Altitude-dependent particle parameters in southern Africa derived from airborne particle counters

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ABSTRACT

The distribution of aerosols in the atmosphere is highly variable in both time and space. Over southern Africa the aerosols are influenced by a highly stratified stable atmosphere [1]. The relative position of the aerosols in the atmosphere as well as their optical characteristics will determine the radiative significance at a local and regional scale. It has been shown that the net radiative effect of aerosols in the atmosphere is most important close to major aerosol sources.

The purpose of this paper is to present first results and to describe the methodology to derive aerosol microphysical and optical properties over southern Africa in terms of the regional vertical stratification, number of particle modes, and particle scattering coefficient, using airborne measurements of aerosol size distribution. A case study shows that aerosols in the boundary layer contribute to around 0.4 in optical thickness at 440 nm while transported biomass burning, composed of particles slightly larger, and located between 2600 and 3700 masl, contributes equally to another 0.4 in optical thickness.

1. DATA AND METHODOLOGY

Data utilised for this paper have been collected over South and southern Africa between 1997 and 2001, covering the dry (June-September) as the wet season (December-March). Aircraft data include flights from the Aerosol Recirculation and Rainfall Experiment (ARREX) and SAFARI 2000 projects. More than 50 flights have given a good spread of data over the subcontinent. The South African Weather Service aircraft were instrumented with either an Active Scattering Aerosol Spectrometer Probe (ASAP 100) or a Passive Cavity Aerosol Spectrometer Probe (PCASP 100x). Both aerosol probes categorise sounded individual particles into one of 15 size channels, based on the angular deviation of the scattered light signal. The ASASP is designed to measure aerosols with diameter between 0.12 and 3.12 µm, while the PCASP detects aerosols of diameter 0.1-3.0 µm (essentially accumulation mode aerosols). The mode of operation of the ASASP and PCASP differs only in the laser used to illuminate particles [2]. Measurement limitations of these probes have been discussed in detail previously [2]. Flights were conducted in order to characterise the ambient aerosol distribution both horizontally and vertically. Typically the flights were conducted from ground level (over the Highveld approximately 1500 masl) to 4000 masl.

In addition to the airborne measurements, ground based CIMEL sunphotometers, part of the AERONET network [3], were located at several positions on the subcontinent between 1997 and 2001. Flights were conducted over the sites at Bethlehem, Inhaca Island, Skukuza, Pietersburg and Maun to provide a reference to our computations.

Measured aerosol size distributions were used with Mie Theory in order to compute aerosol optical properties. The aerosol scattering optical thickness and its spectral dependence were calculated from the vertically-integrated size distribution to compare with ground-based measurements of aerosol optical thickness (AOT) when available. The aerosol scattering coefficient was also derived for discrete levels in the atmosphere. Computations are made with a refractive index of 1.59-0i in order to be consistent with the calibration conditions.

2. AEROSOL CHARACTERISTICS OVER SOUTHERN AFRICA FROM IN-SITU MEASUREMENTS

2.1 Seasonal variations in atmospheric aerosol loading

At a sub-continental scale there are four main sources, namely, mineral dust, biomass burning, sea spray and industrial emissions. Biomass burning and industry account for the largest fraction of fine particulate matter that is the focus of this study. Biomass burning has a strong seasonal signature with maximum emission occurring during the burn season that extends between late May and the beginning of October. Industrial emissions (the largest being power generation) are a constant year round source of primary and secondary aerosol. During the biomass burning season aerosol concentrations increase in the northern parts of southern Africa. Boundary layer aerosol concentrations as measured by the aircraft probes show distinct increases in atmospheric loading during the winter season, associated mostly with regional scale biomass burning. Over the South African industrialized Highveld, elevated concentrations of aerosols are detected year round. During the austral summer the gradient of aerosols over the subcontinent changes so that the highest concentrations are over the south.

2.2 Vertical distribution of aerosol over southern Africa

The vertical dispersion of pollutants over southern Africa is strongly controlled by the thermodynamic structure of the atmosphere. Absolutely stable layers persist at 850, 700 and 500 hPa [1]. The 700 hPa absolutely stable layer exercises the most important control over vertical aerosol structure. Throughout the

© Proceedings of the 8th International Symposium on Tropospheric Profiling, ISBN 978-90-6960-233-2
Delft, The Netherlands, October 2009. Editors, A. Apituley, H.W.J. Russchenberg, W.A.A. Monna

year, aerosols accumulate in the atmosphere in a layer capped at 3000 masl (meters above sea level). The absolutely stable layers are a result of the subsidence stability over the subcontinent. Peak concentrations of aerosols are frequently located just below the stable layer. Due to the calm atmospheric conditions as a result of general atmospheric subsidence over the region, multiple layers of aerosols form in the atmosphere below the top of the boundary layer. During winter aerosol concentrations above the subsidence stability at 700 hPa (approximately 3000 masl) drops of dramatically. In summer the boundary layer may extend up to 500 hPa a few days per month. Cosijn and Tyson [1996] found absolutely stable layers to occur over the entire subcontinent [1]. Aerosol measurements confirm this finding. A blanket of aerosols is observed throughout the region on the vast majority of observation days.

2.3 Aerosol size distribution from in-situ measurements

Aerosol size distributions were derived from the PCASP measurements for different source regions and in the different vertical layers. On any given day each layer in the atmosphere has a distinctive size distribution profile. Distinctive characteristics of each size distribution can be associated with the following characteristics: age of the air mass, direct source contributions (industry, biomass burning, sea salt), and the relative vertical position of the layer of sampled aerosols [3]. The in-situ PCASP size distributions can typically be represented by a tri-modal log normal distribution.

3. AEROSOL CHARACTERISTICS FROM SUN PHOTOMETER MEASUREMENTS

Similar seasonal trends have been identified in the sun photometer measurements made at the various sites over the subcontinent. Monthly average Aerosol optical thickness at 500 nm ($AOT_{(500nm)}$) values range between approximately 0.15 during the summer months to 0.8 during the biomass burning season in Zambia [4]. Towards the southern part of the subcontinent $AOT_{(500nm)}$ values reach a mean monthly maximum of about 0.225. The Angström exponent varies only slightly through the year at Mongu, Zambia (close to 2). The marine site, Inhaca island has a much larger coarse mode fraction as a result of the sea spray with an Angström exponent that is consistently below 1.2 [4].

4. DERIVED AEROSOL OPTICAL PROPERTIES

Aerosol scattering coefficient at 440, 675, 870, and 1020 nm is computed according to Mie theory using 1-minute averages of size distribution acquired while the airplane was making a descent above the Pietersburg's station, on 6 September 2000, in a plume of transported biomass burning emissions [5]. Figure 1 shows the vertical profile of aerosol scattering coefficient, between the ground level (1200 masl) and 4000 masl. Aerosol scattering coefficient at 440 nm ($ASC_{(440nm)}$) increases from 200 Mm^{-1} at ground level to 270 Mm^{-1} at around 2000 masl. A slight decrease is observed around 2500 m (down to 260 Mm^{-1}) and a further increase is observed up to the maximum value of $ASC_{(440nm)}$ of 300 Mm^{-1} at around 3000 masl. The particle concentration is 2100 cm^{-3}. Considering 200 Mm^{-1} as a threshold on $ASC_{(440nm)}$ to define the aerosol layer, it is included between surface level and 3700 masl. The Ansgtröm exponent decreases from 1.9 at ground level to 0.9 at 4000 masl, indicating that the number of smallest aerosols decreases with altitude, faster than the number of largest particles.

The aerosol scattering coefficient is integrated over the altitude to provide an aerosol scattering optical thickness of 0.7 at 440 nm. It agrees with direct ground-based measurements from the AERONET station of Pietersburg which gives an aerosol optical thickness at 440 nm of 0.8. The difference can be due to i) aerosol absorption which is not considered in our computation, ii) missing particles above 4000 masl, and iii) unknown value of the real part of the refractive index. The spectral dependence of aerosol scattering is also well reproduced as the agreement is similar at all wavelengths (e.g. aerosol scattering optical thickness is 0.17 at 1020 nm while $AOT_{(1020nm)}$ is 0.19).

The airborne sounding is consistent with the ground-based observation despite the aircraft measurements being restricted to an altitude of 4000 masl, confirming the aerosol layer is confined underneath. About half aerosol extinction occurs in the boundary layer, below 2600 masl, and another half between 2600 and 3700 masl

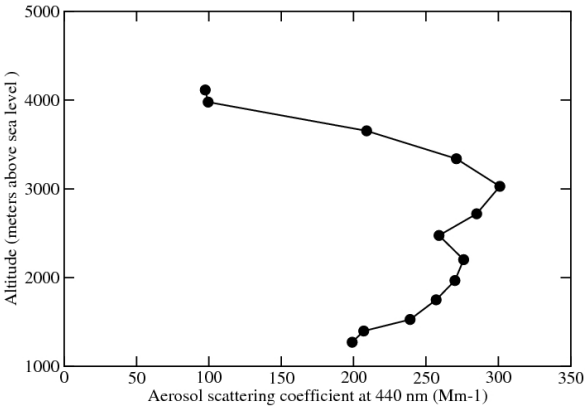

Figure 1. Vertical profile of the aerosol scattering coefficient (Mm^{-1}) computed at 440 nm from measured aerosol size distribution and according to Mie theory. Measurements are made over Pietersburg (29.45°E, 23.88°S) on 6 September 2000.

5. CONCLUSIONS

In-situ measurements over southern Africa have proved to be invaluable to characterise the variability in atmospheric aerosols. The aerosol vary greatly in their spatial (also vertical) and temporal distribution across the subcontinent. The size distribution of aerosol is highly dependant on the age and position of the plume relative to the sources. Total column aerosol optical thickness can mostly be accounted for by aerosols below 4000 masl. Individual layers of aerosols can account for a large proportion of the total column optical thickness.

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