternoon) observations only yield slightly higher AOT values than the Terra (morning) observations, as was also noted by Ichoku et al. (2005). These differences are much smaller than the differences between AERONET and MODIS AOT presented here.

3. MODIS AOT distribution

Yearly average maps are calculated from the monthly average maps, using only those months for which a monthly average is available. In winter months, no data are available for latitudes $>50°$N, because the solar elevation is too low to allow retrieval of AOT. In addition, snow cover hampers the AOT retrieval in the Alps and Central Europe during several (winter) months. Hence, 'yearly' averages for these regions are in fact averages excluding 1 or more months during the winter season. Our analysis is based on both MODIS-Terra (morning) and MODIS-Aqua (afternoon) observations.

The yearly average AOT (at 0.55 μm) in 2003 is shown in Fig. 2. In Europe, high AOT is retrieved over densely populated and industrialised regions and over large areas in Central Europe. Particularly high AOT values are found for the Benelux countries, the Ruhr area, the Po-valley, Northern and Eastern Germany, and Poland. Several major cities like Athens, Paris, and Rome can be recognised. Also the Rhone-valley in France and the Danube River in Central Europe are clearly distinguishable. Low values are observed in Scandinavia and over mountain areas. Furthermore, high AOT values are also observed over semi-arid rural areas in Southern Europe (particularly in Spain and Turkey).

Although the features described above appear to be realistic, a number of issues arise when these distributions are examined in more detail. A discontinuity in retrieved AOT is observed going from ocean to land with lower values over the ocean. Although higher AOT is expected over land (as source areas are located over land), a sudden change at the land–sea border is unlikely to be realistic realising that at least in north-western Europe, fair weather conditions associated with successful retrievals are mostly characterised by continental (offshore) flow conditions. Although high concentrations of sea spray aerosol produced in the surf zone (De Leeuw et al., 2000), these sudden changes are more likely due to the application of different algorithms and the inclusion of a fraction of either land or sea surfaces in the sea or land algorithms, respectively. Along coastal areas like the Netherlands, sometimes unrealistically high AOT values are observed. Suspended sediments in shallow water may give rise to high AOT retrievals (Robles Gonzalez et al., 2000; Chu et al., 2002;

Table 1
Comparison between annual average AOT from MODIS and AERONET stations over Europe for 2003. The correlation coefficient (*R*) between MODIS and AERONET is also listed. Stations are listed by country in which they are located

Country	Station	Observation		Correlation
		AERONET	MODIS	AERONET–MODIS
Algeria	Blida	0.09	0.39	0.42
Belarus	Minsk	0.16	0.27	0.70
Belgium	Oostende	0.25	0.37	0.80
Estonia	Toravere	0.14	0.23	0.26
France	Avignon	0.19	0.32	0.80
France	Bordeaux	0.11	0.28	0.89
France	Dunkerque	0.18	0.34	0.74
France	Fontainebleau	0.24	0.26	0.80
France	Lille	0.20	0.39	0.88
France	Rossfeld	0.30	0.29	0.63
France	Toulouse	0.18	0.35	0.80
France	Palaiseau	0.23	0.29	0.78
Germany	Hamburg	0.17	0.34	0.81
Germany	Helgoland	0.17	0.22	0.89
Germany	IFT-Leipzig	0.21	0.42	0.75
Germany	Munich_Maisach	0.20	0.28	0.70
Greece	Forth Crete	0.18	0.39	0.51
Italy	Etna	0.23	0.33	0.83
Italy	IMC_Oristano	0.22	0.32	0.82
Italy	ISDGM_CNR	0.26	0.51	0.81
Italy	Ispra	0.32	0.28	0.68
Italy	Lampedusa	0.21	0.24	0.91
Italy	Lecce_University	0.20	0.29	0.76
Italy	Rome_Tor_Vergata	0.22	0.33	0.77
Italy	Venise	0.24	0.31	0.62
Moldova	Moldova	0.22	0.23	0.84
Netherlands	The_Hague	0.24	0.33	0.91
Poland	Belsk	0.20	0.31	0.01
Portugal	Cabo_da_Roca	0.08	0.31	0.89
Portugal	Evora	0.14	0.27	0.83
Russia	Moscow_MSU_MO	0.18	0.34	0.19
Spain	El_Arenosillo	0.14	0.29	0.81
Spain	Palencia	0.14	0.31	0.59
Sweden	Gotland	0.13	0.23	0.77
Switzerland	Laegeren	0.13	0.26	0.49
Turkey	IMS-METU-ERDEMLI	0.23	0.33	0.70

Ichoku et al., 2005). Also, a discontinuity in AOT can be observed near the 25°E meridian, which coincides with a different assumption on aerosol type in the MODIS algorithm. Apparently, this gives rise to a systematic difference in the retrieved AOT.

4. Results and preliminary discussion

Below we present the results of the comparison between MODIS and AERONET AOT focusing on the retrieved AOT values, their differences and correlation. Furthermore, we have used a score analysis to assess the possible extent of cloud contamination.

4.1. Annual averages

Table 1 lists the annual average AOT values of MODIS and AERONET for the 36 individual stations. It is evident that MODIS overestimates the AOT values measured by AERONET at almost all stations. The difference is variable for the different stations. On average, the annual mean AOT observed by MODIS across all stations is 0.30, while that observed at AERONET stations is 0.20, providing a mean annual difference of 0.10 across all stations. Relatively, this means a significant overestimation of 50%.

We have also considered the spatial correlation of the annual average optical thicknesses. This is particularly important, as the spatial gradients of (yearly) average AOT play an important role if the AOT is used for improvement of mapping of PM levels. It was found that the spatial correlation based on annual mean co-located values between AERONET AOT and MODIS AOT is 0.64.

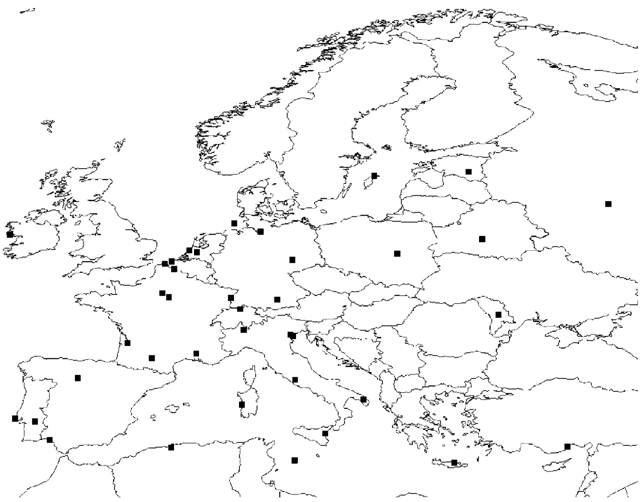

Fig. 1. The locations of the AERONET stations in Europe.

4.2. Temporal correlation

The temporal correlations between MODIS and AERONET AOT at the AERONET stations are also listed in Table 1. The time-series of MODIS and AERONET AOT show a good correlation of $R = 0.72$, averaged over all stations. The median of the correlation coefficients is $R = 0.77$. Poor correlation coefficients are found for stations with sparse data (e.g., Belsk, Poland). Stations with <15 days with simultaneous measurements have therefore been removed from the analysis.

In general, MODIS AOT exhibits a similar seasonal variation to that observed at AERONET stations. Examples are given in Fig. 3a–c for Avignon (France), ISDGM-CMR (Italy), and Lampedusa (Italy). Generally high AOT values are being observed in summer and low AOT values in winter. The MODIS AOT, however, exhibits a stronger seasonal trend than AERONET AOT over land. The difference between MODIS and AERONET AOT is summarised for all sites in Fig. 4. In winter and summer, average residuals for AOT are

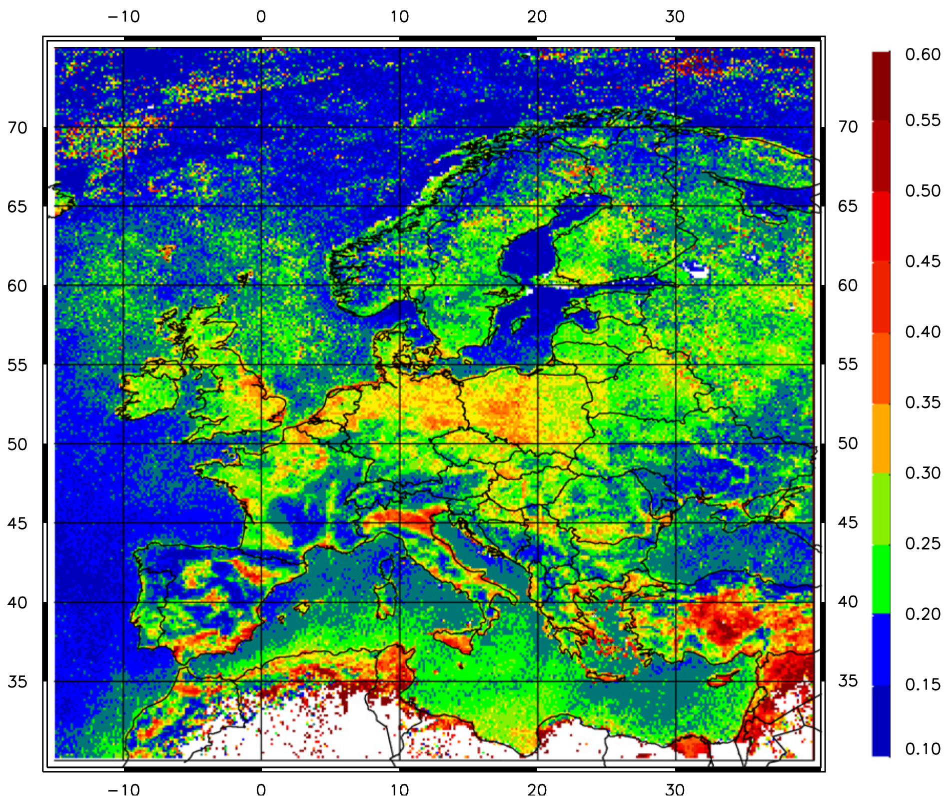

Fig. 2. Annual average composite map of MODIS AOT.

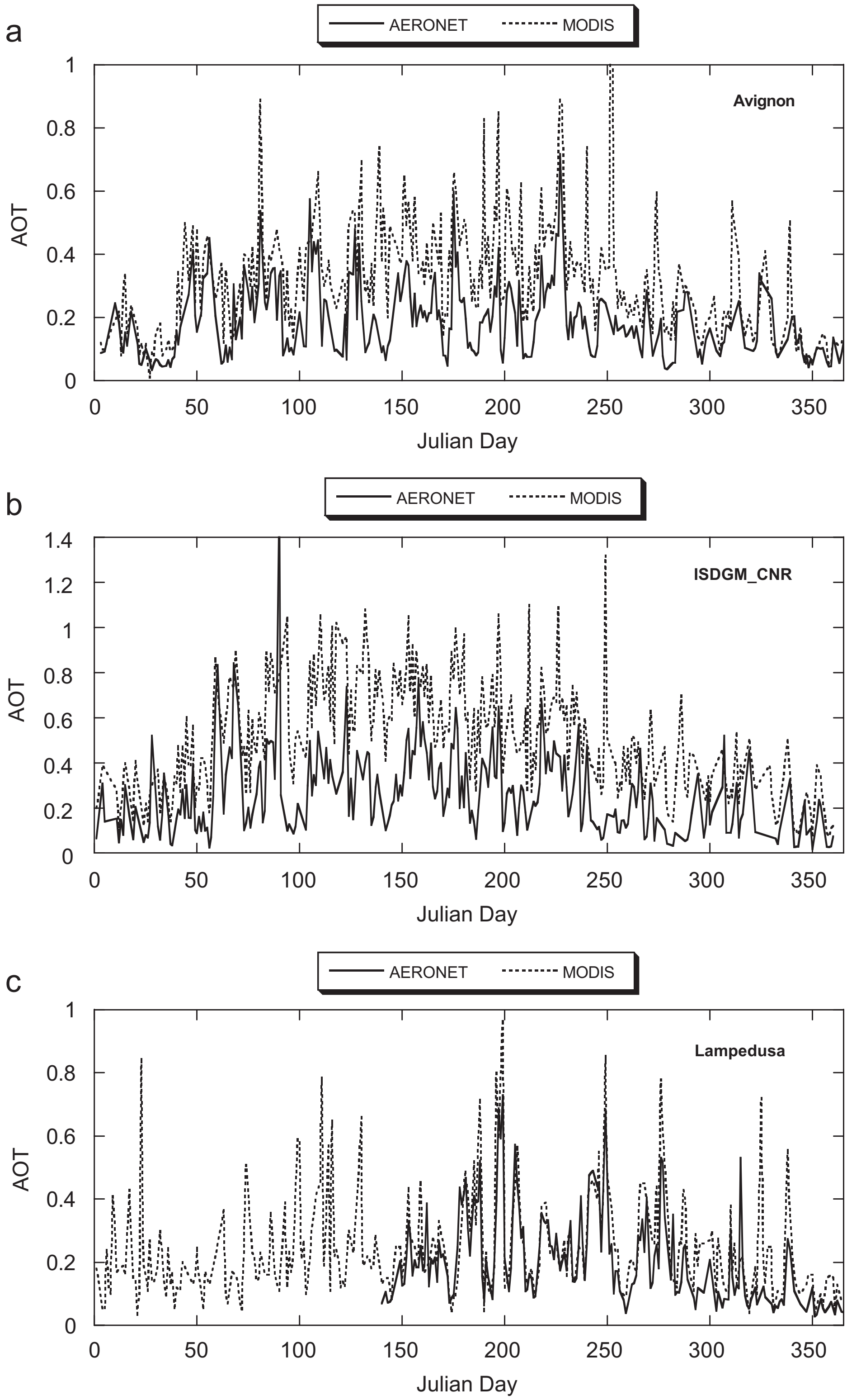

Fig. 3. Temporal variation of AERONET AOT and MODIS AOT during 2003 at (a) Avignon (France), (b) ISDGM-CNR (Italy), and (c) the island of Lampedusa (Italy).

0.05 and 0.16, respectively. In a relative sense, the difference remains more or less constant throughout the year (see Fig. 5). On the whole, the AERONET AOT is about 70% of MODIS AOT.

Over sea, the MODIS AOT appears to have no bias. This is illustrated by the site Lampedusa, which is located on a small island in the Mediterranean Sea. There we find a close agreement between

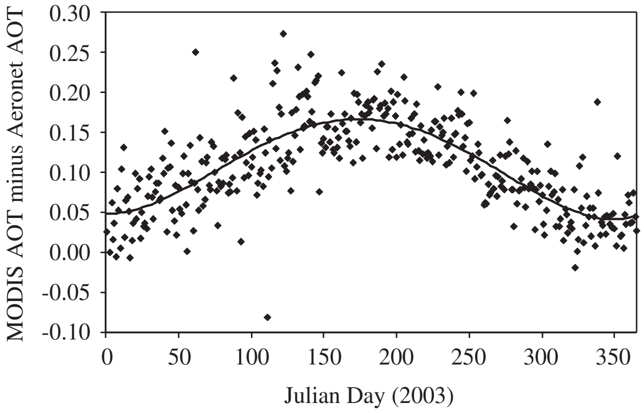

Fig. 4. Seasonal variation of the absolute difference between MODIS and AERONET AOT for all sites averaged.

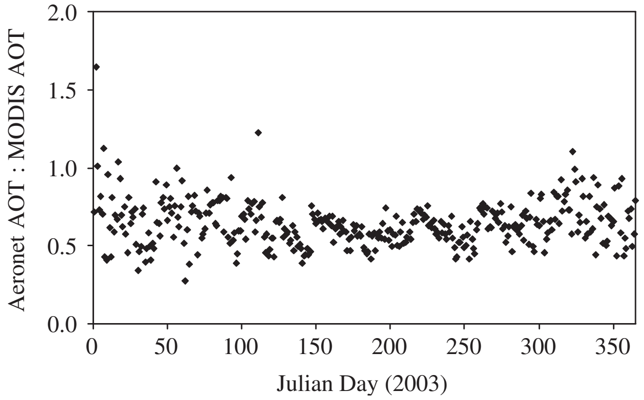

Fig. 5. Relative difference between MODIS and AERONET AOT for all sites averaged.

AERONET and MODIS retrievals (see Fig. 3c). Although Lampedusa is the only site in Europe that is more or less representative of a sea area, the difference in the performance is so large that we suspect that the treatment of the surface refection throughout the year in the retrieval algorithm may be responsible for the seasonal bias that is observed over land.

4.3. Sigma range

We have investigated whether the MODIS AOT over Europe meets the accuracy mentioned in the literature ($\sigma = \pm 0.05 \pm 0.20 \times \text{AOT}$). In Table 2, we summarise the percentage of MODIS retrievals for which the AERONET observation is within 1σ or 2σ from the MODIS retrieval. From a statistical point of view, 68% of the data should be within the 1σ and 95% within the 2σ range, if the residuals follow a normal distribution. The differences between MODIS (v4) and AERONET do not fall within the quoted uncertainty range, which we attribute to the relatively high bias in these data. After correction for the bias in AOT (simply by multiplication with 0.7), the accuracy of AOT does agree with $\sigma = \pm 0.05 \pm 0.20 \times \text{AOT}$. From visual inspection (see Fig. 6), we conclude that the residuals are exhibiting a normal distribution.

Table 2
Percentage of MODIS retrievals for which the AERONET observation is within 1σ or 2σ ($\sigma = \pm 0.05 \pm 0.20 \times \text{AOT}$) from the MODIS retrieval

	% within 1σ	% within 2σ
AOT	55	91
AOT × 0.7	73	96

4.4. Cloud contamination

The validation with AERONET is by definition biased to situations where both the MODIS as well as the AERONET cloud screening identify a cloud-free situation. Situations in which only one of the two retrievals identifies a cloudy situation are not taken into account. Hence, the influence of cloud contamination on the AOT climatology by MODIS is not addressed above. Therefore, we have assessed the number of MODIS retrievals over AERONET stations, which do have a satellite AOT value but miss an AERONET retrieval. In this manner, we determine the number of observations that could erroneously be identified as cloud free in the cloud detection procedure of MODIS. The results of this analysis are shown in Table 3. The table shows that averaged over all AERONET stations, 161 cases occurred where both MODIS and AERONET have AOT measurements, indicating clear-sky conditions. However, there are 80 cases where MODIS provides an AOT retrieval where AERONET does not have a measurement. This might indicate that the MODIS cloud detection is not strict enough in these cases. So, on average, 33% of the MODIS data might suffer from cloud contamination to an unknown extent. We refer to this percentage as the percentage of cloud-contaminated data (PCCD). Note that there are only a few cases (11) in which AERONET does have a measurement and MODIS does not have a measurement. Table 4 shows the PCCDs for the individual stations. The PCCD values range between 10% and nearly 60%. With exception of Evora, Portugal, the lower values tend to be associated with stations in Southern Europe.

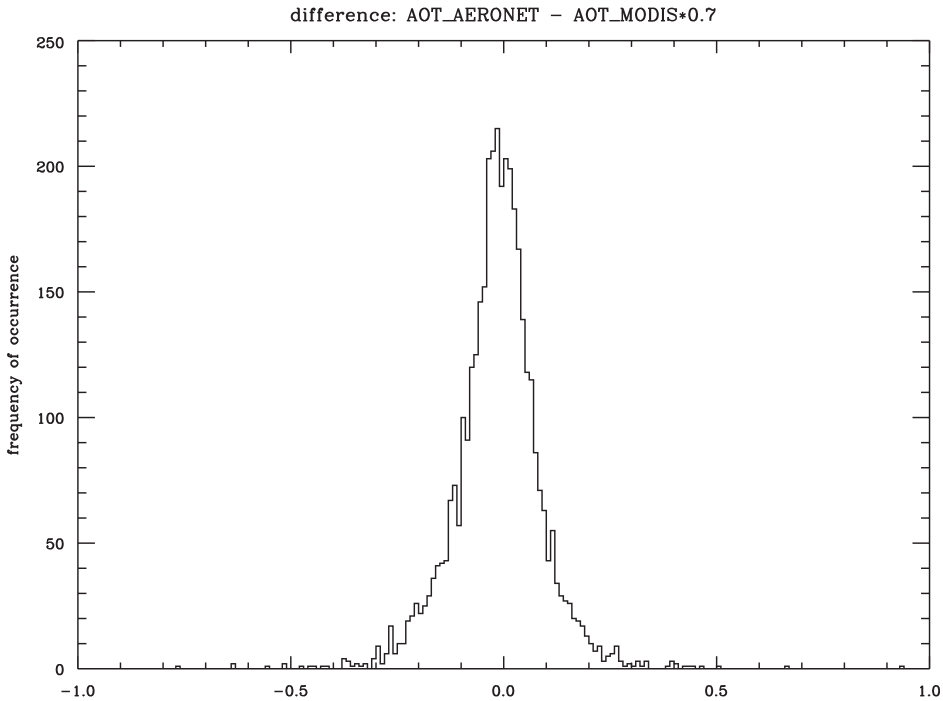

Fig. 6. Frequency distributions of the difference between AERONET and MODIS AOT (bias corrected).

Table 3
"Score card" for MODIS cloud detection

	MODIS observation	
	Yes	No
AERONET observation		
Yes	161	11
No	80	112

Table 4
Percentage of cloud-contaminated data (PCCD) for single AERONET stations

Country	Name	MODIS (%)
Belgium	Oostende	44
Belarus	Minsk	48
France	Lille	47
France	Fontainebleau	45
Portugal	Evora	60
France	Palaiseau	38
Germany	Hamburg	39
France	Toulouse	30
Turkey	IMS-METU	42
Italy	Ispra	28
Italy	IMC_Oristano	32
Italy	Lecce_University	31
Greece	FORTH_CRETE	22
Spain	El_Arenosillo	29
Italy	Rome_Tor_Vergata	23
Italy	Venise	14
France	Avignon	16
Italy	ISDGM_CNR	14
Mean all stations		33

5. Discussion

We have found a large mean bias of MODIS AOT over Europe of about 50%. The extend of the overestimation is very similar to the 48% overestimation reported by Remer et al. (2005) for August 2000 to August 2002. We highlight the strong seasonal signature in the overestimation of AOT by MODIS with a maximum during summer. A likely explanation may be found in the treatment of the surface reflectivity. The AOT is only retrieved for cloud-free pixels and over surfaces that are not too reflective. The reflectivity measured at 2.1 μm at the top-of-atmosphere is used to determine the surface reflectivity at that wavelength. Fine-mode particles, which dominate the AOT in most of Europe, have a negligible optical thickness at 2.1 μm, allowing almost direct observation of the surface (Chu et al., 2003). In the v4.2 retrievals, the surface reflectivity at visible wavelengths is then obtained by assuming a constant ratio between surface reflectivity at 2.1 μm, and that at 0.47 and 0.66 μm. In reality, this ratio depends on surface type and its time-dependent characteristics that determine the reflectivity (e.g., vegetation and soil moisture).

Levy et al. (2005) showed that by using a higher surface reflectivity at 0.47 and 0.66 μm, the resulting lowered AOT over land significantly reduced the discontinuity between land and sea over the coastline in the north-east of the USA. In addition, the agreement between MODIS and AERONET improved in that area. Another important indication comes from the second generation MODIS algorithm (v5.2; Levy et al., 2007) from which data became available during the review of this paper. One of the major improvements to the new algorithm is the inclusion of a variable surface reflectivity. Compared to v4.2, the surface reflection used in the algorithm has increased over more vegetated areas in the mid-latitudes (especially in summer). Indeed, a first inspection of the new annual average distribution shows significantly lower AOT over Europe and only slight discontinuities between land and sea areas.

Although the surface reflection is a likely candidate, it is not straightforward to pinpoint the exact reason for the overestimation. Moreover, it is expected to be a combination of several factors. The new MODIS algorithm (v5.2; Levy et al., 2007) has been updated rigorously. The major changes incorporate the new surface reflectivity assumptions, a new set of aerosol model optical properties, a new aerosol lookup table and a more elaborate inversion scheme. The cloud-screening procedure was not changed. As a consequence of all the improvements, the new product generally yields significantly lower AOT. A preliminary evaluation shows better agreement with AERONET. However, the data from the new collection should be evaluated in detail, which we will do for Europe in the near future. Despite the availability of the new collection, the presented evaluation of collection 4 is still useful as it has been widely used in the (European) research community.

We have used the AERONET data as a standard for validating the MODIS retrievals over Europe. To use AERONET data for the purpose of evaluating co-located satellite derived AOT appears warranted due to their relatively high accuracy (AOT uncertainty $<\pm0.01$ at wavelengths >440 nm) (Holben et al., 1998). We have introduced a simple evaluation of the possible extent of cloud contamination (PCCD) in the MODIS climatology in which we use the availability of AERONET data as an indicator of cloud-free situations. Hence, in that analysis the cloud screening of AERONET is also used as a standard. Some studies indicate that the AERONET cloud-screening algorithms are overly stringent and classify more dense, variable aerosol layers as clouds (Kaufman et al., 2006; O'Neill et al., 2003). In that case our PCCD analysis would yield an overestimation. On the other hand, some evaluations of sun photometer data in combination with other optical instruments (Lidar/Ceilometer) find contamination of the AERONET data with thin, low level clouds or thin cirrus (de Meij et al., 2007). These cloud types are also difficult to detect in (the MODIS) retrieval algorithms (Kaufman et al., 2005). Hence, for now we assume the AERONET cloud screening to be accurate but acknowledge that miss-classified aerosol layers or undetected clouds complicate the analysis.

The PCCD analysis indicates that on average, one third of the MODIS retrievals may still be cloud contaminated. However, the PCCD analysis might be too pessimistic because of various reasons. (1) In the analysis, it is assumed that the AERONET stations were fully operational throughout the year. We have therefore selected those AERONET stations that had data available throughout 2003. This selection coincided with the AERONET stations that had >100 days with observations in 2003. Nevertheless, it might happen that AERONET has no data because of technical reasons, such as power failure, and not because of cloudy conditions. (2) It is also possible that AERONET has no data, while the satellite has valid data, because of partly cloudy conditions. In these conditions, it may happen that clouds occur in the line of sight between the sun and the AERONET station, whereas >12 pixels above and around the AERONET station were identified to be cloud free, which allows a MODIS retrieval. In theory, AERONET observations could also be present when MODIS detects <12 cloud-free pixels. However, the "score card" (Table 3) shows that this feature does not occur. We acknowledge that abovementioned situations happen sometimes and that the PCCD should be interpreted as an upper limit. Hence, assuming we can use the AERONET data as ground-truth, we hypothesise that a relatively high percentage of the retrievals may potentially be cloud contaminated, especially over central Europe.

The bias between satellite and in situ data has important consequences for their use in the mapping of aerosol levels using AOT or the assimilation of the product in a chemistry transport model. In a mapping exercise using geo-statistical techniques, relations are established between AOT and the PM variable that is mapped. In these studies, a bias does not influence the resulting PM distribution as the gradients in the AOT data are actually used rather than the absolute value. The spatial gradients in the retrieved AOT distributions are evaluated with the spatial correlation. The spatial correlation found in this study is similar or better than the spatial correlation that chemistry transport models reach for single aerosol components. Hence, an added value of these observations is expected in mapping procedures, as shown by van de Kassteele et al. (2006). Application of mapping procedures is limited to areas with similar retrieval approaches, because sudden transitions in the AOT fields due to the use of a different aerosol model in the retrieval or sudden changes in the surface properties across the coastline lead to discontinuities in the data base. These issues indicate a more latent issue around the influence of the assumed aerosol characteristics in the retrieval which may not be ideal in all cases and influence the gradients in the distribution.

In case the AOT data are combined with model results using a quantitative, independently obtained relation between AOT and PM, the issue of the bias becomes very relevant. For example, van Donkelaar et al. (2006) calculated the relation between AOT and ground level $PM_{2.5}$ with a model and multiplied this with the retrieved AOT to arrive at an estimate for the ambient $PM_{2.5}$ level. Hence, the uncertainty in the AOT data (bias and spatial gradients) directly influences the result of this exercise. The same applies to more detailed assimilation experiments. Hence, in such studies a bias correction should be made (Koelemeijer et al., 2006b). As a result of this study, we propose to use a multiplication factor of 0.7 for MODIS collection 4 retrievals over the European continent.

6. Conclusions

We have compared the AOT retrieved by MODIS to sun photometer data from the AERONET network in Europe. We have found a large bias of MODIS AOT over Europe of about 50% on average, which is in accordance with earlier findings. We highlight the strong seasonal signature in the overestimation of AOT by MODIS with a maximum during summer. The temporal correlation between MODIS and AERONET is good. After correction for the bias, the accuracy of AOT retrievals agrees with reported uncertainties and the residuals show a normal distribution.

We have introduced a simple method for the evaluation of the possible extend of cloud contamination. Assuming we can use the AERONET data as ground-truth, we hypothesise that a relatively high percentage of the retrievals (one-third) may potentially be cloud contaminated, especially over central Europe. For some stations in central Europe, percentages of $>50\%$ were found.

As a result of this study, we propose to use a bias correction factor of 0.7 for use of MODIS AOT (collection 4) in assimilation exercises over central and Western Europe.

Acknowledgements

This work was funded by the Netherlands Agency for Aerospace Programmes (NIVR) through grant 53.516RI and internal TNO research programmes.

References

Al-Saadi, J., et al., 2005. Improving national air quality forecasts with satellite aerosol observations. Bulletin of the American Meteorological Society 86, 1249–1261.

Anderson, T.L., Charlson, R.J., Schwartz, S.E., Knutti, R., Boucher, O., Rodhe, H., Heintzenberg, J., 2003. Climate forcing by aerosols—a hazy picture. Science 16, 1103–1104.

Brunekreef, B., Holgate, S.T., 2002. Air pollution and health. The Lancet 360, 1233–1242.

Builtjes, P.J.H., ten Brink, H.M., de Leeuw, G., van Loon, M., Robles Gonzales, C., Schaap, M., 2001. Aerosol air quality satellite data. BCRS USP-2 Report 00-33, BCRS, Delft, The Netherlands.

Charlson, R.J., Schwartz, S.E., Hales, J.M., Cess, R.D., Coakley, J.A., Hansen, J.E., Hofmann, D.J., 1992. Climate forcing by anthropogenic aerosols. Science 255, 423–430.

Chu, D.A., Kaufman, Y.J., Ichoku, C., Remer, L.A., Tanré, D., Holben, B.N., 2002. Validation of MODIS aerosol optical thickness retrieval over land. Geophysical Research Letters 29 (12).

Chu, D.A., Kaufman, Y.J., Zibordi, G., Chern, J.D., Mao, J., Li, C., Holben, B.N., 2003. Global monitoring of air pollution over land from the Earth Observing System—Terra Moderate Resolution Imaging Spectroradiometer (MODIS). Journal of Geophysical Research 108, 4661.

Collins, W.D., Rasch, P.J., Eaton, B.E., Khattatov, B.V., Lamarque, J-F., Zender, C.S., 2001. Simulating aerosols using a chemical transport model with assimilation of satellite aerosol retrievals: methodology for INDOEX. Journal of Geophysical Research 106, 7313–7336.

De Leeuw, G., Neele, F.P., Hill, M., Smith, M.H., Vignati, E., 2000. Sea spray aerosol production by waves breaking in the surf zone. Journal of Geophysical Research 105 (D2), 29397–29409.

de Meij, A., Wagner, S., Gobron, N., Thunis, P., Cuvelier, C., Dentener, F., Schaap, M., 2007. Scale issues in aerosol modeling: a case study on chemical and optical properties over the greater Milan area (Italy), June 2001. Atmospheric Research 85, 243–267.

Dockery, D.W., Pope III, C.A., Xu, X., Spengler, J.D., Ware, J.H., Fay, M.E., Ferris, B.G., Speizer, F.E., 1993. An association between air pollution and mortality in six US cities. The New England Journal of Medicine 329, 1753–1759.

Dubovik, O., Lapyonok, T., Kaufman, Y.J., Chin, M., Ginoux, P., Remer, L., Holben, B.N., 2004. Retrieving sources of fine aerosols from MODIS and AERONET observations by inverting GOCART model. Proceedings of SPIE—The International Society for Optical Engineering 5652, 66–75.

Engel-Cox, J.A., Holloman, C.H., Coutant, B.W., Hoff, R.M., 2004. Qualitative and quantitative evaluation of MODIS satellite sensor data for regional and urban scale air quality. Atmospheric Environment 38, 2495–2509.

Hansen, J.E., Sato, M., 2001. Trends of measured climate forcings agents. Proceedings of the National Academy of Science 98, 14778–14783.

Holben, B.N., Eck, T.F., Slutsker, I., Tanre, D., Buis, J.P., Setzer, A., Vermote, E., Reagan, J.A., Kaufman, Y.F., Nakajima, T., Lavenu, F., Jankowiak, I., Smirnov, A., 1998. AERONET—a federated instrument network and data archive for aerosol characterisation. Remote Sensing of the Environment 66, 1–16.

Hutchison, K.D., 2003. Applications of MODIS satellite data and products for monitoring air quality in the state of Texas. Atmospheric Environment 37, 2403–2412.

Ichoku, C., Chu, D.A., Mattoo, A., Kaufman, Y.J., Remer, L.A., Tanre, D., Slutsker, I., Holben, B.N., 2002. A spatio-temporal approach for global validation and analysis of MODIS aerosol products. Geophysical Research Letters 29.

Ichoku, C., Remer, L.A., Eck, T.F., 2005. Quantitative evaluation and intercomparison of morning and afternoon moderate resolution imaging spectroradiometer (MODIS) aerosol measurements from Terra and Aqua. Journal of Geophysical Research 110, D10S03.

Kaufman, Y.J., Tanré, D., 1998. Algorithm for remote sensing of tropospheric aerosol from MODIS. Product ID MOD04.

Kaufman, Y.J., Tanré, D., Remer, L.A., Chu, A., Vermote, E., Holben, B.N., 1997. Operational remote sensing of tropospheric aerosol over landfrom EOS moderate resolution imaging spectroradiometer. Journal of Geophysical Research 102, 17051–17067.

Kaufman, Y.J., Remer, L.A., Tanré, D., Li, R., Kleidman, R., Mattoo, S., Levy, R.C., Eck, T.F., Holben, B.N., Ichoku, C., Vanderlei-Martins, J., Koren, I., 2005. A critical examination of the residual cloud contamination and diurnal sampling effects on MODIS estimates of aerosol over ocean. IEEE Transactions on Geoscience and Remote Sensing 43 (12), 2886–2897.

Kaufman, Y.J., Gobbi, G.P., Koren, I., 2006. Aerosol climatology using a tunable spectral variability cloud screening of AERONET data. Geophysical Research Letters 33, L07817.

Koelemeijer, R.B.A., Schaap, M., Timmermans, R.M.A., Homan, C.D., Matthijsen, J., van de Kassteele, J., Builtjes, P.J.H., 2006a. Mapping aerosol concentrations and optical thickness over Europe—PARMA final report. MNP Report 555034001, Bilthoven, The Netherlands.

Koelemeijer, R.B.A., Homan, C.D., Matthijsen, J., 2006b. Comparison of spatial and temporal variations of aerosol optical thickness and particulate matter in Europe. Atmospheric Environment 40, 5304–5315.

Koren, I., Kaufman, Y.J., Rosenfeld, D., Remer, L.A., Rudich, Y., 2005. Aerosol invigoration and restructuring of Atlantic convective clouds. Geophysical Research Letters 32 (14), 1–4.

Levy, R.C., Remer, L.A., et al., 2005. Evaluation of the MODIS aerosol retrievals over ocean and land during CLAMS. Journal of Atmospheric Sciences 62 (4), 974–992.

Levy, R.C., Remer, L.A., Mattoo, S., Vermote, E.F., Kaufman, Y.J., 2007. Second-generation operational algorithm: retrieval of aerosol properties over land from inversion of moderate resolution imaging spectroradiometer spectral reflectance. Journal of Geophysical Research D: Atmospheres 112 (13) (art. no. D13211).

O'Neill, N.T., Eck, T.F., Smirnov, A., Holben, B.N., Thulasiraman, S., 2003. Spectral discrimination of coarse and fine mode optical depth. Journal of Geophysical Research 108 (D17), 4559.

Pope III, C.A., Dockery, D.W., Schwartz, J., 1995. Review of epidemiological evidence of health effects of particulate air pollution. Inhalation Toxicology 7, 1–18.

Remer, L.A., Kaufman, Y.J., Tanré, D., Mattoo, S., Chu, D.A., Martins, J.V., Li, R.-R., Ichoku, C., Levy, R.C., Kleidman, R.G., Eck, T.F., Vermote, E., Holben, B.N., 2005. The MODIS aerosol algorithm, products, and validation. Journal of Atmospheric Sciences 62, 947–973.

Robles Gonzalez, C., Veefkind, J.P., de Leeuw, G., 2000. Aerosol optical depth over Europe in August 1997 derived from ATSR-2 data. Geophysical Research Letters 27 (7), 955–958.

van de Kassteele, J., Koelemeijer, R.B.A., Dekkers, A.L.M., Schaap, M., Homan, C.D., Stein, A., 2006. Statistical mapping of PM_{10} concentrations over Western Europe using secondary information from dispersion modeling and MODIS satellite observations. Stochastic Environmental Research and Risk Assessment, doi:10.1007/s00477-006-0055-4.

van Donkelaar, A., Martin, R.V., Park, R.J., 2006. Estimating ground-level $PM_{2.5}$ using aerosol optical thickness determined from satellite remote sensing. Journal of Geophysical Research 111, D21201.

Wang, J., Christopher, S.A., 2003. Intercomparison between satellite-derived aerosol optical thickness and $PM_{2.5}$ mass: implications for air quality studies. Geophysical Research Letters 30 (21), 2095.

Wen, G., Marshak, A., Cahalan, R.F., Remer, L.A., Kleidman, R.G., 2007. 3-D aerosol-cloud radiative interaction observed in collocated MODIS and ASTER images of cumulus cloud fields. Journal of Geophysical Research D: Atmospheres 112 (13) (art. no. D13204).

Yu, H., Kaufman, Y.J., Chin, M., Feingold, G., Remer, L.A., Anderson, T.L., Balkanski, Y., Bellouin, N., Boucher, O., Christopher, S., DeCola, P., Kahn, R., Koch, D., Loeb, N., Reddy, M.S., Schulz, M., Takemura, T., Zhou, M., 2006. A review of measurement-based assessments of the aerosol direct radiative effect and forcing. Atmospheric Chemistry and Physics 6 (3), 613–666.