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**Trend analysis of the
Aerosol Optical
Thickness**

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Trend analysis of the Aerosol Optical Thickness and Ångström Exponent derived from the global AERONET spectral observations

J. Yoon, W. von Hoyningen-Huene, A. A. Kokhanovsky, M. Vountas, and J. P. Burrows

Institute of Environmental Physics, University of Bremen, Bremen, Germany

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Correspondence to: J. Yoon (yoon@iup.physik.uni-bremen.de)

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Abstract

Regular aerosol observations based on well-calibrated instruments have led to a better understanding of the aerosol radiative budget on Earth. In recent years, these instruments have played an important role in the determination of the increase of anthropogenic aerosols by means of long-term studies. Only few investigations regarding long-term trends of aerosol optical characteristics (e.g. Aerosol Optical Thickness (AOT) and Ångström Exponent (ÅE)) have been derived from ground-based observations. This paper aims to derive and discuss linear trends of AOT (440, 675, 870, and 1020 nm) and ÅE (440–870 nm) using AErosol RObotic NETwork (AERONET) spectral observations. Additionally, temporal trends of Coarse- and Fine-mode dominant AOTs (CAOT and FAOT) have been estimated by applying an aerosol classification based on accurate ÅE and Ångström Exponent Difference (ÅED). In order to take into account the fact that cloud disturbance is having a significant influence on the trend analysis of aerosols, we introduce a weighted least squares regression depending on two weights: (1) monthly standard deviation and (2) Number of Observations (NO) per month.

Temporal increase of FAOTs prevails over regions dominated by emerging economy or slash-burn agriculture in East Asia and South Africa. On the other hand, insignificant or negative trends for FAOTs are detected over Western Europe and North America. Over desert regions, both increase and decrease of CAOTs are observed depending on meteorological conditions.

1 Introduction

Aerosols directly influence air quality and solar light extinction as well as indirectly influence the cloud microphysics and cloud radiative forcing (Ackerman et al., 2000; Haywood and Boucher, 2000; Penner et al., 2001; Ramanathan et al., 2001; Léon et al., 2001; Ortore and Francione, 2008). Considerable increase of anthropogenic aerosol is complicating the situation, which leads to the fact that the impact of aerosols

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on climate change still remains at “med-low” or “low” level of scientific understanding (IPCC, 2007).

Recently, several studies (Li et al., 2009; Yu et al., 2009; Zhang et al., 2010; Street et al., 2009; Karnieli et al., 2009; Mishchenko et al., 2007; Mishchenko and Geogdzhayev, 2007; Zhao et al., 2008; Massie et al., 2004) based on well-validated aerosol retrieval algorithms (Remer et al., 2005; Kaufman et al., 1997; Higurashi and Nakajima, 1999; Mishchenko et al., 1999a; Jeong et al., 2005; Higurashi et al., 2000; Stowe et al., 1999; Heidinger et al., 2004; Torres et al., 2002; DiGirolamo and Wilson, 2003; Martonchik et al., 2004; Diner et al., 2006; Martins et al., 2002; von Hoyningen-Huene et al., 2011) using both well-calibrated observations (e.g., Sea-viewing Wide Field-of-view Sensor (SeaWiFS), MEdium Resolution Imaging Spectrometer (MERIS), Multi-angle Imaging SpectroRadiometer (MISR), and Moderate Resolution Imaging Spectroradiometer (MODIS)) and long-term records from space instruments (e.g., Advanced Very High Resolution Radiometer (AVHRR), Total Ozone Mapping Spectrometer (TOMS), and Along Track Scanning Radiometer (ATSR)) have contributed significantly to the understanding of global aerosol trends. Related to these trends, a hypothesis of global brightening or dimming has been discussed as well (Wild et al., 2005, 2007; Ohmura, 2006; Stanhill, 2007; Norris and Wild, 2007). However, aerosol retrievals based on satellite observations often have serious uncertainties caused by instrument calibration and assumptions within the algorithms (Li et al., 2009; Higurashi and Nakajima, 1999; Ignatov and Stowe, 2002). It was found that improvements of the retrieval algorithms need to be accompanied by the reduction of uncertainties due to unscreened clouds, a priori aerosol characteristics (e.g. Single Scattering Albedo (SSA) and phase function), and minimization of the error due to separation techniques to discriminate spectral surface signals (Mishchenko et al., 1999a; Ignatov and Nalli, 2002; Jeong et al., 2005; Myhre et al., 2004; Jeong and Li, 2005; Kahn et al., 2005, 2007; Chen et al., 2008; Kalashnikova and Kahn, 2006; Li et al., 2009; Yu et al., 2009; Zhou et al., 2005; Husar et al., 1997; Haywood et al., 2001; Kokhanovsky and de Leeuw, 2009; von Hoyningen-Huene et al., 2011). For polar-orbiting satellite observations, it is difficult to

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avoid the bias in aerosol sampling caused by frequent cloud disturbance (Remer et al., 1997; Dubovik et al., 2001; Jeong and Li, 2005; Jeong et al., 2005; Yoon et al., 2011) and coarse temporal resolution of the observation. Therefore, it is also necessary to investigate aerosol trends based on ground-based observations.

5 The AErosol RObotic NETwork (AERONET) program (<http://aeronet.gsfc.nasa.gov/>) aims to provide a global distribution of aerosol optical properties and to validate satellite retrievals. Despite aerosols below clouds being underrepresented in the AERONET observation database as well (Remer et al., 1997; Dubovik et al., 2001), this network of ground observations provides suitable data for trend analysis of Aerosol Optical Thick-
10 ness (AOT) based on continuous long-term observations with high temporal resolution as well as good retrieval accuracy (Holben et al., 1998, 2001; Eck et al., 1999; Smirnov et al., 2000). Recently, Karnieli et al. (2009) and de Meij et al. (2010) have discussed AOT trends using long-term AERONET data and have compared them with satellite ob-
15 servations (e.g., Multi-angle Imaging SpectroRadiometer (MISR) and Moderate Reso- lution Imaging Spectroradiometer (MODIS)) and model simulations (e.g., Co-operative Programme for Monitoring and Evaluation of the Long-range Transmission of Air Pollu-
20 tants (EMEP), the Region Emission Inventory (REAS) and the Intergovernmental Panel on Climate Change (IPCC, RCP 3PD)). However, they have published no further in- formation about the uncertainty of cloud disturbances and the influences of aerosol classification in the AOT trend analysis.

Cloud disturbances introduce serious uncertainties to trend analysis by decreasing the Number of Aerosol Observations (NO) per temporal interval during persistent cloudiness, thus leading to problems with the statistical representativeness (Yoon, et al., 2011). Therefore, this paper introduces weighted trends using monthly standard
25 deviation and NO to reduce cloud uncertainties in the trend analysis.

Additionally, without applying a classification of aerosol types, the trend studies are only of limited use in order to understand why the aerosol loading changes in time. To support such studies, spectral AOT observations, i.e., the Ångström Exponent (ÅE) (Ångström, 1929), provide a useful quantity to estimate the mean size of particles.

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In general, submicron or supermicron aerosols have higher or lower ÅEs accordingly. However, ÅE is not a perfect indicator to show the exact average size of particles as it also is dependent on aerosol absorption and size distribution. Even though the AERONET inversion process provides useful aerosol optical properties (e.g., volume size distribution and SSA), these could hardly be used to classify aerosols as they are only valid for AOT(440 nm) >0.4 and solar zenith angle >50° (Dubovik et al., 2000). Over the last two decades, there have been several studies of ÅE curvature related to the aerosol size distribution (Kaufman, 1993; Eck et al., 1999; O'Neill et al., 2001a, b, 2003, 2005; Schuster et al., 2006; Gobbi et al., 2007). Relationships described in these papers provide a more suitable framework to classify aerosol types using ÅE and Ångström Exponent Difference (ÅED). Therefore, this paper attempts to analyze the temporal trends of Coarse- and Fine-mode dominant AOT (CAOT and FAOT) separately by applying such aerosol classification.

The present study aims to investigate and analyze the long-term trends of AOT, CAOT, FAOT (440, 675, 870, and 1020 nm), and ÅE (440–870 nm) at several AERONET stations. For this purpose, the second section describes in detail the methodology used for the selection of suitable AERONET stations, the weighted least squares regression to consider the cloud uncertainty, and the classification of coarse- and fine-mode dominant aerosols. In the third section, the aerosol trends at the specific AERONET stations are discussed regionally. The conclusions are summarized in the final section.

2 Methodology

For a reliable analysis of the aerosol trends based on the ground observation, new approaches are introduced: (1) the selection criteria for the AERONET stations having sufficient and nearly-complete multi-year data sets, (2) the weighted least squares regression to consider cloud uncertainty, and (3) the classification of coarse- and fine-mode dominant aerosols.

2.1 Selection criteria for suitable AERONET stations

The AERONET program has provided high quality aerosol products for the past decades over roughly 850 stations globally. However, not all stations distribute a sufficiently large temporal record suitable for a trend analysis. Firstly, we distinguished suitable AERONET stations having a sufficiently large NO record per month. The NO basically depends on the seasonal daytime length, the station's location, the operational instrument status, the cloud disturbance, and the data quality verification process. To get statistically meaningful monthly average values, a large NO is required as the sample average based on a larger sample number is closer to the real average. Therefore, we have defined the minimum NO of 300 per month (10 observations per day) to consider a reliable monthly average value. Another important issue in the trend analysis is the annual completeness of the data set, which needs to consist out of reliable monthly averages. In other words, the absence of continuous monthly averages in the yearly data sets can cause a serious uncertainty in the trend analysis.

Basically, we have set up the following set of criteria to choose suitable AERONET stations:

1. The qualified monthly average is calculated with a NO larger than 300 per month (10 observations per day).
2. The sufficiently-complete yearly set consists out of more than seven qualified monthly averages.
3. A suitable AERONET station needs to have more than five sufficiently-complete years of observation history.

Although a five-years time series may be insufficiently short for a statistically significant trend analysis, it is a first, pragmatic time span to investigate aerosol temporal change from AERONET observations. Figure 1 shows the NO since 1993 for suitable AERONET stations shown in Table 1. Because each station has a different observation

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history as well as differently qualified (with respect to the above listed conditions) data sets, it is difficult to perform the investigation of aerosol trends during the same time period. The research periods when the data sets satisfy the selection criteria for each station are indicated by blue fields in Fig. 1. Detailed information about the geolocation and the research periods for the selected AERONET stations are listed in Table 1.

2.2 Weighted least squares regression

A simple linear model, which is used by minimizing chi-square error statistics, has been adopted in this study. Let Y_t be the monthly AOT averages. The linear trend model is given by the following equation:

$$Y_t = A + BX_t + \epsilon_t, \quad t = 1, \dots, T, \quad (1)$$

where A is a constant term, B is the magnitude of the trend per year ($X_t = t/12$), ϵ_t is the noise, t is the month index, and T is the total number of months. However, as mentioned before, in order to analyze reliable trends, this simple model needs to take into account cloud disturbance.

Each monthly AOT average has been calculated with different NO, which is directly related to the number of cloud occurrence. The trend analysis based on monthly averages during cloudy season may strongly be biased through poor sampling, so that a weighting factor is used to derive the respective trends. Figure 2 depicts the removal ratio of cloud (black line) and quality-unassured (red line) observations of AERONET data. If NO is large enough to ignore the other effects (daytime length, station location, and operational instrument status), then main factors affecting NO could be the cloud disturbance and the data quality verification process. In Fig. 2, the NO of monthly level 2.0 data (n_t) correlate negatively with the cloud removal ratio for most of the stations.

We introduce a weighting factor for the trend analysis.

$$\chi^2(A, B) = \sum_{t=1}^T (wt_t \times (Y_t - A - BX_t))^2 \quad (2)$$

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where, $wt_t (= \sqrt{n_t}/\sigma_t)$ is the monthly weighting factor defined as ratio of NO (n_t) and monthly standard deviation (σ_t). Monthly standard deviation is by itself a suitable weight as it statistically shows the representativeness (variability or diversity) of the average. In the following we will estimate the cloud uncertainty through the comparison between the weighted and unweighted trends.

2.3 Classification of coarse- and fine-mode dominant aerosols

In order to quantify the change in anthropogenic (generally, fine-mode dominant) and natural (coarse-mode dominant) aerosols, an aerosol classification needs to be applied as well. The AERONET inversion process (Dubovik and King, 2000; Dubovik et al., 2000, 2001, 2002, 2006; Sinyuk et al., 2007) generates various aerosol characteristics such as volume size distribution and SSA. However, the data are only provided for the following conditions: AOT(440 nm) >0.4 and solar zenith angle >50° (Dubovik et al., 2000). Figure 3 shows the normalized frequency of AOT (440 nm) and solar zenith angle at the selected AERONET stations. Generally, the normalized frequency (histogram) distributions of AOT (440 nm) are skewed and have long tails towards larger values of AOT. The percentage of AERONET level 2.0 inversion data to the total observations is indicated as a pie chart on the lower-left hand side in Fig. 3. In most cases, it is difficult to use the AERONET inversion data for aerosol classification because of a low proportion to total observations meeting the conditions mentioned above. Alternatively, we propose a classification of coarse- and fine-mode dominant aerosols using ÅE and ÅED retrievals from AERONET direct sun data. ÅE and ÅED are defined as:

$$\text{ÅE} = -\frac{\ln(\tau_{440\text{nm}}/\tau_{870\text{nm}})}{\ln(440\text{nm}/870\text{nm})} \quad (3)$$

$$\text{ÅED} = -\frac{\ln(\tau_{440\text{nm}}/\tau_{675\text{nm}})}{\ln(440\text{nm}/675\text{nm})} + \frac{\ln(\tau_{675\text{nm}}/\tau_{870\text{nm}})}{\ln(675\text{nm}/870\text{nm})} \quad (4)$$

where, τ_λ is AOT at wavelength (λ).

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Several investigations have been previously devoted to the curvature of the spectral dependence of the optical depth in order to derive more accurate aerosol size information. For example, Kaufman (1993) found that the spectral curvature shows a transition from mixed accumulation and coarse particle modes to a dominant accumulation mode. Eck et al. (1999) investigated the wavelength dependence of the optical depth of biomass burning, urban, and desert dust aerosols. O'Neill et al. (2001a, b, 2003, 2005) and Schuster et al. (2006) presented a detailed analysis and compared simulations and observations in order to investigate the relationship between aerosol size distribution and spectral dependence of the AOT. Gobbi et al. (2007) have set up a useful straight-forward graphical framework applicable to classify aerosol properties (effective radius and fine mode fraction to total AOT) using ÅED as a measure of the curvature. They have applied the graphical framework to AERONET data and were able to characterize different aerosol types such as pollution, mineral dust, and biomass burning. However, none of the above mentioned publications involved their methods in trend analyses.

Here, we build up a similar classification as Gobbi et al. (2007). With Fig. 4, using the Mie theory, we tested the relationship between ÅE and ÅED simulated with many bimodal volume size distributions consisting out of mode radii, widths, fine volume fractions, and refractive indices shown Table 2. Negative ÅED shows a high proportion of fine mode aerosol for the same ÅE. In this study, we set up the classification using both ÅE and ÅED to consider aerosol mean size and fine volume fraction together.

In this study, coarse-mode (fine-mode) dominant aerosols are classified by lower (higher) values than ÅE of at least 1.0 (Kaufman, 1993) and a fine volume fraction of 50 %. For positive ÅED values (larger than 0.17), the fine volume fraction of 50 % scatters significantly (Fig. 4). In order to be able to classify even for these values, additional Mie simulations (Mishchenko et al., 1999b, 2002) have been performed based on aerosol characteristics of typical aerosols (urban-industrial and mixed, biomass burning, desert dust, oceanic from Dubovik et al. (2001)) shown in Table 3 and Fig. 5. The red line shown in Fig. 5, especially the upper part relevant for positive ÅED, could be

set to an ÅE value of 1.4 (Tanré et al., 2001; Pereira et al., 2011; Shinozuka et al., 2011). This Fig. shows the red line discriminates coarse- and fine-mode dominant aerosols well. It also demonstrates that coarse-mode dominant aerosols (desert dust and maritime aerosols) have smaller ÅE and positive ÅED according to the increase of aerosol loading. As already mentioned, the mean particle size of fine-mode dominant aerosols could increase due to the increase of aerosol loading, despite of larger fine volume fractions. These tendencies of ÅE and ÅED for coarse- and fine-mode dominant aerosols are more apparent when looking at application of AERONET data.

Figure 6 shows a scatterplot of ÅE and ÅED derived from AERONET datasets at fifteen stations including the red classification line. After applying the classification, the percentages of coarse-mode (C) and fine-mode (F) dominant aerosols are shown as a pie chart at the upper-left hand side of Fig. 6. To avoid large errors in ÅE and ÅED from low AOTs, we only take into account those observations having an AOT (440 nm) larger than 0.15 (Gobbi et al., 2007). Most ÅE and ÅED from AERONET observations in Fig. 6 are generally in good agreement with Mie simulations in Fig. 5. In other words, they are positioned within the simulation border (thick black line) and their variations according to increase of aerosol loading are similar to the simulations. As can be seen in Fig. 6 for practically all AERONET stations, a different percentage of coarse- and fine-mode dominant aerosols is observed due to different regional aerosol sources and atmospheric conditions. All stations over West Africa (Banizoumbou (C 99 % >> F 1 %), Dakar (C 99 % >> F 1 %), and Ouagadougou (C 99 % >> F 1 %)) and Middle East (SEDE_BOKER (C 85 % > F 15 %) and Solar_Village (C 99 % >> F 1 %)) are influenced by coarse-mode dominant aerosols because the regions are close to deserts. In contrast, industrial pollutant and biomass burning aerosols are dominant over Western Europe (Avignon (C 25 % < F 75 %) and Ispra (C 14 % < F 86 %)), South Africa (Mongu (C 7 % << F 93 %) and Skukuza (C 22 % < F 78 %)), and North America (GSFC (C 14 % < F 86 %) and MD_Science_Center (C 15 % < F 85 %)). Especially, typical anthropogenic aerosols caused by urbanization and industrialization as well as natural aerosols brought in by strong westerly winds are observed over East Asia

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(Beijing (C 64 % > F 36 %) and Shirahama (C 44 % < F 56 %)). The classification is not applicable to data observed at Mauna_Loa and Sevilleleta because most AOTs (440 nm) over these stations were less than 0.15.

By applying the aerosol classification, it is possible to analyze more reliable trends separately for coarse- and fine-mode dominant aerosols. However, stations dominated by coarse-mode aerosols (such as Banizoumbou, Dakar, Ouagadougou, and Solar_Village) or those with an extremely small aerosol load (such as Mauna_Loa and Sevilleleta) are neglected.

3 Trend analysis

For this part of study, we have selected fifteen AERONET stations providing datasets meeting the requirements specified in Sect. 2.1. In the following sections, the trends for the stations located in several regions (Western Europe, West Africa, South Africa, Middle East, East Asia, North America, and Free troposphere/Pacific) are discussed. The trends of ÅE (440–870 nm) and AOT (440 nm) at the AERONET stations are shown in Fig. 7. For clarity, the error bar is scaled by a factor 10 of the standard error ($\sigma_t/\sqrt{n_t}$), which is inversely used for the weighted trend analysis. Comparison between unweighted (blue line and text on the left upper part) and weighted trends (red line and text on the right upper part) allows to estimate the uncertainty caused by cloud disturbance in the trend analysis.

In this paper, we would like to discuss the aerosol trends mainly on the basis of the weighted trend of AOT (440 nm) and ÅE (440–870 nm). As previously mentioned, we also introduce a classification of coarse- and fine-mode dominant aerosols based on the Mie theory in the trend analysis in Fig. 9. Finally, the weighted trends of ÅE (440–870 nm), AOT, CAOT, and FAOT (440 nm) over all AERONET stations are summarized in Figs. 8 and 10.

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3.1 Western Europe

The averages of $\dot{A}E$ shown in Fig. 7, for Avignon ($\langle \dot{A}E \rangle = 1.43$) and Ispra ($\langle \dot{A}E \rangle = 1.51$) over Western Europe, were mainly influenced by urbanization and industrial pollutants, such as ammonium salts of sulphate and nitrate (Robles González et al., 2000; Benkovitz et al., 1996). The seasonal variation of $\dot{A}E$ is small as the major source during the year is industrial pollutant. On the other hand, the AOTs over Western Europe exhibit a significant seasonal variation, which is increasing from spring to summer and decreasing from autumn to winter (Fig. 7). Basically, the AOT depends on the aerosol extinction coefficient (influenced, e.g., by aerosol types, emission intensity, and relative humidity) and boundary layer height. Especially industrial pollutants composed of sulphur are enhanced during summer due to stronger solar radiation (Marmer et al., 2007; Karnieli et al., 2009). Additionally, the absence of removal process (e.g., rain/monsoon) as well as a high boundary layer height causes higher AOTs over Europe in summer (Gerasopoulos et al., 2003; Bergamo et al., 2008; Venzac et al., 2009). The weighted AOT trends over both stations are insignificant or decreasing (+0.98 %/year at Avignon and -2.30 %/year at Ispra), most likely due to strict environmental regulations for mitigating climate change and improving air quality (Smith et al., 2001; Streets et al., 2006; Zhao et al., 2008). There is little difference between weighted and unweighted trends of $\dot{A}E$ and AOT (Fig. 7). The AOTs at Avignon and Ispra over Western Europe, where most aerosols are classified as fine-mode dominant caused by industrial activity, tend to be negligible or decreasing (+0.24 %/year at Avignon and -2.30 %/year at Ispra as shown in Fig. 9).

3.2 West Africa

Mineral dust mainly from the Saharan and Sahel regions is the most abundant aerosol type over West Africa influencing Banizoumbou, Dakar, and Ouagadougou stations year-round (Prospero and Lamb, 2003; Washington and Todd, 2005; Moulin and Chiappello, 2004; Reeves et al., 2010). Besides, biomass burning is frequently advected

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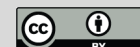
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by the West African monsoon in summer from central Africa (Hao and Liu, 1994). Additionally, low AOT might occur as a result of efficient wet removal of aerosol particles due to heavy precipitation (Reeves et al., 2010; Huang et al., 2009). In Fig. 7, series of these phenomena over West Africa represent the seasonal pattern in ÅE and AOT. Insignificant or decreasing trends of dust aerosol due to decreasing dust activity (Evan et al., 2007) are observed over most stations in West Africa (+0.22%/year at Banizoumbou, -1.53%/year at Dakar, and -1.95%/year at Ouagadougou), which is consistent with the results from the Global Aerosol Climatology Project (GACP) data (Mishchenko and Geogdzhayev, 2007; Mishchenko et al., 2007), in situ measurement, AVHRR, and TOMS observations (Chiapello and Moulin, 2002; Chiapello et al., 2005; Zhao et al., 2008). The weighted trends of ÅE and AOT at Dakar and at Ouagadougou are different compared to unweighted trends due to frequent cloud disturbance. CAOT trends over West Africa (+0.29%/year at Banizoumbou, -1.47%/year at Dakar, and -1.86%/year at Ouagadougou in Fig. 9) are generally similar with AOT trends.

3.3 South Africa

ÅE as well as AOT over stations in South Africa (i.e., Mongu and Skukuza) exhibit a strong seasonality due to pronounced wet and dry seasons (see Fig. 7), and the presence of biomass burning aerosol (Tyson, 1986; Swap et al., 1996). Biomass burning leads to a large aerosol load in warm and dry seasons. The cloud uncertainty in the trend analysis is insignificant because the biomass burning generally happens before the beginning of the rain seasons. A noticeable increase of AOT at Mongu (+2.26%/year) is most likely affected by biomass burning (Mishchenko and Geogdzhayev, 2007; Zhao et al., 2008). Accordingly, positive trends of FAOT over South Africa are observed (+1.76%/year at Mongu and +0.75%/year at Skukuza) in Fig. 9.

3.4 Middle East

The stations SEDE_BOKER and Solar_Village are located within the Middle East and provide a long record of measurements because of stable, clear-sky weather conditions (Basart et al., 2009) (see Fig. 2). In this region, aerosol size and composition are dominated by the regional petroleum industry (Zhao et al., 2008; Basart et al., 2009) and mineral dust which is transported from the Anatolian plateau, Sahara, Negev, and Arabian deserts (Andreae et al., 2002; Kubilay et al., 2003; Derimian et al., 2006; Basart et al., 2009; Sabbah et al., 2001, 2006; Smirnov et al., 2002; Tafuro et al., 2006; Sabbah and Hasan, 2008). The latter explains the clear periodical pattern of ÅE and AOT seen in Fig. 7. The AOT over SEDE_BOKER tend to decrease ($-2.16\%/year$) due to a decrease in small particles (Karnieli et al., 2009), while AOT over Solar_Village show a strong increase ($+3.29\%/year$) in the weighted trend probably related to change of atmospheric conditions (e.g., increase of wind speed and relative humidity) (Sabbah and Hasan, 2008). Interestingly, there are severe differences between the unweighted and weighted trends at both stations due to a large standard error caused by high variability of ÅE and AOT. The weighted trend of CAOT of SEDE_BOKER and Solar_Village is -2.30% and $+3.39\%/year$ respectively (as seen in Fig. 9).

3.5 East Asia

Many emerging economies are found in East Asia, where, as a consequence, large amounts of anthropogenic aerosols are emitted. Additionally, mineral dust from the deserts in Mongolia and in Western and Northern China (mainly the Taklimakan and Badain Juran deserts) contributes around 70% of the total dust emissions in mid-latitude regions. Rapid desertification caused by climatic variation and human activities additionally increases the aerosol burden due to mineral dust transport (Zhang et al., 2003). ÅE and AOT at Beijing in Fig. 7 exhibit very clear seasonal cycles, which have been explained by the complex combination of natural and anthropogenic aerosols, stagnant synoptic meteorological patterns, secondary aerosol formation, and

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hygroscopic growth (Kotchenruther et al., 1999; Dubovik et al., 2001; Kim et al., 2007). Because of industrialization, urbanization, and desertification over East Asia in the last twenty years, the aerosol loading over Beijing increased rapidly and the magnitude of the weighted trend is +4.59%/year. This trend is consistent with many previous studies (Streets et al., 2000, 2003, 2006; Smith et al., 2001, 2003; Massie et al., 2004; Mishchenko and Geogdzhayev, 2007; Zhao et al., 2008). The weighted trend is larger than the unweighted one (+1.06%/year), as there are smaller weighting factors during summer (in Beijing, frequent cloud disturbance and strong mineral dust events are characteristic during summer time). Shirahama is in the middle of Japan, far from large cities, facing the Pacific Ocean (Mukai et al., 2006). Hence, maritime aerosol is predominant, but there are occurrences of mineral dust and/or industrial aerosol transported by strong westerly winds from China (Sano et al., 2003; Mukai et al., 2005). The seasonal cycles of ÅE and AOT in Fig. 7 are similar to those at Beijing due to similar meteorological conditions and aerosol sources. The upward trend of AOT is small (+0.44%/year), while FAOT increases (+2.07%/year) clearly. Due to comparatively small NO over Shirahama, only slight differences between weighted and unweighted trends of ÅE and AOT are observed all year round (see Fig. 2). The magnitude of the trends is, after classification, small for Shirahama, +1.83%/year for CAOT and +0.28%/year for FAOT. Such increase is also observed for Beijing, but is even more pronounced for weighted trends of CAOT and FAOT with increases of +5.12% and +8.72%/year, respectively.

3.6 North America

The stations GSFC and MD_Science_Center are located on urban and built-up land, while Sevilleita is positioned at shrub land over North America (Liu et al., 2004). Therefore, they basically measure aerosols from different sources. The main aerosol type measured at GSFC and MD_Science_Center is due to urban-industrial pollution, mainly due to vehicles and industries. The seasonal cycles of ÅE and AOT in Fig. 7 demonstrate that the variabilities are strongly dependent on the combination of

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natural and anthropogenic aerosols, fuel types, emission characteristic, relative humidity, boundary layer depth, and scavenging by precipitation (Glen et al., 1996; Chen et al., 2001; Dubovik et al., 2001; Andronache, 2004). The negative trends of AOT (−0.54 %/year at GSFC and −0.88 %/year at MD_Science_Center) are consistent with the decrease of industrial emissions in the United States of America (Smith et al., 2001; Streets et al., 2006; Zhao et al., 2008).

Sevilleta station measures a relatively small aerosol loading for the considered time span. The weighted trends are strongly negative (−1.66 %/year for ÅE and −3.79 %/year for AOT), clearly different from the unweighted ones (+0.43 %/year for ÅE and +3.28 %/year for AOT). In most cases of monthly averaged AOT at Sevilleta, the values are lower than 0.15, so that the classification for the trend analysis was not applied to data of this station. The majority of retrieved aerosol type at GSFC and MD_Science_Center is fine-mode, and the weighted trends of FAOT are −0.45 % and −0.18 %/year, respectively (see Fig. 9).

3.7 Mauna_Loa

Aerosols measured at Mauna_Loa (alt. ~3397 m) in the Pacific are representative for free tropospheric aerosols. However, the stations is also under some influence of long-range transport of Asian mineral dust and pollution (Dubovik et al., 2001). The free troposphere is characterized by being almost cloud-free in the subsiding branch of the Hadley cell as well as a pathway for long-distance transport of aerosols (Garstang and Fitzjarrald, 1999; Schmeissner et al., 2011). In most cases, free tropospheric AOT (440 nm) does not exceed values of 0.05 except when affected by volcano eruption or transported mineral dust and pollution. Therefore, it is difficult to analyze size and type, as the error of ÅE is larger than 30 % with AOTs smaller than 0.15 (Gobbi et al., 2007). The main factors affecting the seasonal pattern of AOT are most likely long-range transported aerosols and seasonal meteorological conditions; the AOT trend for this station is positive (+1.73 %/year).

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The relative weighted trends of ÅE (440–870 nm), AOT, CAOT, and FAOT (440 nm) in percent for the above described stations are indicated on the global map in Figs. 8 and 10. In general, total aerosol load tends to decrease over Western Europe and North America, while it is increasing over West and South Africa, Middle East, and East Asia.

The classification of aerosol types explains why the aerosol load changes in time. Consistent with our expectations, fine-mode dominant aerosol loading, primarily created by human activities, is decreasing over those countries having strict environmental regulations, while it is increasing over regions in emerging economies without such strict environmental regulations. Temporal variation of the loading of coarse-mode dominant aerosol depends strongly on meteorological conditions varying with climate change. In particular, those AERONET stations close to the regions where rapid desertification plays a role are characterized by a considerable increase of coarse-mode dominant aerosol. All values (i.e., average, unweighted, and weighted trends of ÅE, AOT, CAOT, and FAOT) are shown in Tables 4 and 5.

4 Summary and conclusion

In this study, long-term trends of the Ångström Exponent (ÅE) (440–870 nm), Aerosol Optical Thickness (AOT), Coarse-, and Fine-mode dominant AOTs (CAOT and FAOT) (440, 675, 870, and 1020 nm) observed at several AERONET stations have been analyzed. Firstly, suitable AERONET stations providing sufficiently long-term data series are chosen in order to make a meaningful trend analysis. Unfortunately, the research periods are different for each AERONET station due to the different observation history and condition (see Table 1 and Fig. 1). Weighted trends have been derived utilizing monthly standard deviation and Number of Observations (NO) providing an estimate of trend uncertainty (primarily) due to cloud disturbance. For example, if there was a high variability of aerosol loading for a small NO, then significant difference between unweighted and weighted trends is observed.

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In general, cloud-free aerosol trends in this study are consistent with results from other papers (Smith et al., 2001; Streets et al., 2006; Zhao et al., 2008; Evan et al., 2007; Mishchenko and Geogdzhayev, 2007; Mishchenko et al., 2007; Chiapello and Moulin, 2002; Chiapello et al., 2005; Karnieli et al., 2009). The aerosol classification using ÅE and ÅED from AERONET spectral observations has been useful to investigate trends of Coarse- and Fine-mode dominant AOTs. The aerosol load is increasing over the stations in/near emerging economies or biomass burning regions and decreasing over the countries under strict environmental regulations. After applying classification, these trends are more apparent. Except Western Europe and North America, most of the considered regions have positive trends of fine-mode dominant aerosol. In particular, the weighted trends of both CAOT and FAOT at Beijing over East Asia show considerable increases. Due to the high density of population in many strongly polluted areas (e.g., 1300 people per 1 km² in Beijing) and the correlation between aerosol load and mortality (Foster and Kumar, 2011), there is an urgent need for measures to reduce the aerosol load in large urban agglomerations (aka megacities) worldwide similar to those already introduced in Western Europe and North America.

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Table 1. Geolocations and research periods of the suitable AERONET stations for aerosol trend analysis in alphabetical order.

Selected AERONET Stations	Regions	Countries	Geolocations (lat.[°]/lon.[°]/ alt.[m])	Research Periods
(a) Avignon	Western Europe	France	43.93/4.88/32	2001~2005
(b) Banizoumbou	West Africa	Niger	13.54/2.66/250	2002 ~ 2008
(c) Beijing	East Asia	China	39.98/116.38/92	2003 ~ 2007
(d) Dakar	West Africa	Senegal	14.39/- 16.96/0	2004 ~ 2008
(e) GSFC	North America	USA	38.99/- 76.84/87	1995 ~ 2008
(f) Ispra	Western Europe	Italy	45.80/8.63/235	2001 ~ 2007
(g) Mauna_Loa	Free troposphere (Pacific)	USA	19.54/- 155.58/3397	1998 ~ 2009
(h) MD_Science_Center	North America	USA	39.28/- 76.62/15	2000 ~ 2006
(i) Mongu	South Africa	Zambia	-15.25/23.15/1107	2000 ~ 2004
(j) Ouagadougou	West Africa	Burkina Faso	12.20/- 1.40/290	2000 ~ 2004
(k) SEDE_BOKER	Middle East	Israel	30.86/34.78/480	2003 ~ 2008
(l) Sevillea	North America	USA	34.35/- 106.89/1477	1998 ~ 2002
(m) Shirahama	East Asia	Japan	33.69/135.36/10	2003 ~ 2009
(n) Skukuza	South Africa	South Africa	-24.99/31.59/150	2000 ~ 2007
(o) Solar_Village	Middle East	Saudi Arabia	24.91/46.40/764	2001 ~ 2007

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Table 2. Bimodal lognormal volume size distribution ($\frac{dV(r)}{d\ln r}$) parameters and refractive indices (Schuster et al., 2006) used to compute ÅE (440–870 nm) and ÅED (ÅE(440–675 nm)–ÅE(675–870 nm)) using Mie code in Fig. 4.

Parameter*	Values
r_{fine}	0.06, 0.09, 0.12, 0.15, 0.18, 0.21, 0.24, 0.27, 0.30
σ_{fine}	0.38, 0.50
r_{coarse}	1.9, 2.2, 2.7, 2.8, 3.0, 3.2, 3.4, 3.6, 3.7
σ_{coarse}	0.75, 1.00
n	1.34, 1.37, 1.40, 1.43, 1.47, 1.50, 1.54
k	0.003
$C_{\text{fine}}/C_{\text{total}}$	0.01, 0.10, 0.20, 0.30, 0.40, 0.50, 0.60, 0.70, 0.80, 0.90, 0.99

* The bimodal lognormal volume size distribution ($\frac{dV(r)}{d\ln r}$) is given by

$$\frac{dV(r)}{d\ln r} = \frac{C_{\text{fine}}}{\sqrt{2\pi}\sigma_{\text{fine}}} \exp\left[-\frac{(\ln r - \ln r_{\text{fine}})^2}{2\sigma_{\text{fine}}^2}\right] + \frac{C_{\text{coarse}}}{\sqrt{2\pi}\sigma_{\text{coarse}}} \exp\left[-\frac{(\ln r - \ln r_{\text{coarse}})^2}{2\sigma_{\text{coarse}}^2}\right],$$

where $C_{\text{total, fine, coarse}}$ represents the particle volume concentration for total, fine and coarse aerosol modes [$\mu\text{m}^3/\mu\text{m}^2$], $r_{\text{fine, coarse}}$ is the median or geometric mean radius [μm], and $\sigma_{\text{fine, coarse}}$ is the variance or width of each mode. n and k represent the real and imaginary parts of the complex refractive index, respectively.

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Table 3. Summary of aerosol optical properties for urban-industrial and mixed, biomass burning, desert dust, and oceanic types from Dubovik et al (2001) based on worldwide AERONET of ground-based radiometers. These properties are used to compute ÅE (440–870 nm) and ÅED (ÅE(440–675 nm)–ÅE(675–870 nm)) using Mie code and depicted by solid, dotted, dashed, dash-dot-dot lines sequentially in Fig. 5.

Urban-industrial and mixed	GSFC, Greenbelt, MD (1993–2000)	Crete-Paris, France (1999)
Range of optical thickness; $\langle \tau \rangle^*$	$0.1 \leq \tau(440) \leq 1.0$; 0.24	$0.1 \leq \tau(440) \leq 0.9$; 0.26
Refractive indices(n ; k)	1.41-0.03 $\tau(440)$; 0.003	1.40; 0.009
$r_{\text{fine}}[\mu\text{m}]$; σ_{fine}	$0.12+0.11 \tau(440)$; 0.38	$0.11+0.13 \tau(440)$; 0.43
$r_{\text{coarse}}[\mu\text{m}]$; σ_{coarse}	$3.03+0.49 \tau(440)$; 0.75	$2.76+0.48 \tau(440)$; 0.79
$C_{\text{fine}}[\mu\text{m}^3/\mu\text{m}^2]$	$0.15 \tau(440)$	$0.01+0.12 \tau(440)$
$C_{\text{coarse}}[\mu\text{m}^3/\mu\text{m}^2]$	$0.01+0.04 \tau(440)$	$0.01+0.05 \tau(440)$
	Mexico City Maldives (INDOEX) (1999–2000)	(1999–2000)
Range of optical thickness; $\langle \tau \rangle$	$0.1 \leq \tau(440) \leq 1.8$; 0.43	$0.1 \leq \tau(440) \leq 0.7$; 0.27
Refractive indices(n ; k)	1.47; 0.014	1.44; 0.007
$r_{\text{fine}}[\mu\text{m}]$; σ_{fine}	$0.12+0.04 \tau(440)$; 0.43	0.18; 0.46
$r_{\text{coarse}}[\mu\text{m}]$; σ_{coarse}	$2.72+0.60 \tau(440)$; 0.63	$2.62+0.61 \tau(440)$; 0.76
$C_{\text{fine}}[\mu\text{m}^3/\mu\text{m}^2]$	$0.12 \tau(440)$	$0.12 \tau(440)$
$C_{\text{coarse}}[\mu\text{m}^3/\mu\text{m}^2]$	$0.11 \tau(440)$	$0.15 \tau(440)$

* $\langle \tau \rangle$ is the average value of Aerosol Optical Thickness (AOT) at λ nm ($\tau(\lambda)$).

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Table 3. Continued.

	Amazonian forest, Brazil (1993–1994); Bolivia (1998–1999)	South American cerrado, Brazil (1993–1995)
Biomass burning		
Range of optical thickness; $\langle \tau \rangle^*$	$0.1 \leq \tau(440) \leq 3.0$; 0.74	$0.1 \leq \tau(440) \leq 2.1$; 0.80
Refractive indices(n ; k)	1.47; 0.00093	1.52; 0.015
$r_{\text{fine}}[\mu\text{m}]$; σ_{fine}	$0.14+0.13 \tau(440)$; 0.40	$0.14+0.01 \tau(440)$; 0.47
$r_{\text{coarse}}[\mu\text{m}]$; σ_{coarse}	$3.27+0.58 \tau(440)$; 0.79	$3.27+0.51 \tau(440)$; 0.79
$C_{\text{fine}}[\mu\text{m}^3/\mu\text{m}^2]$	$0.12\tau(440)$	$0.1\tau(440)$
$C_{\text{coarse}}[\mu\text{m}^3/\mu\text{m}^2]$	$0.05\tau(440)$	$0.04+0.03 \tau(440)$
	African savanna, Zambia (1995–2000)	Boreal forest, United States and Canada (1994–1998)
Range of optical thickness; $\langle \tau \rangle$	$0.1 \leq \tau(440) \leq 1.5$; 0.38	$0.1 \leq \tau(440) \leq 2.0$; 0.40
Refractive indices(n ; k)	1.51; 0.021	1.50; 0.0094
$r_{\text{fine}}[\mu\text{m}]$; σ_{fine}	$0.12+0.025 \tau(440)$; 0.40	$0.15+0.015 \tau(440)$; 0.43
$r_{\text{coarse}}[\mu\text{m}]$; σ_{coarse}	$3.22+0.71 \tau(440)$; 0.73	$3.21+0.2 \tau(440)$; 0.81
$C_{\text{fine}}[\mu\text{m}^3/\mu\text{m}^2]$	$0.12\tau(440)$	$0.01+0.1 \tau(440)$
$C_{\text{coarse}}[\mu\text{m}^3/\mu\text{m}^2]$	$0.09\tau(440)$	$0.01+0.03 \tau(440)$

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Table 3. Continued.

Desert dust and oceanic	Bahrain-Persian Gulf (1998–2000)	Solar-Vil.-Saudi Arabia (1998–2000)
Range of optical thickness; $\langle \tau \rangle^*$	$0.1 \leq \tau(1020) \leq 1.2$; 0.22	$0.1 \leq \tau(1020) \leq 1.5$; 0.17
Refractive indices(n)	1.55	1.56
Refractive indices($k(440/670/870/1020$ nm))	0.0025/0.0014/0.001/0.001	0.0029/0.0013/0.001/0.001
$r_{\text{fine}}[\mu\text{m}]$; σ_{fine}	0.15; 0.42	0.12; 0.40
$r_{\text{coarse}}[\mu\text{m}]$; σ_{coarse}	2.54; 0.61	2.32; 0.60
$C_{\text{fine}}[\mu\text{m}^3/\mu\text{m}^2]$	$0.02+0.1 \tau(1020)$	$0.02+0.02 \tau(1020)$
$C_{\text{coarse}}[\mu\text{m}^3/\mu\text{m}^2]$	$-0.02+0.92 \tau(1020)$	$-0.02+0.98 \tau(1020)$
	Cape Verde (1993–2000)	Lanai, HI (1995–2000)
Range of optical thickness; $\langle \tau \rangle$	$0.1 \leq \tau(1020) \leq 2.0$; 0.39	$0.01 \leq \tau(1020) \leq 0.2$; 0.04
Refractive indices(n)	1.48	1.36
Refractive indices($k(440/670/870/1020$ nm))	0.0025/0.0007/0.0006/0.0006	0.0015
$r_{\text{fine}}[\mu\text{m}]$; σ_{fine}	0.12; $0.49+0.10 \tau(1020)$	0.16; 0.48
$r_{\text{coarse}}[\mu\text{m}]$; σ_{coarse}	1.90; $0.63-0.10 \tau(1020)$	2.70; 0.68
$C_{\text{fine}}[\mu\text{m}^3/\mu\text{m}^2]$	$0.02+0.02 \tau(1020)$	$0.40\tau(1020)$
$C_{\text{coarse}}[\mu\text{m}^3/\mu\text{m}^2]$	$0.9\tau(1020)$	$0.80\tau(1020)$

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Table 4. Climatological averages, and statistical parameters for weighted and unweighted trend of Ångström Exponent (440–870 nm) and AOTs (440, 675, 870, and 1020 nm) at (a) Avignon, (b) Banizoumbou, (c) Beijing, (d) Dakar, (e) GSFC, (f) Ispra, (g) Mauna.Loa, (h) MD.Science_Center, (i) Mongu, (j) Ouagadougou, (k) SEDE_BOKER, (l) Sevillea, (m) Shira-hama, (n) Skukuza, and (o) Solar_Village.

(a) Avignon			(b) Banizoumbou			
	$\langle \tau \rangle^a$	Unweighted trend[/year]	Weighted trend[/year]	$\langle \tau \rangle$	Unweighted trend[/year]	Weighted trend[/year]
α^b	1.43	+0.02242(+1.57%)	+0.01364(+0.95%)	0.37	-0.00777(-2.08%)	-0.01130(-3.03%)
τ_{440}^c	0.20	+0.00020(+0.10%)	+0.00194(+0.98%)	0.52	-0.00166(-0.32%)	+0.00117(+0.22%)
τ_{675}^c	0.11	-0.00024(-0.22%)	+0.00037(+0.34%)	0.47	+0.00038(+0.08%)	+0.00353(+0.76%)
τ_{870}^c	0.08	-0.00066(-0.85%)	-0.00079(-1.02%)	0.43	+0.00119(+0.28%)	+0.00437(+1.01%)
τ_{1020}^c	0.06	+0.00021(+0.33%)	-0.00035(-0.55%)	0.41	+0.00056(+0.14%)	+0.00347(+0.85%)
(c) Beijing			(d) Dakar			
α	1.12	-0.02200(-1.97%)	-0.02308(-2.07%)	0.36	-0.01415(-3.95%)	-0.03264(-9.12%)
τ_{440}	0.68	+0.00717(+1.06%)	+0.03100(+4.59%)	0.46	-0.01186(-2.60%)	-0.00710(-1.56%)
τ_{675}	0.43	+0.00636(+1.48%)	+0.02448(+5.71%)	0.41	-0.00938(-2.31%)	-0.00251(-0.62%)
τ_{870}	0.32	+0.00665(+2.06%)	+0.02023(+6.26%)	0.38	-0.00697(-1.86%)	+0.00104(+0.28%)
τ_{1020}	0.27	+0.00673(+2.46%)	+0.01706(+6.25%)	0.36	-0.00649(-1.83%)	+0.00218(+0.61%)

$\langle \tau \rangle^a$: Average of Aerosol Optical Thickness at λ nm (τ_λ).

α^b : Ångström Exponent (440–870 nm).

τ_λ^c : AOTs at wavelegnth, λ (440, 675, 870, and 1020 nm).

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Table 4. Continued.

(e) GSFC			(f) Ispra			
	$\langle \tau \rangle^a$	Unweighted trend[/year]	Weighted trend[/year]	$\langle \tau \rangle$	Unweighted trend[/year]	Weighted trend[/year]
α^b	1.59	+0.00816(+0.51%)	+0.00442(+0.28%)	1.51	-0.00038(-0.03%)	-0.00178(-0.12%)
τ_{440}^c	0.22	-0.00085(-0.38%)	-0.00122(-0.54%)	0.30	-0.00337(-1.13%)	-0.00687(-2.30%)
τ_{675}^c	0.11	-0.00095(-0.85%)	-0.00074(-0.67%)	0.16	-0.00206(-1.26%)	-0.00418(-2.57%)
τ_{870}^c	0.07	-0.00080(-1.07%)	-0.00037(-0.50%)	0.11	-0.00101(-0.90%)	-0.00262(-2.34%)
τ_{1020}^c	0.06	-0.00062(-1.05%)	-0.00020(-0.34%)	0.09	-0.00094(-1.06%)	-0.00266(-2.99%)
(g) Mauna.Loia			(h) MD.Science.Center			
α	1.21	+0.01180(+0.97%)	+0.01504(+1.24%)	1.68	-0.01228(-0.73%)	-0.01480(-0.88%)
τ_{440}	0.02	+0.00026(+1.36%)	+0.00033(+1.73%)	0.24	-0.00177(-0.74%)	-0.00003(-0.01%)
τ_{675}	0.01	+0.00003(+0.39%)	+0.00009(+1.11%)	0.12	-0.00089(-0.75%)	-0.00049(-0.41%)
τ_{870}	0.01	+0.00004(+0.46%)	+0.00014(+1.41%)	0.08	-0.00020(-0.26%)	+0.00010(+0.13%)
τ_{1020}	0.01	+0.00003(+0.28%)	+0.00008(+0.91%)	0.06	-0.00053(-0.83%)	-0.00083(-1.30%)

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Table 4. Continued.

(i) Mongu			(j) Ouagadougou			
	$\langle \tau \rangle^a$	Unweighted trend[/year]	Weighted trend[/year]	$\langle \tau \rangle$	Unweighted trend[/year]	Weighted trend[/year]
α^b	1.53	+0.01601(+1.05%)	-0.00122(-0.08%)	0.42	+0.01331(+3.14%)	+0.01695(+4.00%)
τ_{440}^c	0.28	+0.01580(+5.70%)	+0.00625(+2.26%)	0.51	+0.01747(+3.42%)	-0.00996(-1.95%)
τ_{675}^c	0.14	+0.00592(+4.30%)	+0.00671(+4.87%)	0.47	-0.00367(-0.78%)	-0.01168(-2.48%)
τ_{870}^c	0.09	+0.00370(+3.99%)	+0.00736(+7.94%)	0.41	+0.01427(+3.48%)	-0.01028(-2.51%)
τ_{1020}^c	0.07	+0.00175(+2.49%)	+0.00582(+8.29%)	0.38	+0.01691(+4.40%)	-0.00720(-1.87%)
(k) SEDE.BOKER			(l) Sevilleta			
α	0.88	+0.00611(+0.69%)	-0.02455(-2.78%)	1.19	+0.00516(+0.43%)	-0.01980(-1.66%)
τ_{440}	0.20	+0.00111(+0.56%)	-0.00427(-2.16%)	0.08	+0.00271(+3.28%)	-0.00313(-3.79%)
τ_{675}	0.14	-0.00007(-0.05%)	-0.00361(-2.60%)	0.05	+0.00096(+1.98%)	-0.00162(-3.35%)
τ_{870}	0.12	+0.00055(+0.45%)	-0.00163(-1.33%)	0.04	+0.00115(+3.01%)	-0.00095(-2.49%)
τ_{1020}	0.11	-0.00032(-0.30%)	-0.00112(-1.05%)	0.03	+0.00124(+3.65%)	+0.00057(+1.68%)

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Table 4. Continued.

	(m) Shirahama			(n) Skukuza		
	$\langle \tau \rangle^a$	Unweighted trend[/year]	Weighted trend[/year]	$\langle \tau \rangle$	Unweighted trend[/year]	Weighted trend[/year]
α^b	1.27	+0.01735(+1.37%)	+0.02617(+2.07%)	1.34	+0.00566(+0.42%)	-0.01080(-0.80%)
τ_{440}^c	0.31	+0.00082(+0.27%)	+0.00137(+0.44%)	0.23	-0.00113(-0.48%)	+0.00118(+0.51%)
τ_{675}^c	0.18	-0.00111(-0.60%)	-0.00012(-0.06%)	0.12	-0.00126(-1.01%)	-0.00037(-0.29%)
τ_{870}^c	0.13	-0.00030(-0.22%)	-0.00006(-0.04%)	0.09	-0.00034(-0.39%)	+0.00006(+0.07%)
τ_{1020}^c	0.11	-0.00150(-1.35%)	-0.00165(-1.49%)	0.07	-0.00039(-0.54%)	-0.00027(-0.37%)
	(o) Solar Village					
α	0.55	-0.02293(-4.16%)	-0.00492(-0.89%)			
τ_{440}	0.31	+0.01881(+6.12%)	+0.01009(+3.29%)			
τ_{675}	0.25	+0.01684(+6.62%)	+0.00541(+2.13%)			
τ_{870}	0.23	+0.01705(+7.35%)	+0.00503(+2.17)			
τ_{1020}	0.23	+0.01373(+6.03%)	+0.00140(+0.62%)			

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Table 5. Climatological averages and statistical parameters for weighted and unweighted trend of Coarse- and Fine-mode dominant AOTs (440, 675, 870, and 1020 nm) at (a) Avignon, (b) Banizoumbou, (c) Beijing, (d) Dakar, (e) GSFC, (f) Ispra, (h) MD_Science_Center, (i) Mongu, (j) Ouagadougou, (k) SEDE_BOKER, (m) Shirahama, (n) Skukuza, and (o) Solar_Village.

	Coarse-mode dominant aerosol			Fine-mode dominant aerosol		
	$\langle \tau \rangle$	Unweighted trend[/year]	Weighted trend[/year]	$\langle \tau \rangle$	Unweighted trend[/year]	Weighted trend[/year]
(a) Avignon						
τ_{440}	0.15	+0.00364(+2.51%)	+0.00313(+2.16%)	0.18	-0.00026(-0.14%)	+0.00043(+0.24%)
τ_{675}	0.09	+0.00207(+2.42%)	+0.00128(+1.50%)	0.09	-0.00058(-0.62%)	-0.00021(-0.23%)
τ_{870}	0.07	+0.00122(+1.82%)	+0.00053(+0.79%)	0.06	-0.00095(-1.46%)	-0.00112(-1.73%)
τ_{1020}	0.06	+0.00184(+3.14%)	+0.00100(+1.71%)	0.05	-0.00006(-0.11%)	-0.00071(-1.38%)
(b) Banizoumbou						
τ_{440}	0.52	-0.00160(-0.31%)	+0.00149(+0.27%)	–	–	–
τ_{675}	0.47	+0.00040(+0.08%)	+0.00377(+0.80%)	–	–	–
τ_{870}	0.43	+0.00118(+0.27%)	+0.00462(+1.07%)	–	–	–
τ_{1020}	0.41	+0.00055(+0.13%)	+0.00374(+0.91%)	–	–	–
(c) Beijing						
τ_{440}	0.51	-0.00099(-0.19%)	+0.02598(+5.12%)	0.70	+0.02039(+2.90%)	+0.06135(+8.72%)
τ_{675}	0.35	+0.00042(+0.12%)	+0.01618(+4.65%)	0.42	+0.01235(+2.97%)	+0.03019(+7.26%)
τ_{870}	0.28	+0.00137(+0.49%)	+0.01189(+4.25%)	0.29	+0.00921(+3.15%)	+0.02000(+6.85%)
τ_{1020}	0.25	+0.00157(+0.64%)	+0.00979(+3.97%)	0.23	+0.00769(+3.27%)	+0.01420(+6.05%)

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Table 5. Continued.

	Coarse-mode dominant aerosol			Fine-mode dominant aerosol		
	$\langle \tau \rangle$	Unweighted trend[/year]	Weighted trend[/year]	$\langle \tau \rangle$	Unweighted trend[/year]	Weighted trend[/year]
(d) Dakar						
τ_{440}	0.46	-0.01223(-2.69 %)	-0.00669(-1.47 %)	–	–	–
τ_{675}	0.41	-0.00990(-2.44 %)	-0.00225(-0.56 %)	–	–	–
τ_{870}	0.38	-0.00754(-2.01 %)	+0.00121(+0.32 %)	–	–	–
τ_{1020}	0.36	-0.00708(-1.99 %)	+0.00231(+0.65 %)	–	–	–
(e) GSFC						
τ_{440}	0.14	-0.00340(-2.51 %)	-0.00027(-0.20 %)	0.22	-0.00025(-0.11 %)	-0.00100(-0.45 %)
τ_{675}	0.07	-0.00293(-3.94 %)	-0.00014(-0.18 %)	0.11	-0.00040(-0.37 %)	-0.00071(-0.66 %)
τ_{870}	0.06	-0.00260(-4.47 %)	+0.00011(+0.19 %)	0.07	-0.00024(-0.35 %)	-0.00040(-0.56 %)
τ_{1020}	0.05	-0.00231(-4.59 %)	+0.00029(+0.57 %)	0.05	-0.00007(-0.14 %)	-0.00023(-0.42 %)
(f) Ispra						
τ_{440}	0.16	+0.00389(+2.36 %)	-0.00174(-1.05 %)	0.29	-0.00324(-1.12 %)	-0.00662(-2.30 %)
τ_{675}	0.10	+0.00166(+1.67 %)	-0.00257(-2.58 %)	0.15	-0.00183(-1.20 %)	-0.00426(-2.80 %)
τ_{870}	0.08	+0.00149(+1.90 %)	-0.00151(-1.92 %)	0.10	-0.00077(-0.76 %)	-0.00236(-2.33 %)
τ_{1020}	0.07	+0.00113(+1.65 %)	-0.00151(-2.20 %)	0.08	-0.00069(-0.87 %)	-0.00202(-2.58 %)

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Table 5. Continued.

	Coarse-mode dominant aerosol			Fine-mode dominant aerosol		
	$\langle \tau \rangle$	Unweighted trend[/year]	Weighted trend[/year]	$\langle \tau \rangle$	Unweighted trend[/year]	Weighted trend[/year]
(h) MD_Science_Center						
τ_{440}	0.14	+0.00073(+0.52 %)	+0.00202(+1.45 %)	0.24	-0.00135(-0.56 %)	-0.00044(-0.18 %)
τ_{675}	0.07	+0.00031(+0.42 %)	+0.00076(+1.01 %)	0.12	-0.00058(-0.49 %)	-0.00003(-0.03 %)
τ_{870}	0.06	+0.00057(+1.02 %)	+0.00106(+1.90 %)	0.08	+0.00006(+0.08 %)	+0.00062(+0.83 %)
τ_{1020}	0.05	+0.00001(+0.03 %)	+0.00017(+0.33 %)	0.06	-0.00029(-0.49 %)	-0.00041(-0.68 %)
(i) Mongu						
τ_{440}	0.20	-0.00608(-3.01 %)	-0.00037(-0.18 %)	0.27	+0.01648(+6.13 %)	+0.00473(+1.76 %)
τ_{675}	0.12	-0.00475(-4.07 %)	+0.00042(+0.36 %)	0.13	+0.00641(+4.94 %)	+0.00533(+4.10 %)
τ_{870}	0.09	-0.00338(-3.69 %)	+0.00144(+1.57 %)	0.08	+0.00415(+4.91 %)	+0.00588(+6.95 %)
τ_{1020}	0.08	-0.00378(-4.93 %)	+0.00109(+1.43 %)	0.06	+0.00223(+3.59 %)	+0.00404(+6.51 %)
(j) Ouagadougou						
τ_{440}	0.51	+0.01764(+3.44 %)	-0.00953(-1.86 %)	–	–	–
τ_{675}	0.47	-0.00333(-0.71 %)	-0.01259(-2.67 %)	–	–	–
τ_{870}	0.41	+0.01469(+3.57 %)	-0.01133(-2.76 %)	–	–	–
τ_{1020}	0.39	+0.01734(+4.50 %)	-0.00830(-2.15 %)	–	–	–



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Table 5. Continued.

	Coarse-mode dominant aerosol			Fine-mode dominant aerosol		
	$\langle \tau \rangle$	Unweighted trend[/year]	Weighted trend[/year]	$\langle \tau \rangle$	Unweighted trend[/year]	Weighted trend[/year]
(k) SEDE.BOKER						
τ_{440}	0.19	+0.00162(+0.83 %)	-0.00445(-2.30 %)	0.13	-0.00014(-0.11 %)	-0.00146(-1.12 %)
τ_{675}	0.14	+0.00021(+0.15 %)	-0.00452(-3.26 %)	0.08	-0.00016(-0.21 %)	-0.00136(-1.75 %)
τ_{870}	0.12	+0.00075(+0.61 %)	-0.00305(-2.46 %)	0.07	+0.00083(+1.27 %)	+0.00013(+0.20 %)
τ_{1020}	0.11	-0.00030(-0.27 %)	-0.00342(-3.15 %)	0.05	+0.00025(+0.49 %)	+0.00040(+0.78 %)
(m) Shirahama						
τ_{440}	0.23	+0.00604(+2.62 %)	+0.00423(+1.83 %)	0.31	+0.00137(+0.44 %)	+0.00088(+0.28 %)
τ_{675}	0.15	+0.00190(+1.29 %)	+0.00210(+1.43 %)	0.17	-0.00100(-0.57 %)	-0.00032(-0.18 %)
τ_{870}	0.12	+0.00165(+1.40 %)	+0.00205(+1.74 %)	0.12	-0.00035(-0.29 %)	-0.00052(-0.42 %)
τ_{1020}	0.10	-0.00006(-0.06 %)	+0.00047(+0.47 %)	0.10	-0.00156(-1.64 %)	-0.00207(-2.17 %)
(n) Skukuza						
τ_{440}	0.14	+0.00119(+0.83 %)	+0.00044(+0.31 %)	0.23	-0.00113(-0.49 %)	+0.00174(+0.75 %)
τ_{675}	0.09	-0.00070(-0.80 %)	-0.00095(-1.08 %)	0.12	-0.00124(-1.03 %)	-0.00020(-0.17 %)
τ_{870}	0.07	-0.00035(-0.51 %)	-0.00005(-0.08 %)	0.08	-0.00030(-0.36 %)	-0.00001(-0.01 %)
τ_{1020}	0.06	-0.00067(-1.11 %)	-0.00039(-0.64 %)	0.07	-0.00033(-0.50 %)	-0.00034(-0.51 %)
(o) Solar_Village						
τ_{440}	0.31	+0.01888(+6.15 %)	+0.01042(+3.39 %)	–	–	–
τ_{675}	0.25	+0.01682(+6.60 %)	+0.00573(+2.25 %)	–	–	–
τ_{870}	0.23	+0.01697(+7.31 %)	+0.00530(+2.28 %)	–	–	–
τ_{1020}	0.23	+0.01367(+6.00 %)	+0.00173(+0.76 %)	–	–	–

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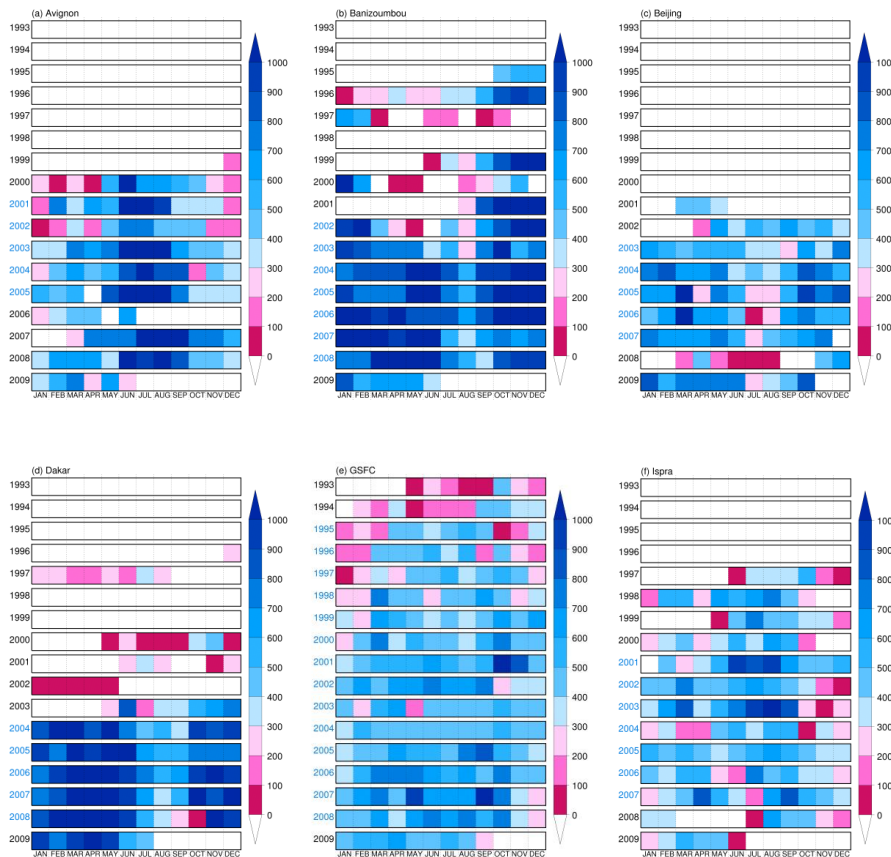


Fig. 1. The monthly observation numbers at the AEROET stations: **(a)** Avignon, **(b)** Bani-zoumbou, **(c)** Beijing, **(d)** Dakar, **(e)** GSFC, **(f)** Ispra, **(g)** Mauna_Loa, **(h)** MD_Science_Center, **(i)** Mongu, **(j)** Ouagadougou, **(k)** SEDE_BOKER, **(l)** Sevilleta, **(m)** Shirahama, **(n)** Skukuza, and **(o)** Solar_Village since 1993. The research period for each station is shown by the blue years at vertical axis.

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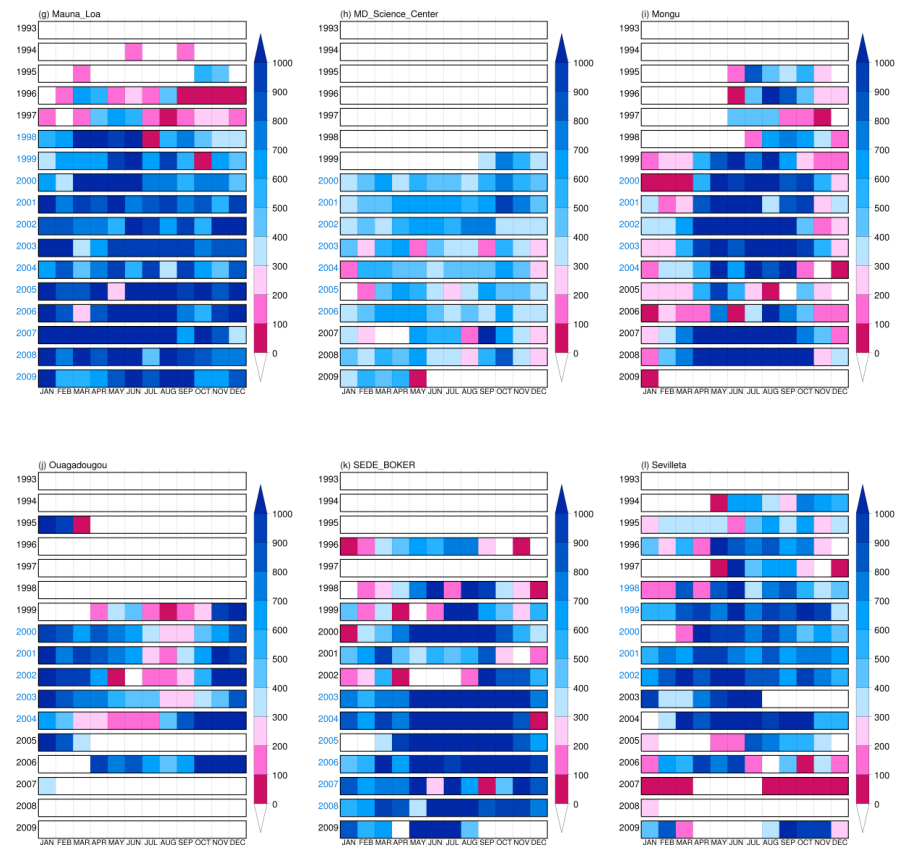


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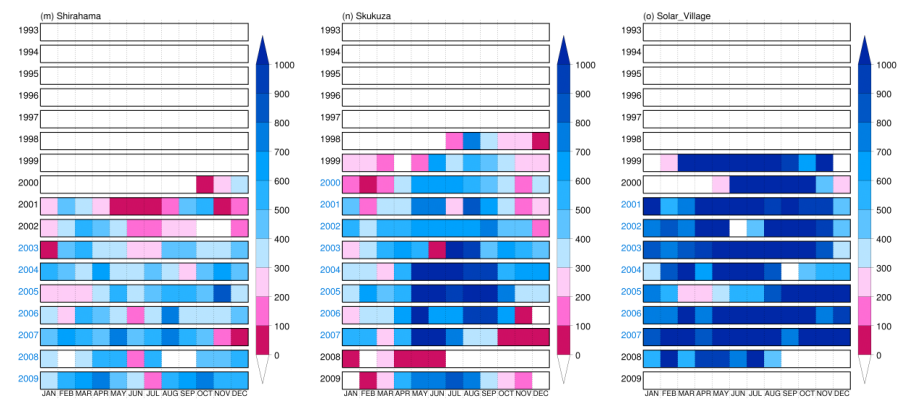


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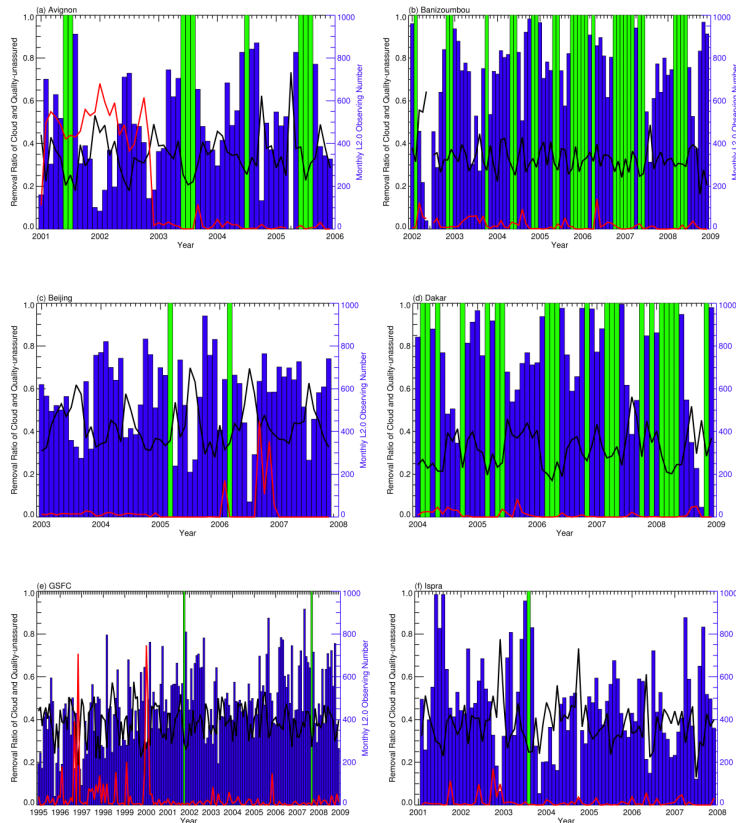


Fig. 2. The removal ratio of cloud (black line) and quality-unassured (red line) cases to Level 2.0 data (blue bar) within each of research period at the AERONET stations: **(a)** Avignon, **(b)** Bamzoumbou, **(c)** Beijing, **(d)** Dakar, **(e)** GSFC, **(f)** Ispra, **(g)** Mauna_Loa, **(h)** MD_Science_Center, **(i)** Mongu, **(j)** Ouagadougou, **(k)** SEDE_BOKER, **(l)** Sevilleta, **(m)** Shirahama, **(n)** Skukuza, and **(o)** Solar_Village. Green bars mean that the monthly observation numbers are over 1000 times.

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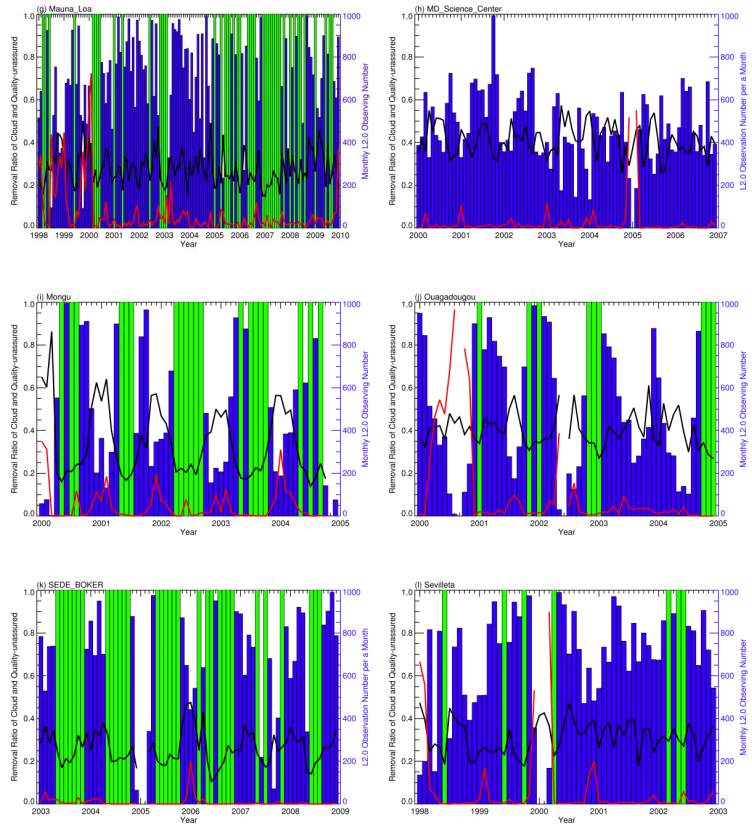


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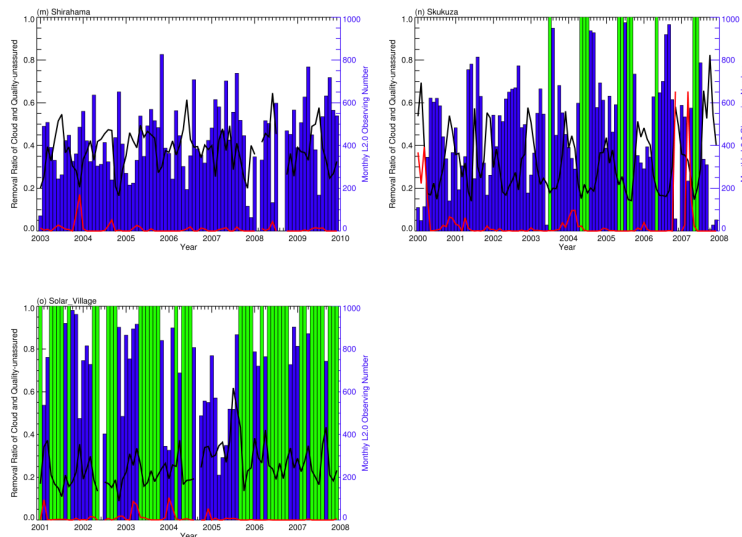


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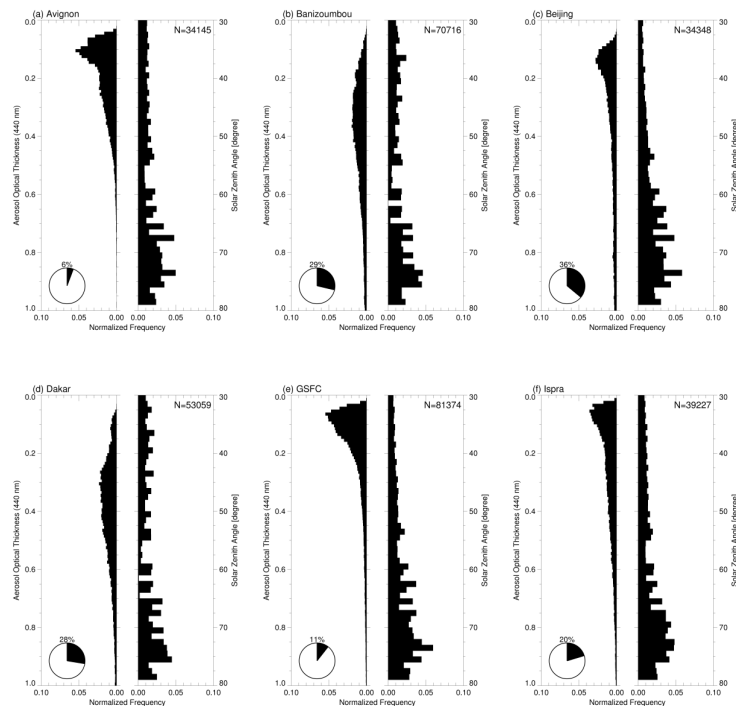


Fig. 3. Normalized frequency of Aerosol Optical Thickness at 440 nm (τ_{440}) and solar zenith angle (θ) to total observation number (N) at the AERONET stations: **(a)** Avignon, **(b)** Bani-zoumbou, **(c)** Beijing, **(d)** Dakar, **(e)** GSFC, **(f)** Ispra, **(g)** Mauna_Loa, **(h)** MD_Science_Center, **(i)** Mongu, **(j)** Ouagadougou, **(k)** SEDE_BOKER, **(l)** Sevillaeta, **(m)** Shirahama, **(n)** Skukuza, and **(o)** Solar_Village. The bin sizes for τ_{440} and θ are 0.01 and 1.0° , respectively. The circle diagram on the lower-left hand means the percentage of AERONET level 2.0 inversion data (e.g. volume size distribution and Single Scattering Albedo (SSA)) to total observations. The AERONET inversion data are provided under the criteria; $\tau_{440} > 0.4$ and $\theta > 50^\circ$.

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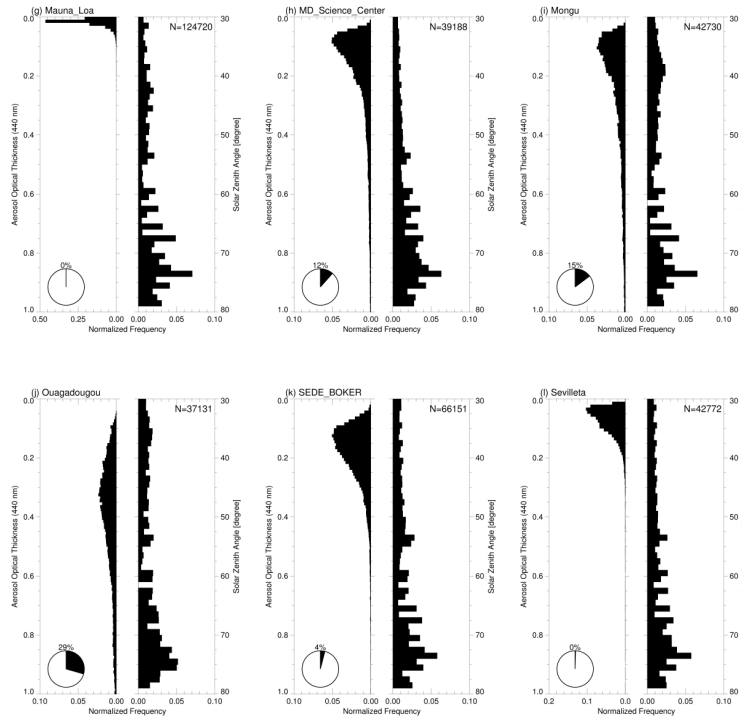


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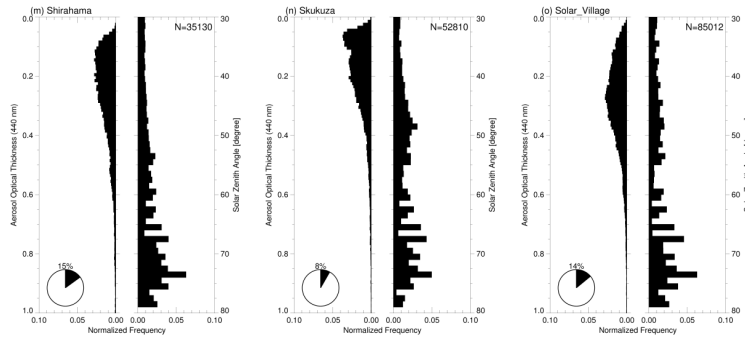


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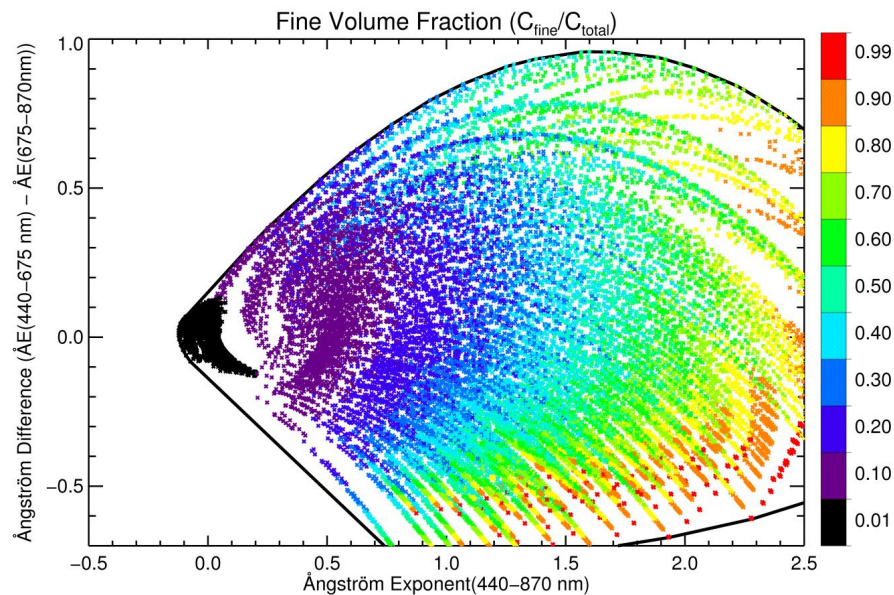


Fig. 4. Simulations of the fine volume fraction as a function of Ångström Exponent (440–870 nm) and Ångström Exponent Difference ($\text{ÅE}(440\text{--}675\text{ nm}) - \text{ÅE}(675\text{--}870\text{ nm})$) using Mie theory with all combinations of volume median radius, standard deviation, refractive indices, and fine volume fractions shown Table 2.

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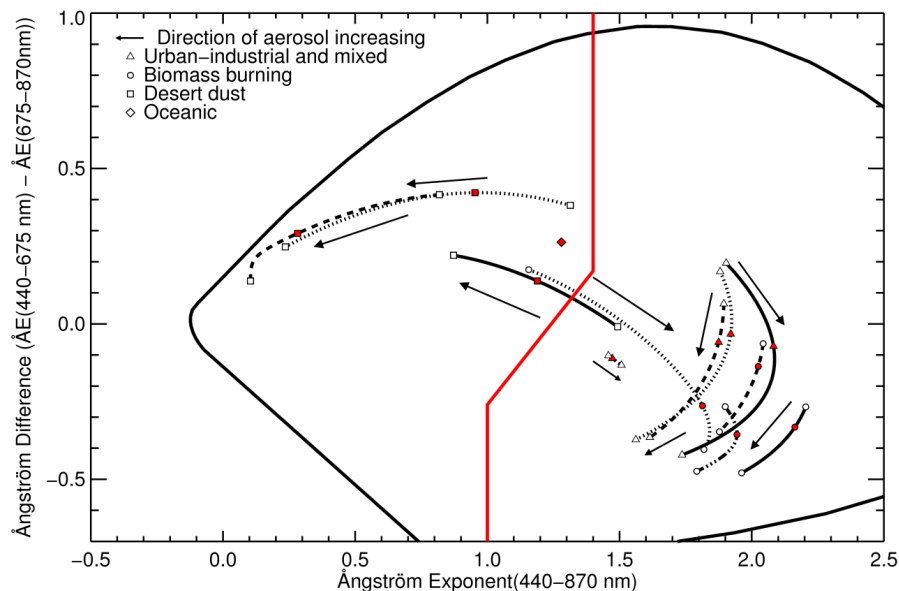


Fig. 5. Mie simulations (solid, dotted, dashed, dash-dot-dot lines) for the typical aerosols (urban-industrial and mixed, biomass burning, desert dust, oceanic) depending on an increase of aerosol loading in Dubovik et al. (2001) shown Table 3. The red spot and red line represent the simulations for AOT average of the typical aerosols and the classification line for two aerosol types (fine- and coarse-mode dominant aerosols), respectively.

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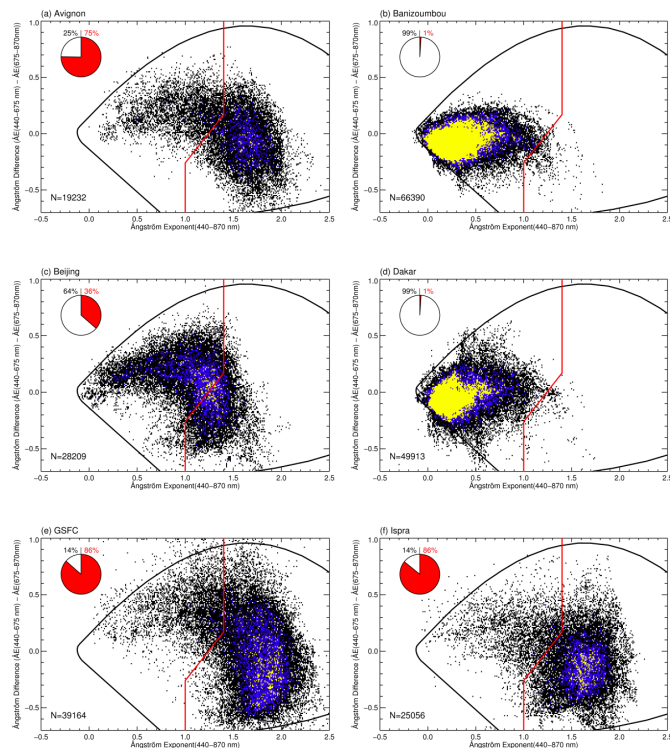


Fig. 6. Applications of the classification method to the AERONET datasets at **(a)** Avignon, **(b)** Banizoumbou, **(c)** Beijing, **(d)** Dakar, **(e)** GSFC, **(f)** Ispra, **(g)** Mauna_Loa, **(h)** MD_Science_Center, **(i)** Mongu, **(j)** Ouagadougou, **(k)** SEDE_BOKER, **(l)** Sevilleta, **(m)** Shirahama, **(n)** Skukuza, and **(o)** Solar_Village, which are characterized by two aerosol types as fine- and coarse-mode dominant aerosols. The circle diagram on the upper-left hand means the percentage of fine-(red) and coarse-mode (black) dominant aerosols to total observations (N). To avoid large errors in Ångström Exponent and Ångström Exponent Difference from low AOTs, only AERONET level 2.0 data with AOT (440 nm) > 0.15 were used.

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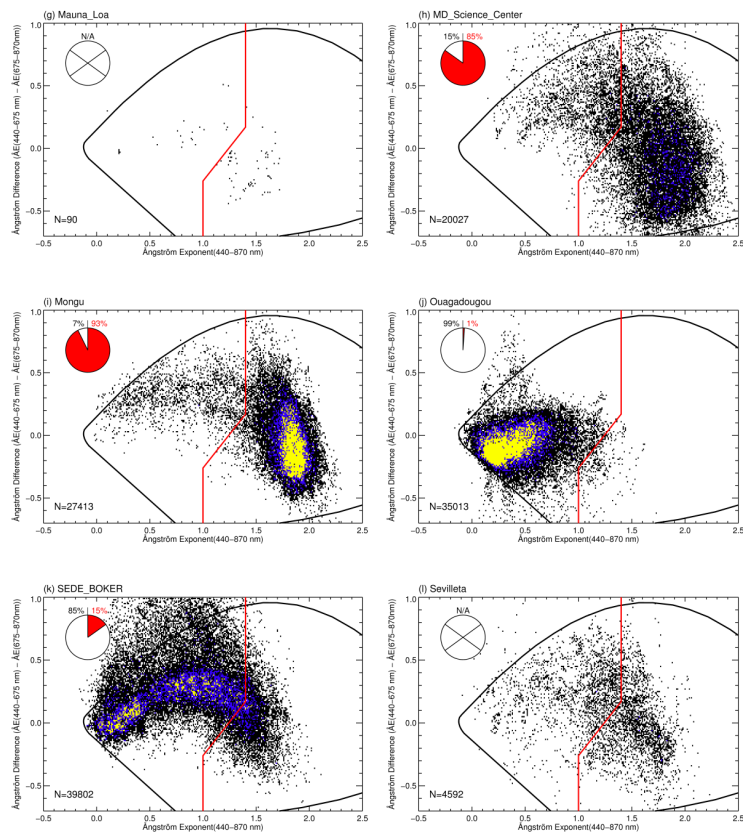


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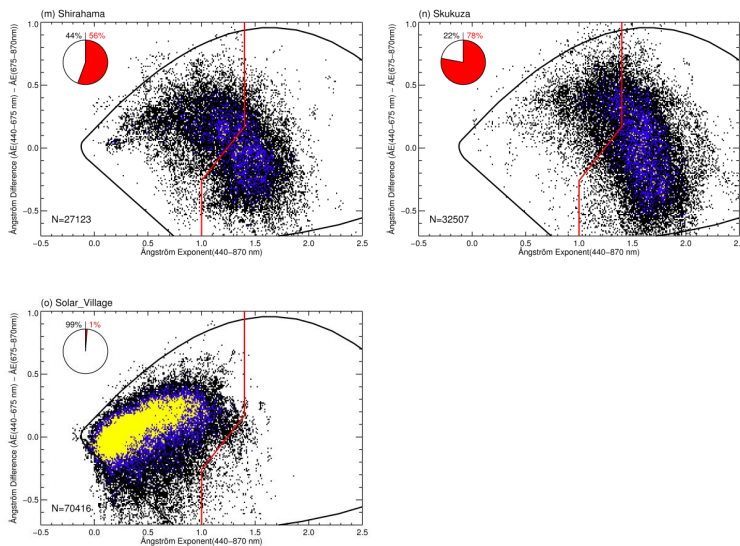


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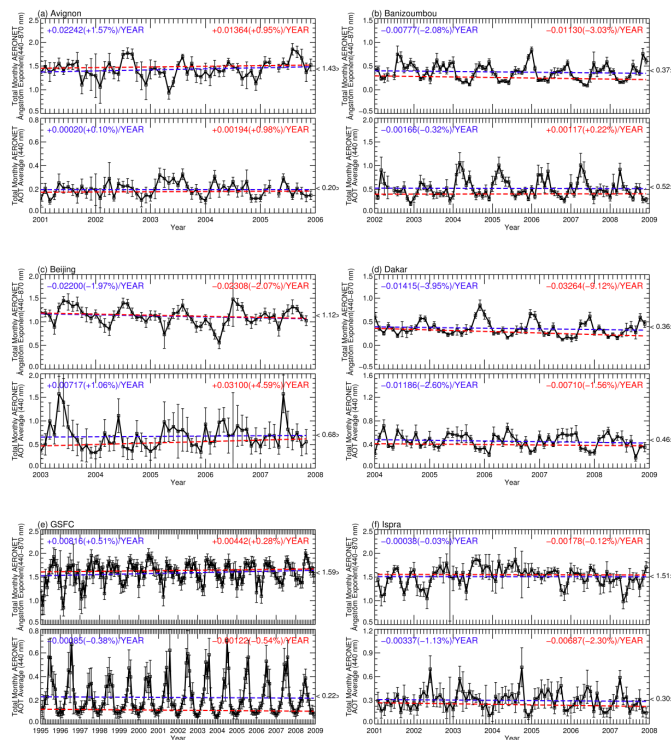


Fig. 7. Total averages (black one enclosed with parentheses at right vertical axis), temporal unweighted (blue one on the left upper part), and weighted (red one on the right upper part) trends of Ångström Exponent (440–870 nm) and AOT (440 nm) at the AERONET stations: **(a)** Avignon, **(b)** Banizoumbou, **(c)** Beijing, **(d)** Dakar, **(e)** GSFC, **(f)** Ispra, **(g)** Mauna_Loa, **(h)** MD_Science_Center, **(i)** Mongu, **(j)** Ouagadougou, **(k)** SEDE_BOKER, **(l)** Sevilleta, **(m)** Shirahama, **(n)** Skukuza, and **(o)** Solar_Village. The error bar means the 10 times of the standard error, which are used for the weighted trend analysis.

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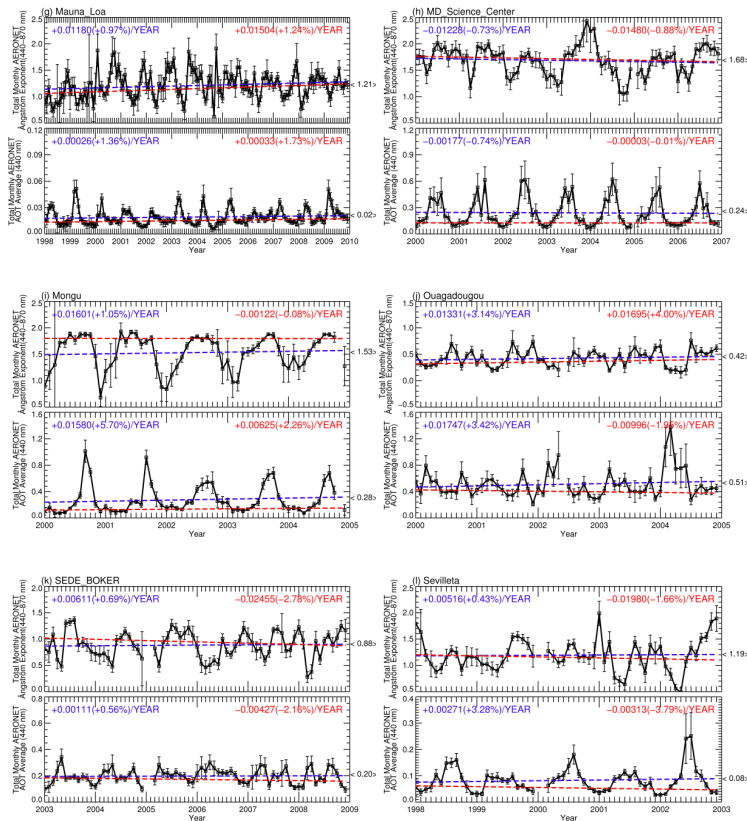


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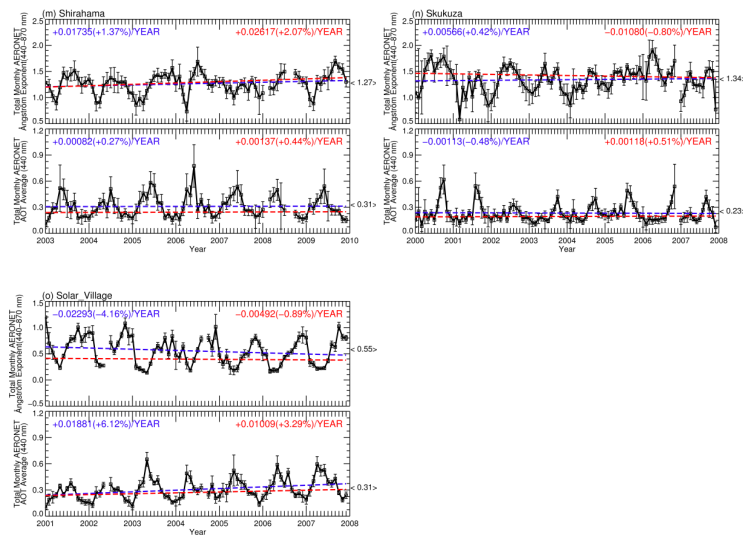


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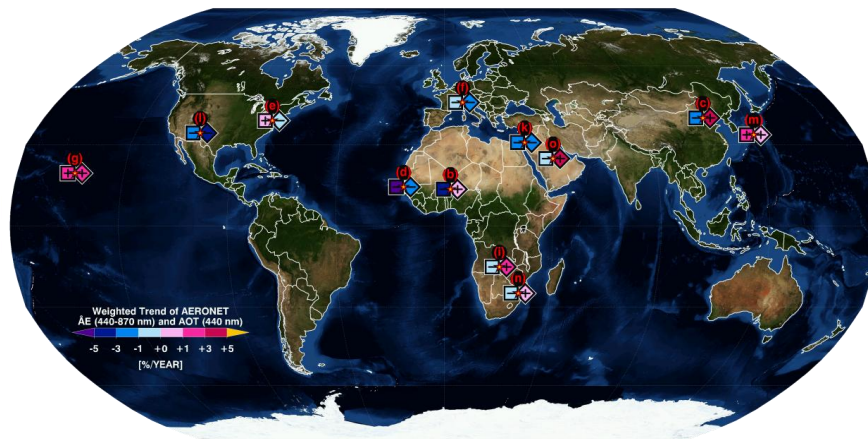


Fig. 8. The weighted trends of Ångström Exponent (440–870 nm) (left square) and AOT at 440 nm (right diamond) in percent at the major stations were indicated except **(a)** Avignon (+0.95%/year and +0.98%/year) over Western Europe, **(h)** MD_Science_Center (−0.88%/year and −0.01%/year) over North America, and **(j)** Ouagadougou (+4.00%/year and −1.95%/year, respectively) over West Africa.

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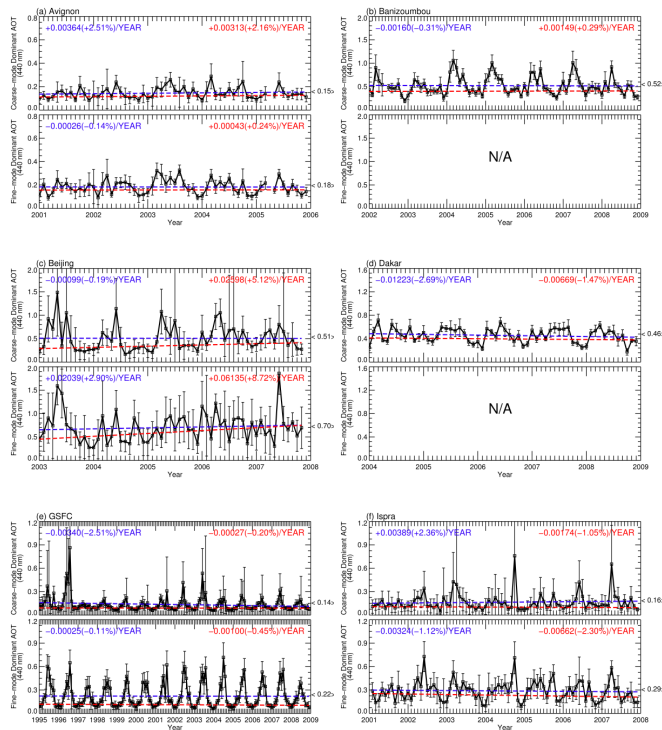


Fig. 9. Total averages (black one enclosed with parentheses at right vertical axis), temporal unweighted (blue one on the left upper part), and weighted (red one on the right upper part) trends of coarse- and fine-mode dominant AOT (440 nm) at the AERONET stations: **(a)** Avignon, **(b)** Banizoumbou, **(c)** Beijing, **(d)** Dakar, **(e)** GSFC, **(f)** Ispra, **(h)** MD_Science_Center, **(i)** Mongu, **(j)** Ouagadougou, **(k)** SEDE_BOKER, **(m)** Shirahama, **(n)** Skukuza, and **(o)** Solar_Village. The error bar means the 10 times of the standard error, which are used for the weighted trend analysis. Trend analysis of fine-mode dominant AOT is non-applicable (N/A) at **(b)** Banizoumbou, **(d)** Dakar, **(j)** Ouagadougou, and **(o)** Solar_Village.

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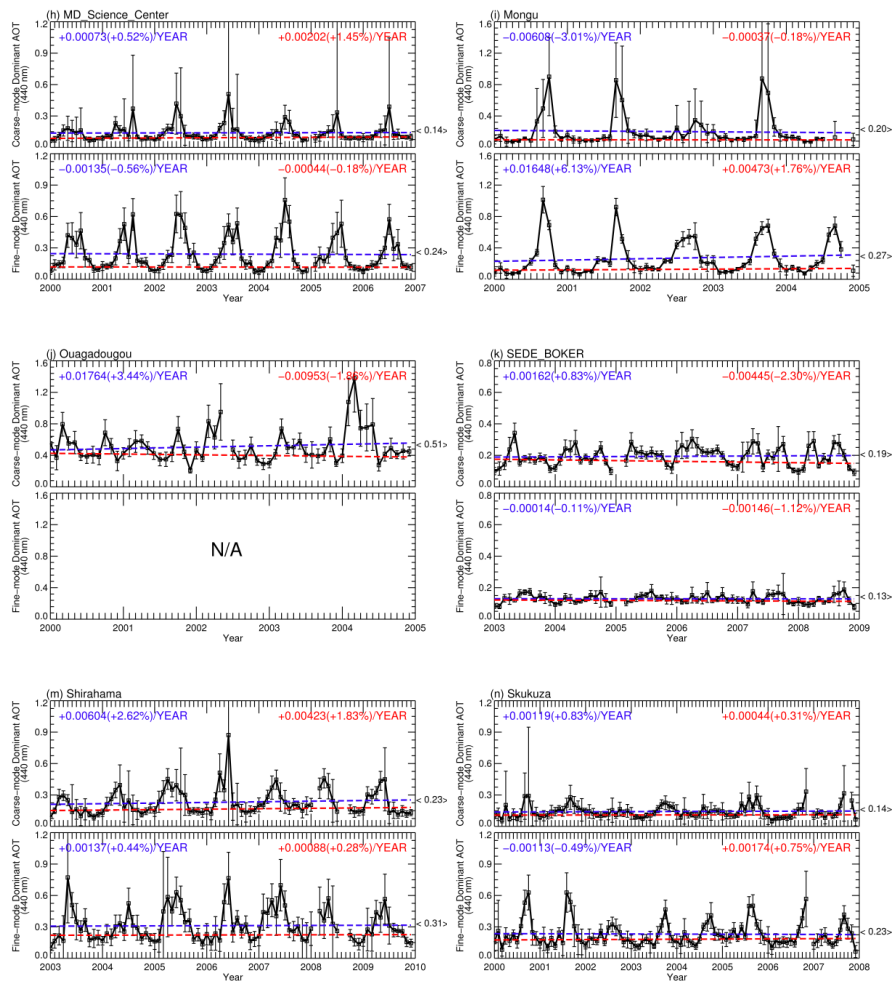


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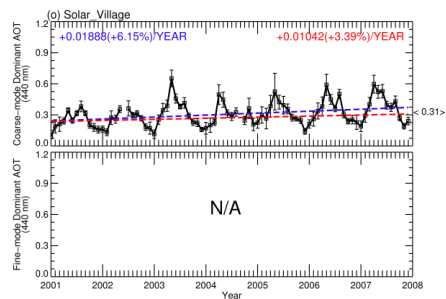


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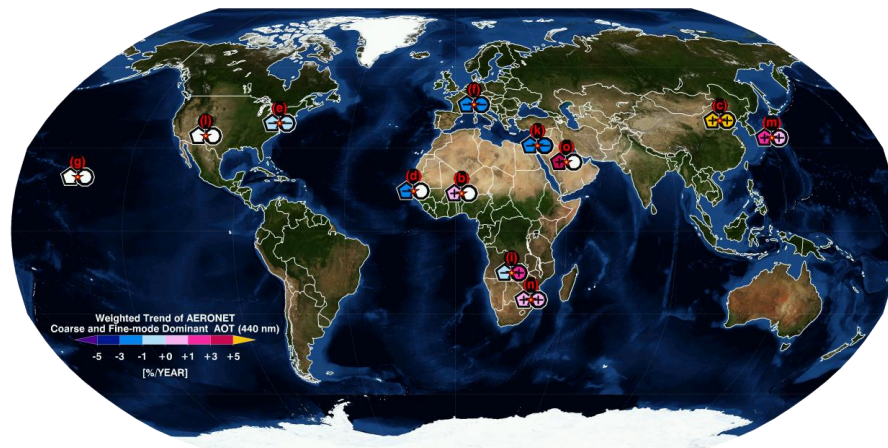


Fig. 10. The weighted trends of coarse- (left pentagon) and fine-mode (right circle) dominant AOT (440 nm) in percent at the major stations were indicated except **(a)** Avignon (+2.16%/year and +0.24%/year) over Western Europe, **(h)** MD_Science_Center (+1.45%/year and -0.18%/year) over North America, and **(j)** Ouagadougou (-1.86%/year and non-applicable, respectively) over West Africa. Non-applicable cases are shown as a white blank.

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