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Seasonal variability of aerosol optical properties in Darwin, Australia

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ABSTRACT

This paper investigates the annual cycle in aerosol optical thickness (AOT) and Angstrom exponent in Darwin, Australia, a coastal site in the Tropical Warm Pool where the major aerosol sources are biomass burning and sea salt. We have used radiometer measurements from the Tropical Western Pacific Atmospheric Radiation Measurement facility for the period March 2002–June 2003. Strong seasonal cycles in AOT and Angstrom exponent were observed, peaking during the burning season (May–November). Investigation of the spectral dependence of optical thickness showed that the Angstrom formula can be satisfactorily fitted to the AOT data during the burning season but not on summer and autumn afternoons due to the presence of sea salt aerosols.

1. Introduction

Aerosols have a profound impact on the climate system: they affect the Earth's radiation budget by scattering or absorbing radiation (Charlson et al., 1992) and by altering cloud properties (Ramanathan et al., 2001). Aerosols also play an important role in air quality and health (Brook et al., 2002). Aerosol optical, physical and chemical properties have been studied in many regions of the globe; however, the large spatio-temporal variability, short lifetimes and mixing processes lead to uncertainties on the effects of aerosols on the climate system (Forster et al., 2007).

Australia, in general, has a low aerosol loading in comparison with the other parts of the globe (Box et al., 2002; Radhi et al., 2006) and in remote areas is mostly affected by biomass burning aerosols from fires in the Northern Territory (Craig et al., 2002; Russell-Smith et al., 2003) and dust outbreaks in central and southern Australia (Prospero et al., 2002). Dust aerosols at selected sites in Australia have been studied using the TOMS Aerosol Index (AI) (Herman et al., 1997; Prospero et al., 2002; Baddock et al., 2009). There have been a number of studies of biomass burning aerosols in northern Australia including the Biomass Burning and Lightning Experiment (BIBLE) (Kondo et al., 2003), airborne measurements by Tsutsumi et al. (1999), Gras et al. (1999) and Gloudemans et al. (2006). O'Brien and Mitchell (2003) used TOMS AI in combination with ground-based measurements to study biomass smoke particles in northern Australia.

Maenhaut et al. (2000) conducted a study of the aerosol composition at Jabiru in the Northern Territory, Australia in 1995–1996 and found that the fine mode contained mostly biomass burning aerosol and the coarse mode contained sea salt and mineral dust. Carr (2004), Carr and Burridge (2004), Carr et al. (2005) conducted a number of field campaign measurements in Kakadu National Park in the Northern Territory of Australia during the 2002 and 2003 dry seasons. The purpose of this study was to characterize the biomass burning aerosol in the 2003 dry season using data on aerosol concentration, light scattering, light absorption and chemical composition. Both ground-based measurements (at Jabiru) and airborne measurements were taken, along with separate aircraft measurements between Adelaide and Darwin. Carr et al. (2005) observed that changes in the smoke-plume level varied markedly with the number and intensity of fires. The aerosol mass loading was found to be lower in June than September, and the coarse mode aerosol for both months was similar, containing mostly sea salt, and some soil.

Sea salt aerosols from the global oceans are a large contributor to the total aerosols and their effects must be added to that of continental aerosols from natural or anthropogenic sources (Piazzola and Despiau, 1997; Green et al., 1992). While the scientific community has focused on the role of anthropogenic aerosols in climate change, there are a fewer studies of the sea salt aerosols in the literature (O'Dowd et al., 1997; Smirnov et al., 2002, 2003) and, to our knowledge the only study in the Northern Territory of Australia is that of Allen et al. (2008).

Field campaigns provide detailed information but long term monitoring of aerosol properties is necessary to understand seasonal and inter-annual variability. Radiometry is one way of providing this continuous coverage. The Aerosol Robotic Network (AERONET) (Holben et al., 1998) and Atmospheric Radiation Measurement (ARM) (Michalsky et al., 2001) radiometers provide

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[1] Now at: IPS Radio and Space Services, PO Box 1386, Haymarket, NSW 1240, Australia.

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continuous monitoring around the world, including the Darwin site in the Northern Territory of Australia. ARM monitoring in Darwin commenced in 2002 and AERONET in 2004; data from both are publicly available.

Darwin is located on the northern coast of Australia and in the absence of industrial pollution the main aerosol sources are biomass burning and sea salt aerosol from the local maritime environment. In this work, we focus on analyzing the seasonal variations of aerosol optical properties in Darwin using ground-based radiometer measurements from the ARM archive for the period March 2002–June 2003. In Section 2, the study area, instrumentation and data analysis methods are described. Section 3 presents the annual patterns of aerosol optical thickness (AOT) and Angstrom exponent. In Section 4, we investigate deviations from the Angstrom law and in Section 5, the seasonal characteristics of aerosol optical properties are examined to obtain more information about aerosol sources. Section 6 contains a discussion of the results and conclusions.

2. Background and methods

2.1. Location

The Tropical Western Pacific (TWP) facility in Darwin was established in 2002 as the third ARM Climate Research Facility operated by the TWP Office at Los Alamos National Laboratory to collect long-term data for the better understanding of the effect of tropical clouds on the Earth's energy budget. The facility is adjacent to the Australian Bureau of Meteorology office near Darwin International Airport at 12.425°S, 130.891°E, 29.9 m. The location was chosen because of the existence of the Pacific warm pool, its influence on weather and climate, and because it meets the scientific goal of the ARM program.

The climate of Darwin falls in the tropical wet and dry category. It experiences three distinctive climate patterns yearly: a dry continental regime from May to September; a wet, monsoonal season from December to March, and transitional periods in April–May and October–November. The summer/wet season climate is hot, very humid and rainy, and is dominated by marine winds. In the dry season the winds are mainly from the east and southeast (continental) in the morning, turning to the north and northwest (marine) in the afternoon.

Northern Australia is covered by tropical savanna with dense grass, scattered trees and grassy woodlands. In the Darwin–Kakadu and Arnhem Land regions the dominant vegetation is open eucalypt forest with an understorey of grasses. Biomass burning is a significant source of aerosol during the dry season. Burning in northern Australia begins in April, and fires become more intense as the dry season progresses, peaking around September–October.

2.2. Instrumentation

The multi-filter rotating shadow-band radiometer (MFRSR) is an instrument which directly measures the global and diffuse components of spectral irradiance using an upward looking diffuser, which is shaded at periodic intervals by a rotating shadowband. At the start of a measurement sequence the shadowband is in the nadir position and the total downward horizontal radiation is measured. Three other measurements are taken: one when the diffuser is shaded, and two others just before and after shading of the diffuser. The measurements before and after shading are used to correct for the error introduced by the shadowband blocking part of the sky during the diffuse measurement. The direct component of the radiation is given by the difference between the global and diffuse components. The direct normal component is then found by correcting for the cosine response of the instrument. A full description of the MFRSR and the algorithm for retrieving optical thickness can be found in Harrison et al. (1994) and Harrison and Michalsky (1994).

In this study we have used direct normal irradiance measurements (product level b1) for the period March 2002–June 2003. The radiometer measurements used were taken at wavelengths centred at 413, 497, 612, 667 and 868 nm by a single filtered detector with a nominal 10 nm full-width at half-maximum bandwidth at a sampling period of 20 s. Surface meteorology data were obtained from the TWP surface meteorology station. Minute-by-minute measurements of wind speed, wind direction and barometric pressure were also used.

2.3. Determination of AOT and errors

The atmospheric optical thickness (τ_A) is a dimensionless quantity which measures the depletion that a beam of radiation undergoes as it passes through a layer of the atmosphere due to suspended molecules and particles. The attenuation of the direct solar beam is given by the Beer–Bouguer–Lambert law:

$$I(\lambda) = I_0(\lambda)\exp(-m\tau_A) \tag{1}$$

where $I(\lambda)$ is the irradiance of wavelength λ at the surface, $I_0(\lambda)$ is the corresponding irradiance at the top of the atmosphere corrected for sun–earth distance, τ_A is the (columnar) optical thickness of the atmosphere and m is the air mass factor which describes the irradiance path through the atmosphere. The air mass factor can be determined from the solar zenith angle θ using $m=[\cos(\theta)]^{-1}$ as long as the sun is not too low in the sky. Otherwise, a modified expression should be used to account for the sphericity of the Earth; the expression adopted here is that of Kasten and Young (1989):

$$m = [\cos\theta + 0.50572(1.46468-\theta)^{-1.6364}]^{-1}$$

where θ is in radians.

Taking the natural logarithm of both sides of Eq. (1) yields

$$\ln(I) = \ln(I_0) - \tau_A m \tag{2}$$

If the logarithm of the irradiance (at a given wavelength) is plotted against m for many data points throughout a morning or an afternoon (Langley plot) a straight line should result. The slope, τ_A, gives the atmospheric optical thickness and the intercept $\ln(I_0)$ represents the logarithm of the extra-terrestrial irradiance at the top of the atmosphere which can be used for self-calibration of the system.

The values of I_0 and τ_A were determined using the least squares regression algorithm of Harrison and Michalsky (1994) for the air mass range $2 < m < 6$. It uses a forward finite difference filter to identify regions of positive dI/dm indicating recovery from cloud passage. The starting time for the cloud passage is estimated and the entire cloud region eliminated. A subsequent finite difference filter tests for regions of strong second derivative and regions where the first derivative is negative and more than twice the mean are eliminated. Two iterations are made on the remaining points to obtain a robust regression: the first iteration is a conventional regression which is used to eliminate points where the residual is more than 1.5 standard deviations from the regression line; the second iteration is a regression on the remaining points. For a successful regression at least 1/3 of the initial data points must remain after filtering and the residual standard deviation about the regression line must be no more than 0.009.