High aerosol optical depth biomass burning events: A comparison of optical properties for different source regions

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[1] The optical properties of aerosols such as smoke from biomass burning vary due to aging processes and these particles reach larger sizes at high concentrations. We compare the spectra of aerosol optical depth (τ_a), column-integrated volume size distributions, refractive indices, and single scattering albedo retrieved from AERONET observations for four selected events of very high smoke optical depth ($\tau_a \sim 2$ at 500 nm). Two case studies are from tropical biomass burning regions (Brazil and Zambia) and two are cases of boreal forest and peat fire smoke transported long distances to sites in the US and Moldova. Smoke properties for these extreme events can be significantly different from those reported in more typical plumes. In particular, large differences in smoke fine mode particle radius (~ 0.17 to 0.25 μm) and single scattering albedo (~ 0.88 to 0.99 at 440 nm) were observed as a result of differences in fuels burned, combustion phase, and aging. INDEX TERMS: 0305 Atmospheric Composition and Structure: Aerosols and particles (0345, 4801); 0345 Atmospheric Composition and Structure: Pollution—urban and regional (0305); 0360 Atmospheric Composition and Structure: Transmission and scattering of radiation. Citation: Eck, T. F., B. N. Holben, J. S. Reid, N. T. O'Neill, J. S. Schafer, O. Dubovik, A. Smirnov, M. A. Yamasoe, and P. Artaxo, High aerosol optical depth biomass burning events: A comparison of optical properties for different source regions, Geophys. Res. Lett., 30(20), 2035, doi:10.1029/2003GL017861, 2003.

1. Introduction

[2] Events with very high aerosol optical depths ($\tau_a > 1.5$ at 500 nm) resulting from biomass burning are not common in most regions. However they are of intense interest due to possible severe respiratory health impacts on portions of the population and due to other adverse effects such as severe visibility reduction that may impede aviation operations. Additionally, the study of high τ_a smoke cases are of interest as growth mechanisms for aging sub-micron smoke particles, such as the rate of coagulation and gas-to-particle conversion, are in part dependent on the concentration of aerosols present [Reid et al., 1998]. Therefore fine mode particle growth should reach a maximum at the highest

concentrations, given enough time for aging processes to act. From such studies upper limits for smoke particle size and subsequent optical properties can be placed. Climate change, in part from increasing greenhouse gas concentrations, has resulted in significant temperature increases at high northern latitudes, which simulations suggest will continue and may result in future increases in the incidence of biomass burning wildfires in northern boreal forest zone [Stocks et al., 1998]. Therefore very high τ_a smoke events may also become more frequent in some regions.

[3] In this study we present analyses of smoke optical properties measured during four episodes of very high τ_a from biomass burning in different regions, two from fires in the boreal zone (forest and peat fires, in Canada and Russia) and two cases from major tropical biomass burning regions (savanna and deforestation fires, in Zambia and Brazil respectively). These are rare events in most regions; that the analysis reported here was possible is a direct consequence of long-term AERONET monitoring in many regions.

2. Instrumentation and Techniques, Measurement Locations, and Fire Types

[4] All data presented in this study were acquired by CIMEL Sun/sky radiometers operated at sites that were in the AERONET global network. Instrument specifications, measurement sequences, and accuracy (spectral τ_a accuracy is $0.01-0.02$) are described in Holben et al. [1998] and Eck et al. [1999]. The algorithm of Dubovik and King [2000] was employed to retrieve the total column integrated volume size distributions, refractive indices, and single scattering albedo (ω_0) from almucantar sky radiance scans and spectral AOD. An uncertainty analysis can be found in Dubovik et al. [2000]. The estimated uncertainty in ω_0 is 0.03 for $\tau_{a440} > 0.50$. For smoke aerosol over Namibia, Haywood et al. [2003] compared co-located and simultaneous in-situ size distributions and ω_0 (from aircraft profiles) with AERONET retrievals and found very good agreement (within ~ 0.01 in mid-visible ω_0). Aircraft in-situ measurements presented by Hartley et al. [2000] for the mid-Atlantic US coast yielded a mean ω_0 of 0.96 at 450 nm (for cases of $\tau_{a450} > 0.3$), which compares well with 0.98 at 440 nm from AERONET retrievals in this region [Dubovik et al., 2002]. During the INDOEX study in the northern Indian Ocean, Ramanathan et al. [2001] have shown that AERONET retrievals of ω_0 agreed with in-situ values within the combined uncertainties and variability of both techniques.

[5] Data from sites in Maryland, USA (Goddard Space Flight Center and Baltimore, sites which are ~ 35 km apart) were analyzed for July 8, 2002. The smoke in this region and day originated from wild fires triggered by lightning

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strikes in a remote region of Quebec east of James Bay. The dry conditions, high winds, and meteorological conditions resulted in almost direct southward advection of smoke for >1600 km to the measurement sites in MD (from back trajectories computed using the Hybrid Single Particle Lagrangian Integrated Trajectory (HYSPLIT) model (www.arl.noaa.gov/ready/hysplit)). This was a unique event that allowed for analysis of smoke that was transported and aged for ~2.5 days from a known fire source. The smoke τ_a on this day was the highest τ_a ever measured at this site (except for the previous day, July 7) in ~10 years of monitoring. On July 7 late afternoon the smoke τ_a was so high that the signal from all channels <675 nm was near zero, and the τ_{a500} was estimated as ~7 (highest ever recorded in AERONET monitoring) from the spectra in the longer wavelengths ($\tau_{a870} \sim 3$).

[6] Measurements made in the capital of Moldova (Chisinau) on September 11, 2002 were analyzed for another case of long distance transport of smoke from a boreal forest region. In this case the smoke was from forests and peat fires from swampland in the vicinity of Moscow that had been drained decades earlier. The measurements in Moldova were located ~1200 km from the peat fires, with the transport distance being significantly greater. Although the aerosol from this event was dominated by smoke, it was likely that some mixing of urban/industrial pollution occurred along the transport path, as transport was >2 days to the instrument, and most of the aerosol was advected at low altitude (as suggested by the back trajectory). The smoke τ_a on this day (and the previous day, Sep 10) was the highest τ_a ever measured at this site, by about a factor of 2, in ~3 years of monitoring.

[7] Data acquired on September 15, 2000 from sites in western Zambia (Mongu and Senanga, ~95 km apart) were analyzed. This is a region with numerous local savanna fires and on this particular day, back trajectories suggest that there was also transport from regions to the northwest where there were also fires in the Democratic Republic of the Congo and southern Tanzania. Therefore the smoke on this day was a mixture of fresh and aged aerosol. The τ_a on this day was the highest smoke τ_a ever recorded in ~7 years of burning season monitoring in Zambia, when we also had good almucantar retrievals on the same day.

[8] Measurements made in the southern Brazilian Amazon on August 15, 2002, were from a site located in the Jaru biological reserve in Rondonia state. This region has undergone rapid conversion from rainforest to agricultural land within the last 3 decades, and burning of forest within the reserve boundary was observed in 2002. There is much local burning in Rondonia of pasture lands (former forests) and new forest clearing. Also trajectories suggest that smoke on this day was likely transported from other regions in the Amazon basin, especially from northern Mato Grosso, ~600 km to the east. Therefore the smoke at this site and day is also a mixture of fresh and aged smoke. Smoke $\tau_{500} \geq 2$ are not uncommon in Rondonia, Brazil during the peak of the burning season of some years (e.g. 1995 and 2002).

3. Comparison of Optical Depth Spectra, Size Distributions, and Absorption

[9] A comparison of the aerosol optical depth spectra for the 4 sites/cases at 7 wavelengths from 340 to 1020 nm is
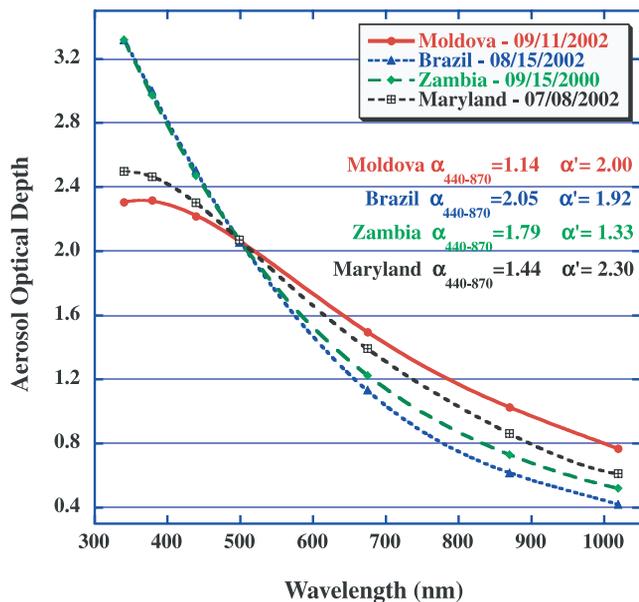

Figure 1. Aerosol optical depth spectra from 340 nm to 1020 nm for the four smoke events studied. The inter-point curves are second order regression polynomials in ln τ_a vs ln λ space.

shown in Figure 1. Although mainly by chance the τ_a for all cases are nearly equal at 500 nm (2.05 to 2.07) there are significant spectral differences of τ_a at other wavelengths. For example, at 340 nm the τ_a difference between the Brazil and Zambia cases and the Maryland and Moldova cases is ~0.8 to 1.0, while at 1020 nm the difference between the Moldova case and the Brazil case is ~0.35. This results in large differences in the Angstrom wavelength exponent (α; computed here from the 440 to 870 nm wavelength (λ) interval), ranging from 2.05 for the Brazil case to 1.14 for the Moldova case. Additionally, the τ_a spectra show a high degree of non-linearity in ln τ_a versus ln λ space. We quantify this non-linearity by a second order fit of ln τ_a versus ln λ and define $\alpha' = d\ \alpha/d\ \ln \lambda$ [*Eck et al.*, 1999]. There is a large range of α' values varying from ~1.3 to 2.3 (all high values), mainly as a result of differences in fine particle size distribution and secondarily due to refractive indices [*Eck et al.*, 2001; *O'Neill et al.*, 2001]. The second order polynomial fit of ln τ_a versus ln λ matched the measured τ_a to within 0.025 or less while the difference between the linear (Angstrom) fit and the measurements was as large as 0.46.

[10] The retrieved aerosol size distributions for these cases (Figure 2) show significant differences in fine mode particle size, with the Zambian smoke having the smallest radius particles (peak volume modal radius, r = 0.17 μm) and the Moldova smoke the largest (r = 0.25 μm). The dynamic (as a function of τ_a) smoke size distribution models of *Dubovik et al.* [2002] are in reasonable agreement (r to within ~0.02 μm) with the fine mode sizes of all cases presented here except the Moldova case which exhibits fine mode radius that is ~0.07 μm greater than the model for boreal forest smoke. The fine mode strongly dominates over the coarse mode for all four cases with the ratio of τ_{fine}/τ_{total} > 0.97 at 440 nm and is >0.90 for all cases and wavelengths
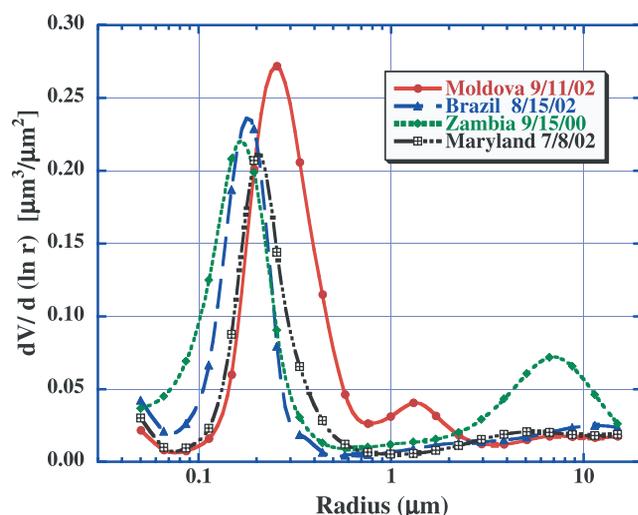
Figure 2. Aerosol volume size distributions retrieved from AERONET measurements for the 4 cases.

(except for the Zambian case at 1020 nm when it is 0.86). The differences in fine mode particle size modulate the wide range in visible-near infrared α, which is often governed mostly by the influence of fine versus coarse mode τ_a. The large differences in fine mode size between the cases (Figure 2) are due to several different factors. First, in tropical biomass burning regions of Brazil and Zambia, burning from numerous fires is so prevalent that fresh and aged smoke are often combined. By nature fresh smoke is smaller [*Reid et al.*, 1998]. Similarly, smoke from flaming combustion tends to have smaller particles than from smoldering combustion [*Reid and Hobbs*, 1998]. This may partly account for Zambia having smaller particle size than Brazil since African savanna fires consume a higher percentage of biomass (mostly grass) in the flaming phase than Amazonian deforestation fires.

[11] Unlike the tropical regions, smoke from the Quebec fires remained isolated during transport. As this smoke was advected >1600 km over two days the probable shift to larger particle size (r = 0.20 μm) by coagulation and condensation was more evident.

[12] The Moldovan case was the most extreme event observed. Here, the very large particle size may result from a combination of smoldering combustion from peat fires and the production of more hygroscopic aerosols. Peat fuels may contain significant amounts of sulfur and combustion may lead to subsequent production of hygroscopic sulfate aerosol species. Additionally the smoke is likely mixed with some urban/industrial pollution from Moscow and other cities during its transport to Moldova, thus potentially adding significant hygroscopic aerosols to the smoke/pollution mixture. It is noted that the more hygroscopic fine mode pollution particles in the US mid-Atlantic region [*Kotchenruther and Hobbs*, 1998] reach sizes similar to the Moldova case, when AOD and humidity are high ($\tau_{a440} > 1.0$, RH > 70%). The lower retrieved real refractive index of the Moldovan smoke, ~1.48 versus 1.53−1.54 for the other 3 cases is consistent with greater hygroscopic growth for that case as well as for the increased organic carbon content of smoldering fires. *Nakajima et al.* [1999]

also retrieved large size accumulation mode particles (peak radius = 0.25 μm), the same size as the Moldova case, for smoke (at similarly high τ_a) from Indonesian fires in 1997 during the El Nino episode when there were many peat fires. The smoke size distribution measurements of *Nakajima et al.* [1999] (also derived from sky almucantar measurements although through a different algorithm) were taken in Singapore where local urban/industrial pollution was also mixed with the smoke, with smoke estimated as ~75% to 90% of the total aerosol. The widths (geometric standard deviations) of the fine mode size distributions for all four cases ranged from ~1.35 to 1.50 which is similar to but somewhat narrower than the values given in the *Dubovik et al.* [2002] smoke aerosol climatology of 1.49 to 1.54.

[13] Aerosol single scattering albedo shows a wide range of absorption magnitude for these cases (Figure 3). The highest ω_0 values were measured for the Moldova case, due to the larger particle size (greater scattering) and the small imaginary refractive index (Table 1). The ω_0 measured at the AERONET site in Moscow on September 7, 2002, in close proximity to the peat fires, was 0.95−0.96, with $\tau_{a500} = 2.15$. The fine mode particle radius was ~0.20 μm on this date in Moscow. Therefore the ω_0 in Moldova on September 11, 2002 increased by ~0.03−0.04 over values measured near Moscow (and radius increased by ~0.05 μm), probably due to aging, hygroscopic growth, and mixing with other pollutants. The smoke transported to Maryland from the Quebec fires also had relatively low absorption ($\omega_0 \sim 0.96−0.97$) suggesting that the majority of the aerosol production from these boreal fires was from the smoldering phase. At the other extreme, the most absorbing case was in Zambia, where the smoke is largely produced by flaming phase combustion of savanna grass, which produces much more black carbon than woody fuel combustion from deforestation fires [*Ward et al.*, 1992, 1996] or from peat fires. Intermediate in magnitude is the spectral ω_0 of smoke from the Brazil case that is produced by fires from both pastures
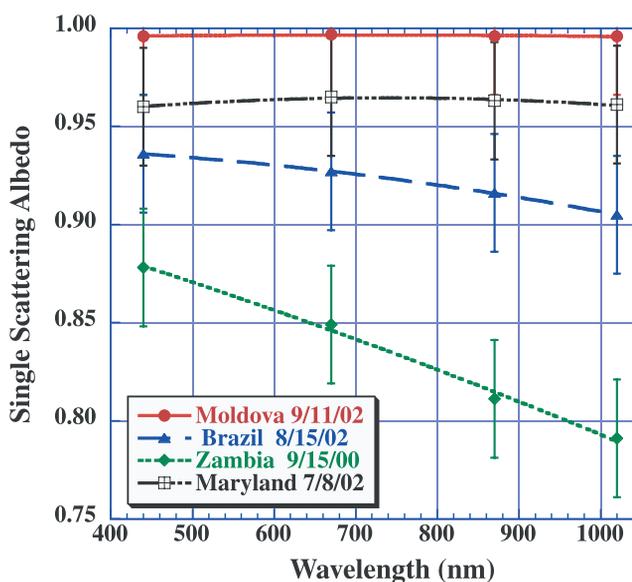

Figure 3. Aerosol single scattering albedo retrieved from AERONET measurements for the 4 cases. The error bars show the 0.03 uncertainty (estimated) in retrieved ω_0.

Table 1. Refractive Index (Imaginary Part) for Sites/Dates Shown in Figures 1–3

Site	440 nm	675 nm	870 nm	1020 nm
Zambia	0.0256	0.0247	0.0248	0.0232
Brazil	0.0138	0.0123	0.0108	0.0102
Maryland	0.0076	0.0060	0.0055	0.0052
Moldova	0.0005	0.0005	0.0005	0.0005

and deforestation sites with the smoke of mixed age. The imaginary refractive index (k_i) shows relatively little spectral variation for all four cases presented here (Table 1) which is consistent with the relatively small spectral dependence of k_i for black carbon in this wavelength region [*Bergstrom et al.*, 2002]. It is noted that the seasonal and multi-year averages of ω_0 given by *Dubovik et al.* [2002] for the African savanna are within ~0.01 of the values retrieved for the Zambia case shown in this study, and similarly the ω_0 given by *Dubovik et al.* [2002] for the Amazonian forest are within ~0.01 of the values retrieved for the Brazil case shown in this study. However, the ω_0 of *Dubovik et al.* [2002] for the boreal forest average is ~0.02 to 0.08 lower (larger differences at the longer wavelengths) than the 2 cases presented here of smoke that originated in the boreal zone and were transported long distances.

4. Conclusions

[14]   1. A comparison of the aerosol optical depth spectra from 4 cases of very high smoke aerosol loading ($\tau_a \sim 2$ at 500 nm) showed a wide range in 440–870 nm Angstrom exponent, ~1.1 to ~2.1 primarily as a result of differences in fine mode particle size and also due to differing refractive indices.

[15]   2. Large differences in smoke fine mode particle radius (~0.17 to 0.25 μm) were observed for these cases as a result of differences in fuels burned, combustion phase, and aging. This compares to a range of radius of ~0.13 to 0.16 μm for moderate to high smoke aerosol loadings ($\tau_a \sim$ 0.5 to ~1.0 at 440 nm) in various biomass burning regions [*Dubovik et al.*, 2002].

[16]   3. Large differences in smoke single scattering albedo (~0.88 to 0.99 at 440 nm) were also observed as a result of differences in fuels burned, combustion phase, and aging.

[17]   4. Both cases of smoke from boreal region fires that had been transported long distances (and therefore aged) had both larger particle size and higher single scattering albedo than for smoke measured within boreal regions. Smoke resulting in part from peat fires in Russia had very high ω_0 of ~0.99 and very large radius (r = 0.25 μm) likely due to smoldering combustion of peat fuels and also subsequent hygroscopic particle growth.

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