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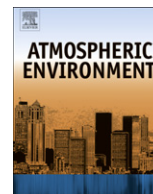
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Climatology of aerosol optical properties in Southern Africa

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ABSTRACT

A thorough regionally dependent understanding of optical properties of aerosols and their spatial and temporal distribution is required before we can accurately evaluate aerosol effects in the climate system. Long term measurements of aerosol optical depth, Angstrom exponent and retrieved single scattering albedo and size distribution, were analyzed and compiled into an aerosol optical properties climatology for southern Africa. Monitoring of aerosol parameters have been made by the AERONET program since the middle of the last decade in southern Africa. This valuable information provided an opportunity for understanding how aerosols of different types influence the regional radiation budget. Two long term sites, Mongu in Zambia and Skukuza in South Africa formed the core sources of data in this study. Results show that seasonal variation of aerosol optical thicknesses at 500 nm in southern Africa are characterized by low seasonal multi-month mean values (0.11 to 0.17) from December to May, medium values (0.20 to 0.27) between June and August, and high to very high values (0.30 to 0.46) during September to November. The spatial distribution of aerosol loadings shows that the north has high magnitudes than the south in the biomass burning season and the opposite in none biomass burning season. From the present aerosol data, no long term discernable trends are observable in aerosol concentrations in this region. This study also reveals that biomass burning aerosols contribute the bulk of the aerosol loading in August–October. Therefore if biomass burning could be controlled, southern Africa will experience a significant reduction in total atmospheric aerosol loading. In addition to that, aerosol volume size distribution is characterized by low concentrations in the non biomass burning period and well balanced particle size contributions of both coarse and fine modes. In contrast high concentrations are characteristic of biomass burning period, combined with significant dominance of fine mode particles.

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1. Introduction

Atmospheric aerosols of both natural and anthropogenic origin contribute substantially to global climate variability (Kondratyev, 1999; IPCC, 2007). Changes in the aerosol content of the atmosphere constitute a major forcing mechanism by affecting the radiative balance of the climate system (Crutzen and Andreae, 1990; Charlson et al., 1992).

A thorough regionally dependent understanding of optical properties of aerosols (e.g. aerosol optical thickness, size distribution, chemical composition) and their spatial and temporal distribution is required before we can accurately evaluate aerosol effects in the climate system (Hsu et al., 2000). However, the prospect of

fully understanding aerosols influence on climate forcing is small without validation and augmentation by ancillary ground based observations that can be provided by radiometers historically known as sun photometers (Holben et al., 1998).

The Southern Africa Science Initiative (SAFARI 2000) formed the foundation for a common strategy for aerosol and trace gas measurements at a number of regionally representative sites in southern Africa (Swap et al., 2003; Annegarn et al., 2002).

Since SAFARI 2000 aerosol measurements have continued in a systematic and continuous manner at Mongu in Zambia and Skukuza in South Africa. These sites are part of AERONET (Holben et al., 1998).

Both sites were established well before the year 2000, with Mongu starting in June 1995 and Skukuza in July 1998. These long term measurements made with CIMEL sun photometers provide a good opportunity to compile a climatology of aerosol optical properties, which in turn will improve our understanding on direct and indirect radiative forcing by aerosols in southern Africa.

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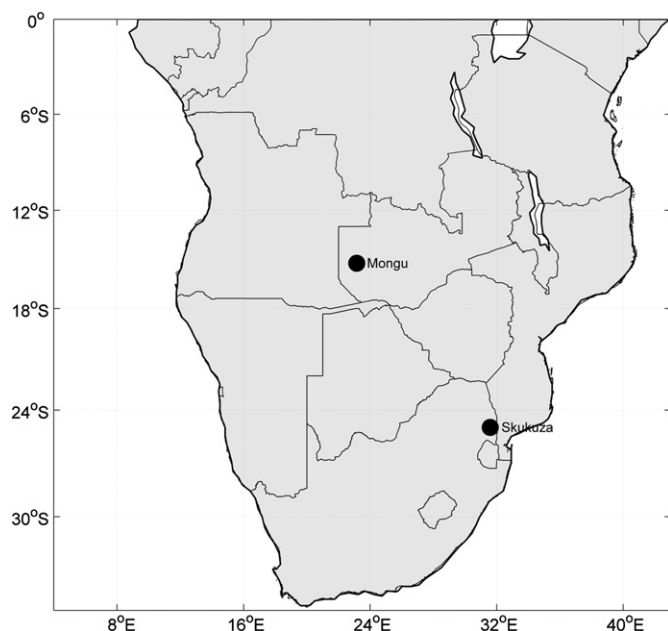


Fig. 1. Geographical position of Mongu at Zambia and Skukuza in South Africa (black dots).

2. Data and methodology

2.1. Site descriptions

Long-term measurements of aerosol data were acquired at two sites in southern Africa, Mongu in Zambia ($15^{\circ} 15' S$; $23^{\circ} 09' E$; elev. 1107 m) and Skukuza in South Africa ($24^{\circ} 59' S$; $31^{\circ} 35' E$; elev. 150 m) as seen in Fig. 1. Mongu is located in the centre of biomass burning activity in southern Africa and far from industrial sources. Skukuza is located in Kruger National Park and due to atmospheric circulation of the region this site can be affected by diverse sources of aerosols including Aeolian dust, industrial, maritime and biomass burning. The aerosol data cover the time period of June 1995 to June 2007, for Mongu and July 1998 to June 2008 for Skukuza.

2.2. Sun photometer measurements

The CIMEL Sun Photometer is a solar-powered, hardy, robotically pointed sun and sky spectral radiometer. A microprocessor computes the position of the Sun based on time, latitude, and longitude, which directs the sensor head to within approximately 1° of the Sun, after which a four-quadrant detector tracks the Sun precisely to a programmed measurement sequence. After the routine measurement is completed, the instrument returns to the “park” position awaiting the next measurement sequence. A “wet sensor” exposed to precipitation will cancel any measurement sequence during rain events. Data in the memory of the Sun photometer are transmitted via a radio transmitter to the METEOSAT geostationary satellite and then retransmitted to a ground receiving station (Holben et al., 1998).

The CIMEL was programmed to make measurements of the direct sun and diffuse sky radiances at Mongu and Skukuza within the spectral range 340–1020 nm and 440–1020 nm respectively. The direct sun measurements are made every 15 min in eight spectral channels at 340, 380, 440, 500, 675, 870, 940 and 1020 nm (nominal wavelengths). Seven of the eight bands are used to acquire aerosol optical thickness data. The eighth band at 940 nm is

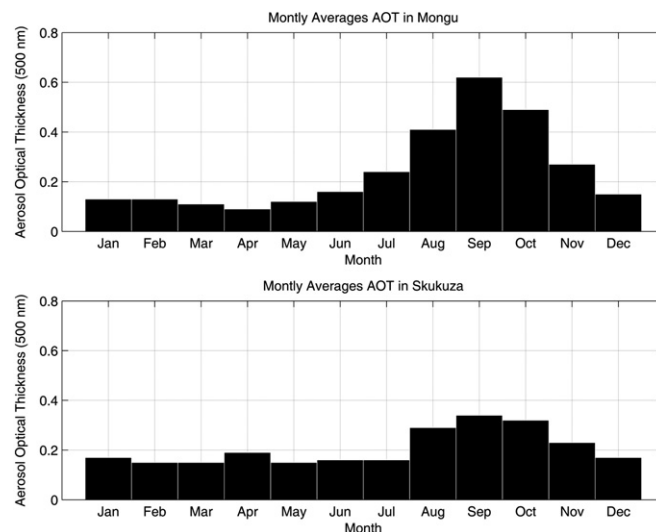


Fig. 2. Seasonal multi year monthly means of aerosol optical thickness computed from 1995 to 2007 at Mongu (upper panel) and from 1998 to 2008 at Skukuza (lower panel).

used to estimate total precipitable water content (Holben et al., 1998).

The diffuse sky radiances, called almucantar, is a series of measurements taken at the elevation angle of the Sun for specified azimuth angles relative to the position of the Sun. These measurements are taken at 440, 675, 870 and 1020 nm. During almucantar measurements, observations from a single channel are made in a sweep at a constant elevation angle across the solar disc and continue through 360° of azimuth in about 40 s. This is repeated for each channel to complete an almucantar sequence (Holben et al., 1998).

The calibration of the instruments were performed regularly at the Goddard Space Flight Center (GSFC) by a transfer of calibration from reference instruments that were calibrated by the Langley method at Mauna Loa Observatory. The combined effects of uncertainties in calibration, atmospheric pressure, and ozone amount (climatology is used) result in a total uncertainty in derived aerosol optical thickness of ~ 0.01 – 0.02 , with the largest errors in the UV (Eck et al., 1999).

Data from the two sites are transmitted via satellite communications to a receiving ground stations and are then downloaded to computers at the Goddard Space Flight Center in Maryland. The AERONET project algorithms compute the aerosol optical thickness retrievals in near real time. Data are quality checked and cloud-screened following the methodology of Smirnov et al. (2000).

A flexible inversion algorithm for the retrieval of optical properties, developed by Dubovik and King (2000) is used for retrieving aerosol volume size distributions, single scattering albedo, refractive index and phase function. Results can be accessed by internet linkage on the AERONET web page (<http://aeronet.gsfc.nasa.gov:8080>).

3. Results and discussion

3.1. Variability of aerosol optical thickness in southern Africa

3.1.1. Seasonal variation of multi year monthly mean of aerosol optical thickness in Mongu and Skukuza

The seasonal variation of monthly average aerosol optical thickness at 500 nm shows maximum values in the months of August, September and October for both sites (Fig. 2). This seasonal peak

Table 1
Seasonal averages of aerosol optical thickness at Mongu and Skukuza.

Site	Seasonal averages of AOT (500 nm)				Annual average
	DJF	MAM	JJA	SON	
Mongu	0.14	0.11	0.27	0.46	0.28
Skukuza	0.17	0.17	0.20	0.30	0.21

occurring in spring for southern Africa is due to the high activity of biomass burning in southern Africa during this period. The multi-year annual means of AOT (500 nm) are 0.28 and 0.21 for Mongu and Skukuza respectively. Monthly mean values in Mongu range from 0.11 to 0.62 and 0.15 to 0.34 in Skukuza. Highest AOT values at Mongu and Skukuza are observed during the biomass burning season (July to November). The onset of the biomass burning season starts one month earlier at Mongu than at Skukuza; however the highest levels are both recorded on the month of September.

A summary of seasonal averages of aerosol optical thickness are given in Table 1. Low aerosol loadings ranging from 0.11 to 0.17 are observed from December to May and values ranging from 0.20 to 0.27 are characteristic of the winter period in southern Africa during June, July and August (JJA). In the peak of biomass burning very high values ranging from 0.30 to 0.46 can be observed. These results point out the large differences in aerosol loadings between the non-biomass burning and biomass-burning seasons and explicitly show that biomass burning is a large source of aerosols in southern Africa.

3.1.2. Inter annual variability of aerosol optical thickness in Mongu and Skukuza

Inter annual variability of AOT (500 nm) show the spring seasonal peaks with maximum values observed in September for both sites (Fig. 3). Significant variations of total aerosol loading from year to year can be observed, these differences are more pronounced at Skukuza (Fig. 3, lower panel). High values were

Table 2
Aerosol loading classification in southern Africa as result of AOT range at 500 nm.

AOT range at 500 nm	Aerosol loading classes
<0.1	Very low
0.1–0.2	Low
0.2–0.3	Medium
0.3–0.5	High
>0.5	Very high

recorded in the biomass burning peak month of September in two consecutive years of 2000 and 2001 suggesting increased biomass burning activity followed by lower values in 2002 and 2003. These significant changes will be investigated in the next sections.

Given the magnitude of the climatological values of AOT and their seasonal variability it is reasonable to suggest a classification of the state of the atmosphere in southern Africa based on aerosol loading. AOT (500 nm) values have been classified as Very low; Low; Medium, High and very High (Table 2)

3.1.3. Monthly inter annual Anomalies of aerosol optical thickness in Mongu and Skukuza

The monthly inter annual anomalies (computed as departures from multi-year monthly means) shows clearly the significant inter annual variability of total aerosol content in atmosphere of southern Africa (Fig. 4). Years of relative abundance of aerosols can be identified by positive anomalies, while negative anomalies represent relative reduction of the aerosol amount. One crucial question arising from the aerosol community is whether atmospheric aerosols concentrations are increasing or not. From the present aerosol data, no long term discernable trends are observable in aerosol concentrations in southern Africa possibly suggesting no anthropogenic induced trends. The slope of the linear regression is nearly zero and very low correlation as observed by the R-squared values of 0.001 and 0.002 for Mongu and Skukuza.

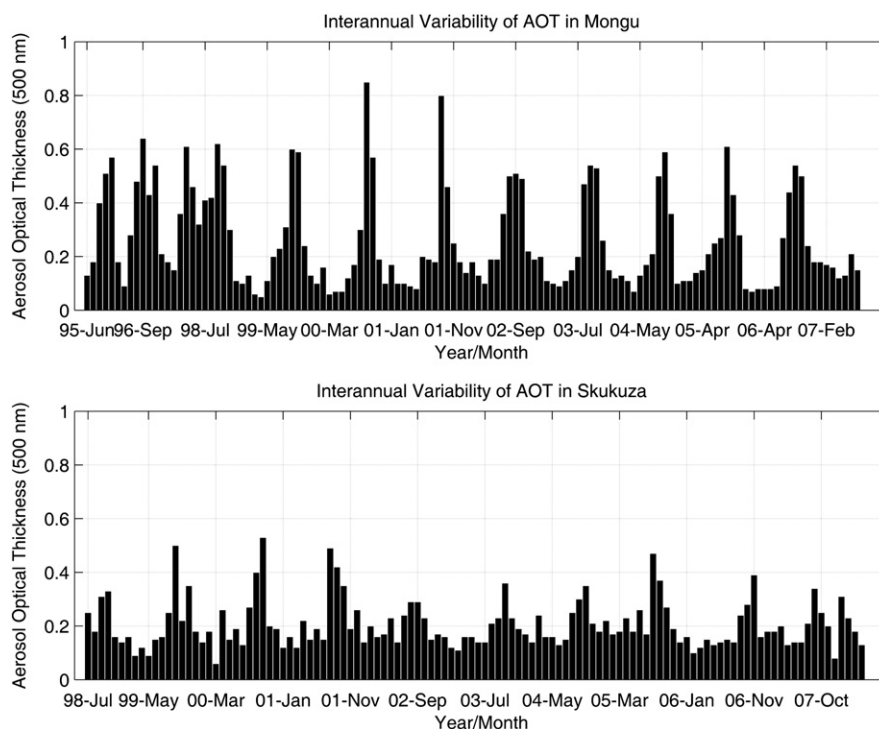


Fig. 3. Inter annual variability of aerosol optical thickness at Mongu (upper panel) and Skukuza (lower panel) showing changes of aerosol loading across different years.

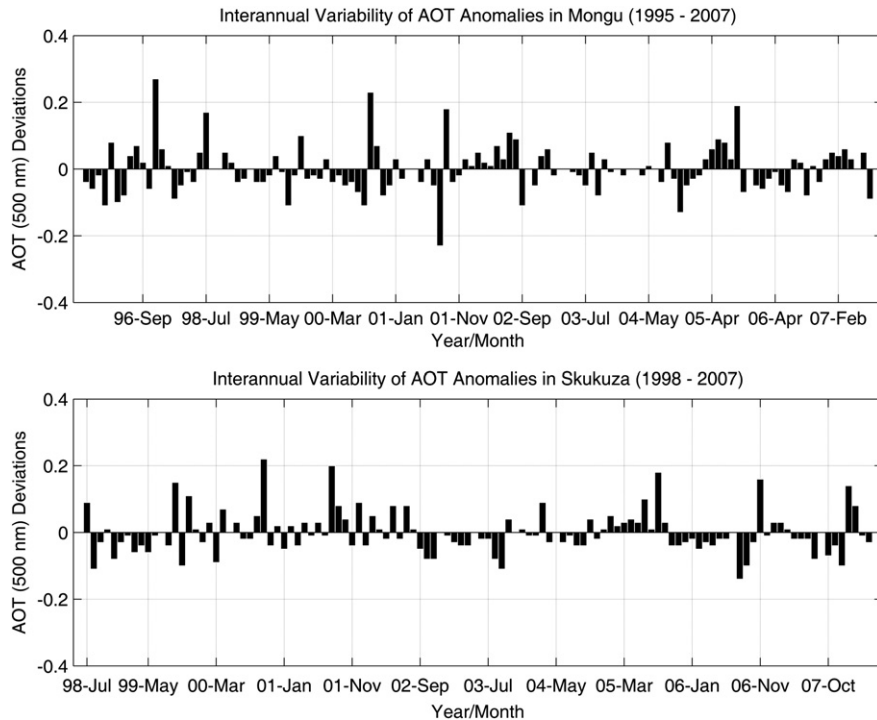


Fig. 4. Inter annual changes of aerosol loading at Mongu (upper panel) and Skukuza (lower panel) given as aerosol optical thickness anomalies across different years.



Fig. 5. Inter annual changes of aerosol loading during the peak of biomass burning season in southern Africa (August, September and October) at Mongu and Skukuza.

However inter annual changes of aerosol loadings are significant in both positive and negative directions. These changes need to be investigated in order to understand the global or regional patterns associated with this year to year variation.

An attempt has been made to find possible trends into biomass burning and non-biomass burning seasons separately. For the

biomass burning season (August, September and October) no discernable trend has been found at both Mongu and Skukuza. It seems that a slight reduction of the total aerosol loading can be observed over the last 5 years of observations through a negative slope between 2003 to 2007 at Skukuza (Fig. 5). During the non-biomass burning dry season (Fig. 6), Mongu has a positive slope

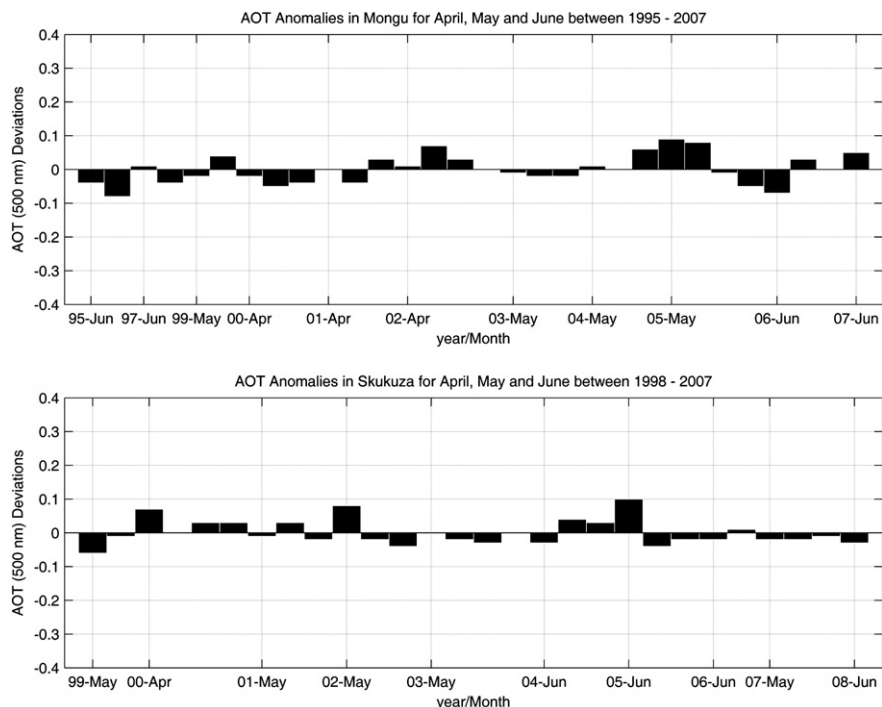


Fig. 6. Inter annual changes of aerosol loading during the non biomass burning season in southern Africa (April, May and June) at Mongu and Skukuza.

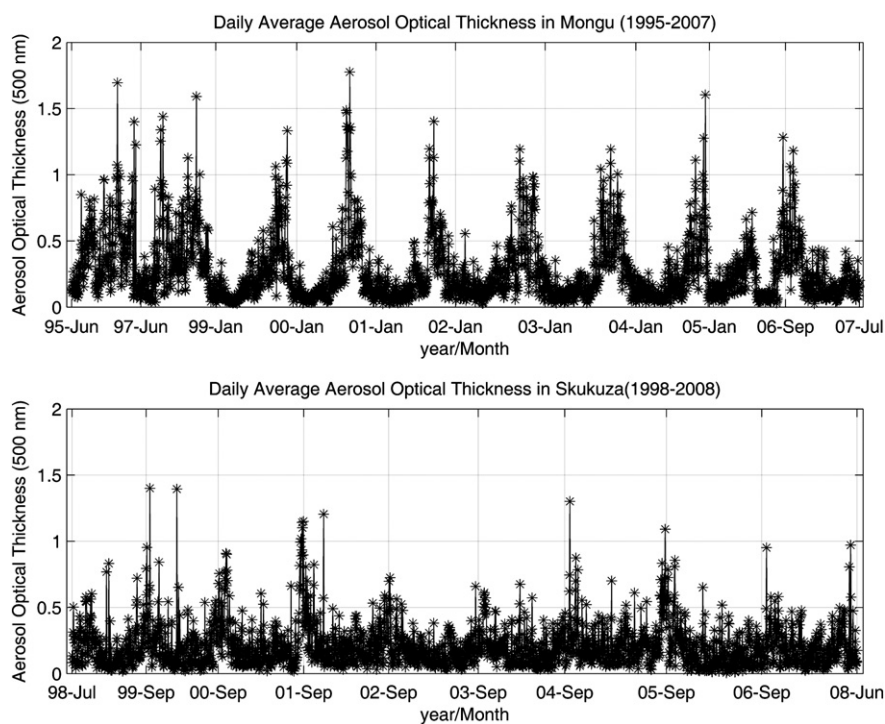


Fig. 7. Day to day variability of aerosol optical thickness from 1995 to 2007 at Mongu (upper panel) and from 1998 to 2008 at Skukuza (lower panel).

(0.02) and Skukuza has a slightly negative slope (-0.001) which are both not statistically significant.

3.1.4. Day to day variability of aerosol optical thickness in Mongu and Skukuza

Daily averages of AOT (500 nm) range from 0.02 to 1.7 at Mongu and 0.01 to 1.4 at Skukuza over the entire monitoring period. The

overall averages for the entire period of observations are 0.28 and 0.21 for Mongu and Skukuza respectively. A high degree of day to day optical variability is evident at both sites, with Mongu showing that episodes of very high turbidity ($AOT > 0.5$) are much more common compared to Skukuza (Fig. 7). This variability is largely attributed to the diverse contributions of aerosol and varying meteorological conditions impacting the tropospheric column

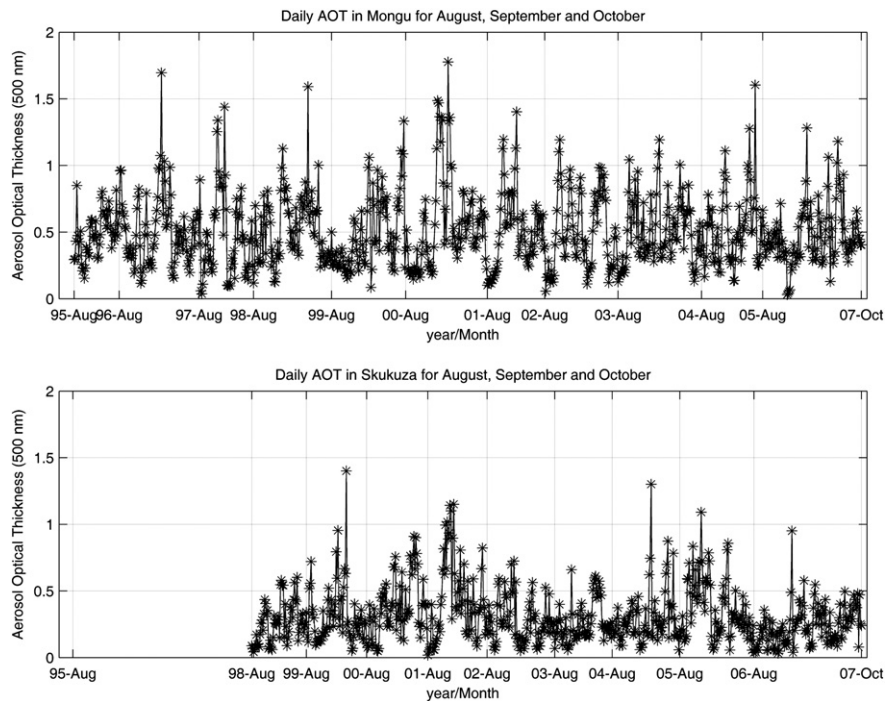


Fig. 8. Day to day variability of aerosol optical thickness for Mongu and Skukuza during the biomass burning season in southern Africa.

above Mongu or Skukuza and is likely to include sources such as local and transported wind blown dust, emissions from domestic biofuel usage, local and transported biomass burning and long-range transport of industrial emissions from the South African Highveld.

Based on the wide range of aerosol loadings and their variability it would be important to understand the frequency

distribution of the different classes of AOT ranges suggested previously.

As observed before that aerosol loadings in southern Africa are characterized by low values from December to May, medium values in June to August and high to very high during September and November. For better understanding of the day to day variability of AOT values in these distinct periods, times series were separated in

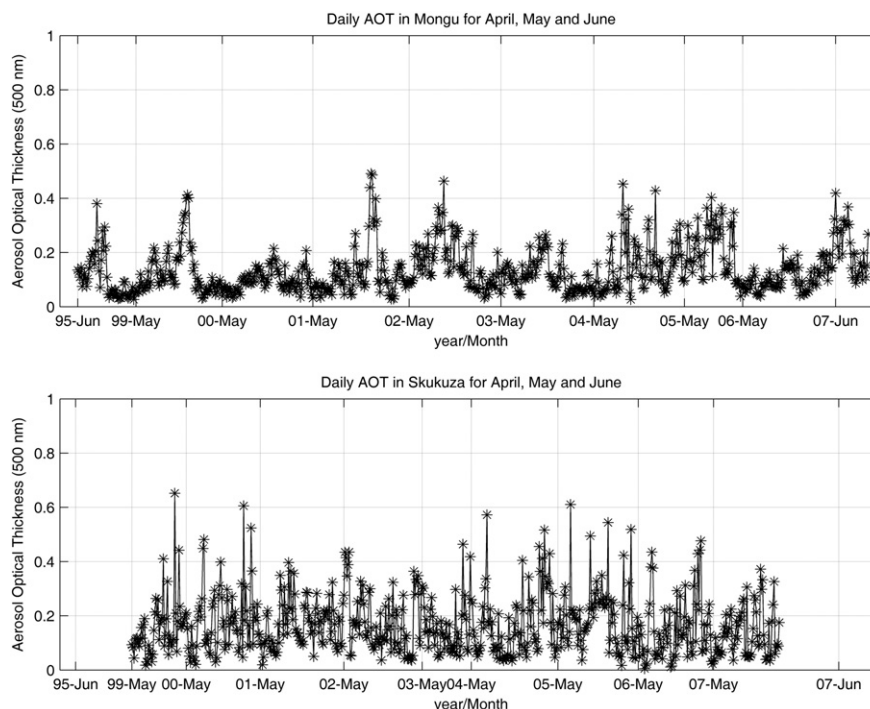


Fig. 9. Day to day variability of aerosol optical thickness for Mongu and Skukuza during the none biomass burning season in southern Africa.

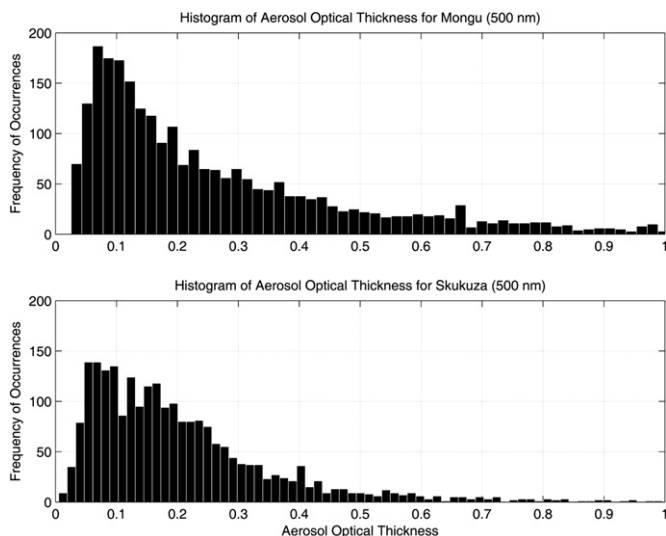


Fig. 10. Frequency distribution of aerosol optical thickness for Mongu and Skukuza.

two main seasons. The first one cover the peak of biomass burning (August, September and October) and the second covering the non-biomass burning season and outside the rain season (April, May and June).

During the biomass burning season, daily values of AOT (500 nm) range from 0.03 to 1.7 in Mongu and 0.02 to 1.4 at Skukuza (Fig. 8), the overall averages are 0.50 and 0.31 respectively. In contrast during non-biomass burning dry season period the AOT (500 nm) varied between 0.02–0.49 in Mongu and 0.01–0.65 at Skukuza (Fig. 9). The overall averages for this period are 0.14 and 0.17 respectively. The AOT ranges presented above reveal that within the biomass burning period there are low episodes of aerosol loading occurring and also during non-biomass burning period high aerosol loading episodes are observed. It is also noted that daily values of AOT at 500 nm greater than 1 only occur in biomass burning period at both sites. Once again this demonstrates the strength of biomass burning source.

The frequency of occurrences histograms of AOT (500 nm) for the entire data recorded at Mongu (1995–2007) and Skukuza (1998–2008) shows typical AOT skewed distributions (O'Neill et al., 2001b). The median values at Mongu and Skukuza are 0.19 and 0.17 respectively (Fig. 10). The difference between the mean and median at Mongu (0.28 and 0.19) is significant compared to Skukuza (0.21 and 0.17), indicating that AOT values at Skukuza are more normally distributed compared to Mongu.

Table 3 summarises the probabilities of certain classes of aerosols loadings that can occur above Mongu and Skukuza throughout

Table 3

Probability of occurrences of aerosol optical thicknesses values within different classes ranges for all data points, biomass burning (ASO) and none biomass burning (AMJ) periods at Mongu and Skukuza.

Aerosol loadings classes	AOT range	Probability within the range (%)					
		Mongu			Skukuza		
		All data points	ASO	AMJ	All data points	ASO	AMJ
Very low	<0.1	23.7	1.2	44.7	27.8	12.0	33.7
Low	0.1–0.2	26.7	7.6	37.2	31.9	23.8	34.5
Medium	0.2–0.3	15.7	14.4	12.0	20.7	23.5	20.4
High	0.3–0.5	17.4	33.1	6.1	13.8	24.9	10.1
Very high	>0.5	16.4	43.7	0	5.8	15.8	1.3

the year (all data points) and during the biomass burning (ASO) and non-biomass burning (AMJ) periods. All five selected ranges do occur at both sites in an almost proportional manner, except for very high loading (AOT > 0.5) where the turbidity levels are significant higher for Mongu. Under the extremes of optical classes defined in previous sections (Table 2), the probability that AOT (500 nm) values fall below 0.1 is 23.7% for Mongu and 27.8% for Skukuza. Whereas the probability that AOT values are greater than 0.5 is 16.4% for Mongu and 5.8% for Skukuza.

Under biomass burning and none biomass burning periods significant changes of column aerosol loading can be observed from one season to another; hence the radiation balance will be strongly affected. In Mongu a huge optical contrast can be observed, with the biomass burning period accounting for >40% probability in very high aerosol loading and nearly zero during the non-biomass burning for the same class. On the other hand, within the very low class during non-biomass burning; Mongu accounts for 45% probability and Skukuza 34%.

The atmosphere above Mongu is cleaner than Skukuza during the non-biomass burning period and the atmospheric aerosol loading at the two sites are reversed during biomass burning season, with highest AOT values recorded at Mongu. This is consistent with a finding by Kirkman et al. (2000), from airborne measurements, that an aerosol gradient exists over southern Africa and it reverses from biomass burning to none biomass burning seasons. The aerosol loading south of 20°S is consistent throughout the year and is probably dominated by anthropogenic fossil fuel combustion sources. This has significant implications for radiative forcing potential over the subcontinent at a seasonal level.

3.2. Particle size

3.2.1. Analysis of Angström exponent over Mongu and Skukuza

The Angstrom exponent (α) parameter provides some basic information about the aerosol size distribution. The α values presented in this study were computed from 440–870 nm wavelengths. In this case when $\alpha < 0.75$, coarse particles dominate, while for $\alpha > 0.75$, fine mode particles exert the greatest influence (Eck et al., 1999; O'Neill et al., 2001a; Eck et al., 2005).

The frequency distribution histograms of Angstrom exponent indicate a wide range of particle sizes with median Angstrom exponent values of 1.73 and 1.41 in Mongu and Skukuza respectively (Fig. 11). These results suggest that the tropospheric aerosol loading over both sites has a diverse number of contributing

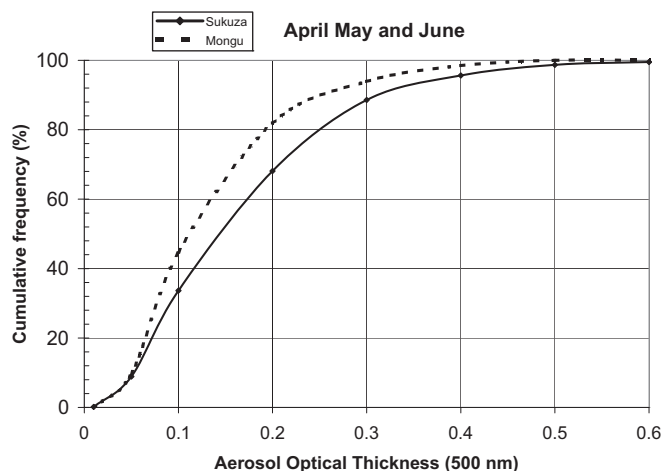


Fig. 11. Angstrom exponent distribution showing the frequency of occurrences for a given size mode of particles.

Table 4

Angstrom exponent ranges and their probabilities within a given range.

Angstrom exponent	Probability within the range (%)	
	Mongu	Skukuza
<0.5	2.6	4.6
0.5–1.0	9.2	16.1
1.0–1.5	18.6	39.6
1.5–2.0	62.4	37.5
>2.0	7.2	2.2

sources. However the dominant range well above 1, indicate that the aerosol particle size is for the most part of time dominated by fine particles, to a larger extent in Mongu than in Skukuza.

Table 4 gives more detailed information about the α values ranges and their probability to be within a given range. Angstrom exponent values very close to 2 (1.5–2.0) accounts for ~60% in Mongu, indicating that very fine particles are dominant in this site, however coarse particles likely from airborne dust or aging process could also be found in small concentrations. In contrast Angstrom exponent values close to 1 (0.5–1.5) represent ~50% in Skukuza, indicating well balanced mixture of coarse and fine particles in this site.

The predominance of each component (coarse or fine) will be investigated in more details in the next section.

3.2.2. Analysis of fine and coarse mode optical thickness

The recognition that the aerosol particle size distribution is effectively bimodal enables one to exploit the spectral curvature information of $\ln \tau_a$ versus $\ln \lambda$ to extract the elemental optical parameters of each mode (O'Neill et al., 2003). The total aerosol optical thickness (τ_a) can be represented as the sum of fine and coarse mode optical thickness ($\tau_a = \tau_f + \tau_c$).

The separation into two components τ_f and τ_c ; yields aerosol optical statistics, which allow the understanding of atmospheric

optical events dominated by fine or coarse mode particle size distribution. Recently AERONET started providing aerosol optical data in these two separate components generated from the latest version of the spectral deconvolution algorithm (developed by O'Neill et al., 2006).

3.2.2.1. Seasonal variation of the fine and coarse mode aerosol optical thickness in Mongu and Skukuza. Atmospheric aerosol loads are significantly dominated by fine mode particle size throughout the year; with monthly averages of AOT (500 nm) for fine mode ranging between 0.07–0.57 and 0.11–0.28 in Mongu and Skukuza respectively (Fig. 12) upper panels. Remarkable increases of fine particles are observed from July to November at Mongu and from August to November at Skukuza. In contrast the coarse mode remain with low AOT values (0.02–0.06) along the year, with the lowest values registered during early dry season and slightly increases towards the wet season at both sites (Fig. 12) lower panels.

In order to assess the contribution of each component to the total aerosol optical thickness, the percentages of the fine and coarse modes are presented in Fig. 13 for Mongu and Skukuza. White and black bars represent the fine and coarse mode percentages respectively. The general picture confirms that fine particles dominate the atmospheric loadings at these sites, with an average percentage of approximately 78%.

At Mongu the presence of fine mode particles is significant from August to October, with percentages between 90% and 94%. During the wet period from December to March the fine particles percentage decrease up to ~ 60% and consequently the coarse mode increases. These results are good indicators of the strong contribution of biomass burning at the end of the dry season to the fine particles in this region.

At Skukuza the contribution of biomass burning to the fine mode can be observed particularly from August to October, with monthly average percentages of ~ 83%. However the strength of

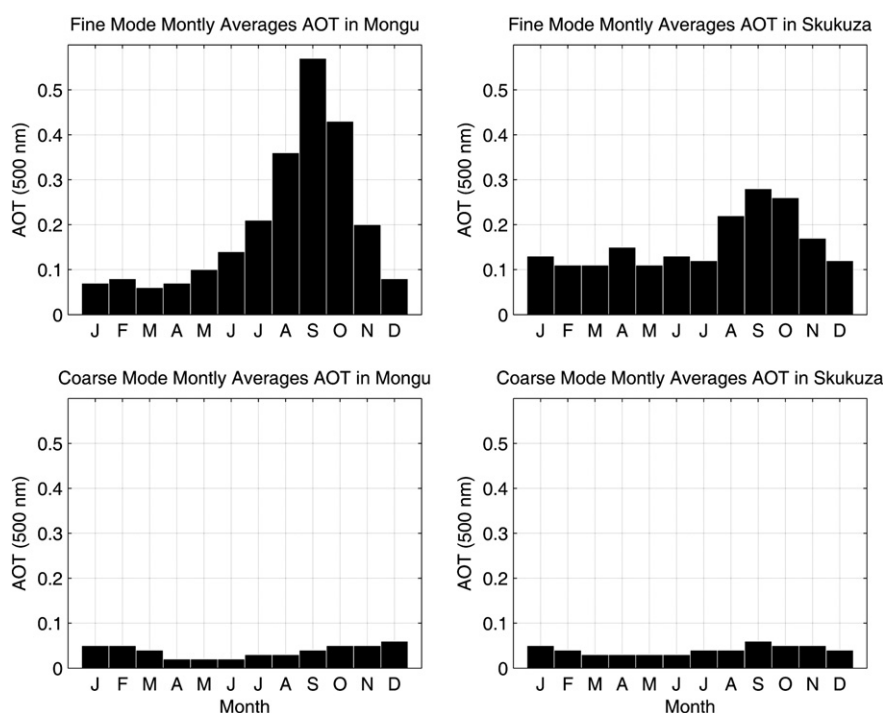


Fig. 12. Seasonal variation of the fine and coarse mode components of aerosol optical thickness at 500 nm at Mongu and Skukuza. Fine mode component are showed in the upper panels while the coarse mode component in the lower panels.

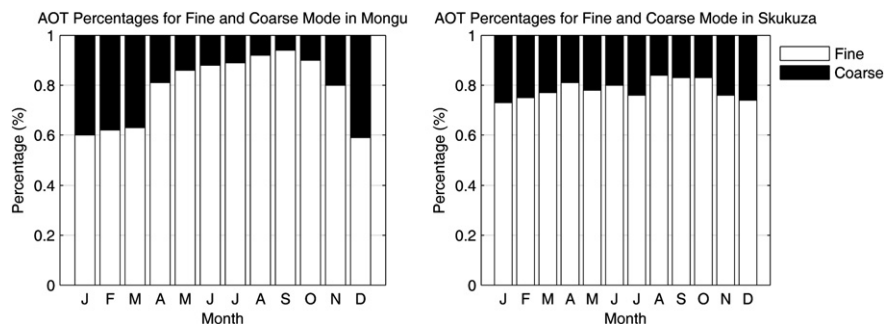


Fig. 13. Monthly percentages distribution of the fine and coarse mode aerosol particles at Mongu and Skukuza. White bars represent the fine mode and the black bars the coarse mode.

Table 5

Seasonal averages of AOT at 500 nm and percentages distribution of the fine and coarse mode at Mongu and Skukuza.

Period	Mongu				Skukuza			
	Seasonal averages for fine and coarse modes							
	Fine	Coarse	(%) of Fine	(%) of Coarse	Fine	Coarse	(%) of Fine	(%) of Coarse
DJF	0.08	0.05	60	40	0.12	0.04	74	26
MAM	0.08	0.02	77	23	0.13	0.03	79	21
JJA	0.24	0.03	90	10	0.16	0.04	80	20
SON	0.40	0.05	88	12	0.24	0.06	80	20

the fine particles sources it's also significant outside the peak of biomass burning, where the lowest percentage for fine mode is 73%, well above the lower value for Mongu (60%). These results suggest that fine mode particles sources impacting over the atmosphere of Skukuza are not limited to biomass burning and are most likely to be fossil fuel combustion sources over the South African Highveld.

Previous studies on air mass transport climatology from the industrial South African Highveld revealed that the transport to the east off the Indian ocean account for 70% (Freiman and Piketh, 2003). This air path way passes over Skukuza and is likely to carry particles from industrial sources.

The summary of seasonal averages of the fine and coarse mode aerosol optical thickness and their percentages distributions are given in Table 5. By applying the same classification of the aerosol column concentrations used in previous sections, it can be seen that aerosol loadings of the coarse component are always limited within the Very low class (AOT at 500 nm < 0.1) compared to the wide range of optical classes covered by the fine component (very low, low, medium and high).

From December to May aerosol loadings for the fine component in Mongu are less than 0.1, in this same period in Skukuza values between 0.1–0.2 are observed. In the winter period (JJA) and early biomass burning season both sites show an increase in the presence of the fine component until the highest values are registered during SON. The changes of aerosol loadings are more pronounced in

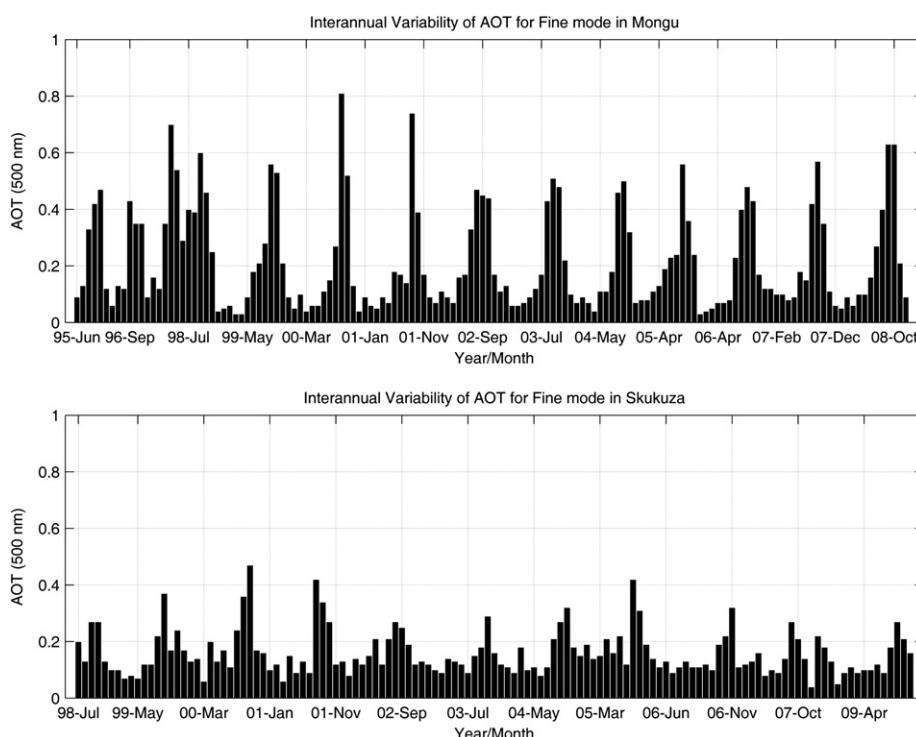


Fig. 14. Interannual variability of the fine mode aerosol optical thickness at Mongu (upper panel) and Skukuza (lower panel).

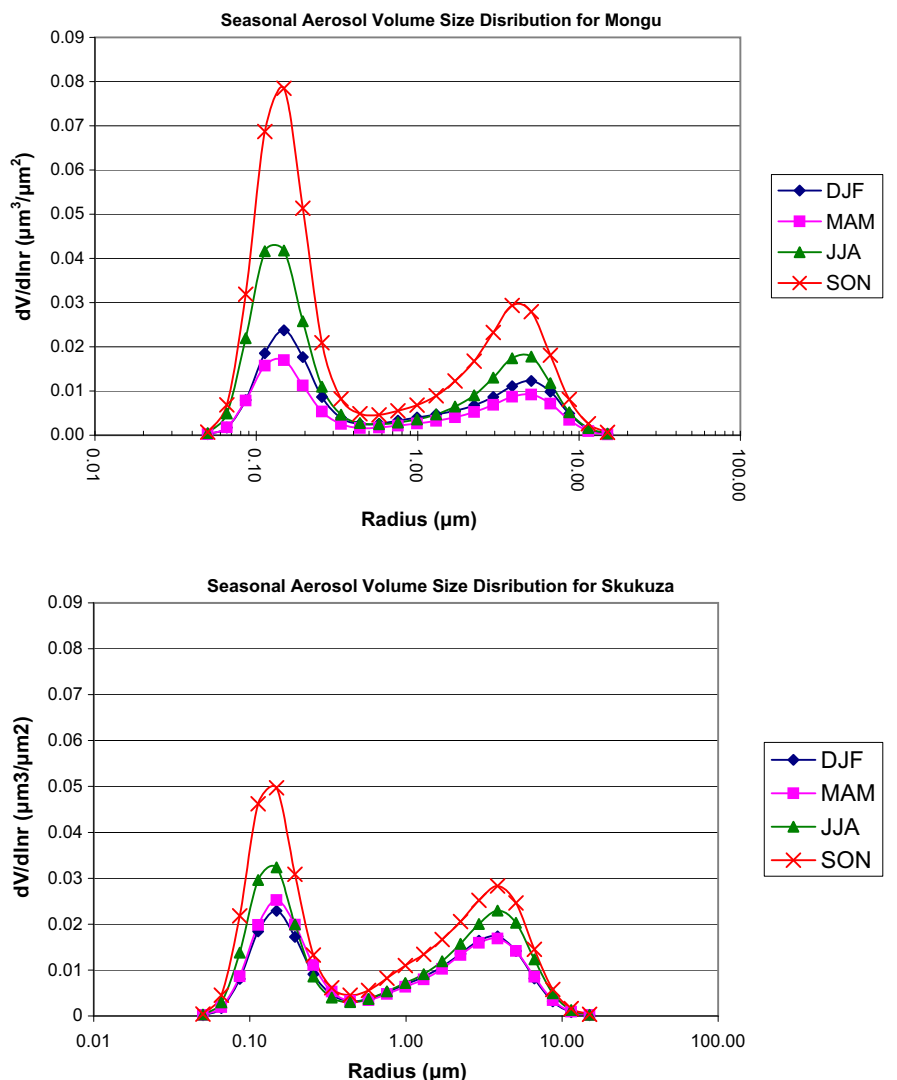


Fig. 15. Volume size distributions retrievals for different seasons at Mongu (upper panel) and Skukuza (lower panel).

Mongu than in Skukuza, where the values fluctuate between 0.1 and 0.3 (low and medium classes).

3.2.2.2. Interannual variability and trends of the fine mode aerosols optical thickness in southern Africa. Previous sections have shown that the coarse component of the aerosol loading does not change significantly and does not contribute to the high aerosol loadings in the region. From this perspective further analysis will be concentrated on the fine component.

Fig. 14 shows the interannual variability of the fine mode aerosol optical thickness in Mongu and Skukuza. The strength of biomass burning as source of fine particles in the atmosphere can be better evaluated by observing the high magnitudes of AOT in Mongu compared to Skukuza. Combining the information given by Fig. 14 and from the previous Table 5; it is reasonable to say that from December to May, southern Africa experiences a relatively clean atmosphere, followed by a transition period from June to August of a moderately turbid atmosphere and finally from September to October with high levels of biomass burning aerosol loadings.

Assuming that most of the fine particles sources including forest and savanna fires are for the most related with human activities. As such changes in behaviours and practices will significantly reduce

the amount of tropospheric aerosols in this region and hence lead to better air quality. However, the socio-economical implications of this are far reaching and need independent investigation.

3.2.3. Analysis of the volume size distribution

Seasonal volume size distribution retrievals for the entire data set reveals differences in aerosol mode concentrations during the year (Fig. 15). During the low aerosol loading period from December to May (with average AOT between 0.11 to 0.17), the concentrations of aerosol particles are quite low (less than $0.02 \mu\text{m}^3 \mu\text{m}^{-2}$) and well balanced on both coarse and fine modes concentrations in both sites Mongu and Skukuza. The winter period and the early stage of biomass burning activities are characterized by the increasing of fine particles concentrations, this increase continues and reach remarkable values during the peak of biomass burning season. Changes in concentrations of fine mode particles from non-biomass burning to the peak of biomass burning periods are clearly observed at both sites; with Mongu contributing more (factor of four), compared to Skukuza by factor of two. From this perspective its clear that biomass burning in southern Africa is a significant source of fine particles, which interact efficiently with the solar radiation and hence can affect the radiation budget of the region.

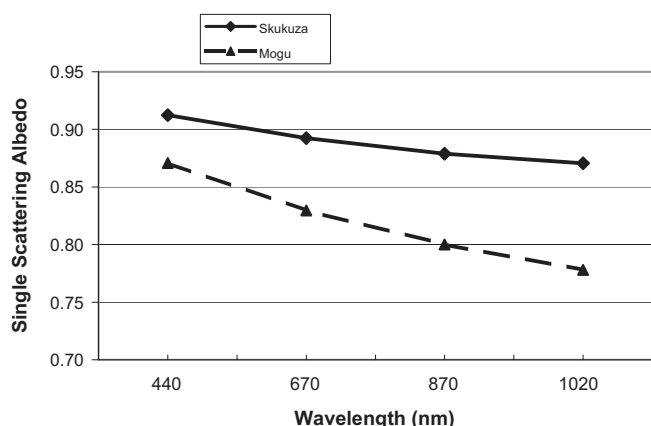


Fig. 16. Spectral single scattering albedo retrieved from AERONET almucantar scans at Mongu and Skukuza.

3.3. Column-integrated aerosol single scattering albedo

The single scattering albedo (SSA) is the probability that, given an interaction between the photon and particle, the particle will be scattered rather than absorbed. Single scattering albedo is equal to zero ($SSA = 0$) for a perfectly absorbing aerosol and equal to one ($SSA = 1$) for pure scatter. Previous analysis of SSA retrievals accuracy from AERONET data have shown that combined systematic uncertainty is ~ 0.01 (Leahy et al., 2007).

Fig. 16, for cases with AOT at 440 nm > 0.4 , shows that SSA decreases with wavelength at both sites and differences in magnitudes can also be observed in all wavelengths. Decrease of SSA with wavelength is characteristic of the absorption by small elemental carbon particles in a mixture of non-absorbing particles (Dubovik et al., 2002; Eck et al., 2001).

Special attention will go to the magnitude of the SSA in the visible range (440–670 nm), to better compare with previous studies results. In this study SSA values vary between 0.91 and 0.89 at Skukuza at 440 and 670 nm respectively, while at Mongu the values are 0.87 and 0.83 at the same wavelengths. Ichoku et al. (2003) suggested using 0.88 and 0.84 within the same wavelength range (440 and 670 nm) instead of constant value of 0.90, suggested by previous studies for southern Africa. Previous radiative forcing studies for southern Africa have debated the values used for SSA. Values used by Ichoku et al. (2003) and Leahy et al. (2007) ($SSA = 0.85 \pm 0.02$) are the most representative for the northern region most affected by biomass burning emissions. It should be noted, however, that these SSA values are not appropriate south of 20° S, where the value for Skukuza (showing less absorbing aerosols) would be more appropriate, i.e. $SSA = 0.90 \pm 0.03$.

From this study it seems reasonable to suggest the use of constant value of 0.90 for the southernmost area (low biomass burning activity) and the lower values of SSA for the northernmost area with high biomass burning activity.

4. Conclusions

A successful ground based long-term monitoring and characterization of aerosols, accessible database for the scientific community combined with good international collaboration has been a crucial factor in the study of atmospheric aerosol properties worldwide. These valuable and unique long term measurements of atmospheric aerosols in southern Africa provided a good opportunity to compile a climatology of aerosol optical properties, which

in turn will improve our understanding on direct and indirect radiative forcing by aerosols in this region.

Seasonal variation of aerosol optical thicknesses at 500 nm in southern Africa are characterized by low values (0.11–0.17) from December to May, medium values (0.20–0.27) between June and August, and high to very high values (0.30–0.46) during September to November. In terms of spatial distribution the north has high magnitudes than the south (north–south gradient) in the biomass burning season and the opposite in none biomass burning season, i.e. the gradient reverse. This has significant implications for radiative forcing potential over the subcontinent at a seasonal level.

Aerosol loadings in southern Africa display significant inter annual variability, showing years with relative abundance or reduction of total aerosol amounts. However, from the present aerosol data, no long term discernable trends are observable in aerosol concentrations in this region. The impact of large scale extreme events on annual aerosols amounts is strongly suggested and should be investigated in more detail.

High Angstrom exponent values (1.5–2.0) account for $\sim 60\%$ of the observations in Mongu, indicating that very fine particles are the most dominant, however coarse particles likely from airborne dust or aging process could also be found in small concentrations. In contrast Angstrom exponent values close to 1 (0.5–1.5) represent $\sim 50\%$ at Skukuza, indicating mixtures of coarse and fine particles in this site and suggesting that, Skukuza has a more diverse number of contributing aerosol sources.

Partition of aerosol optical thickness in fine and coarse components in southern Africa also reveal significant dominance of fine mode particles throughout the year in both sub regions; with remarkable increases of fine particles observed from July to November in the northernmost region and from August to November in the southernmost part.

It is also noted that high magnitudes of aerosol optical thickness are associated with fine particles mainly attributed to biomass burning in this region. In contrast clean episodes are related to coarse mode particles which remain within very low class throughout the year.

The volume size distribution is characterized by low concentrations (less than $0.02 \mu\text{m}^3 \mu\text{m}^{-2}$) during the low aerosol loading period in southern Africa from December to May (with average AOT between 0.11 to 0.17), and well balanced on both coarse and fine modes concentrations. The winter period and the early stage of biomass burning activities (June, July and August) are characterized by the increasing of fine particles concentrations, this increase continues and reach remarkable values during the peak of biomass burning season. These results add credence that biomass burning in southern Africa is a significant source of fine particles, which interact efficiently with the solar radiation and hence can affect the radiation budget of the region.

In this study, the single scattering albedo in the visible range (440 and 670 nm) vary between 0.91 and 0.89 in Skukuza respectively, while in Mongu the values are 0.87 and 0.83 at the same wavelengths. Ichoku et al. (2003) suggested using 0.88 and 0.84 within the same wavelength range (440 and 670 nm) instead of constant value of 0.90, suggested by previous studies for southern Africa. From this study it seems reasonable to suggest the use of constant value of 0.90 for the southernmost area (low biomass burning activity) and the lower values of SSA for the northernmost area with high biomass burning activity.

In general southern Africa experience a relatively clean atmosphere from December to May, followed by a transition period from June to August of a moderately turbid atmosphere and finally from September to October with high levels of biomass burning aerosol loadings.

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