Optical properties of boreal region biomass burning aerosols in central Alaska and seasonal variation of aerosol optical depth at an Arctic coastal site

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Earth's land surface or ~16 million square kilometers across northern Eurasia and North America. The arctic and boreal zones currently store the globe's largest reservoir of soil carbon, at 25-35% of the total, largely in organic soil layers in the permafrost [Bonan and Shugart, 1989; Melillo et al. 1995]. Both climate change simulations of the effects of increasing atmospheric greenhouse gas concentrations and recent observations show that global temperature increases are the largest in the arctic and boreal regions [Soja et al., 2007; IPCC, 2007]. As a result of these temperature increases (and drier conditions) models predict future increases in area burned in the boreal zone. For example Flannigan et al. [2005] simulated a 74-118% increase in burned area in Canada by 2100 in a 3XCO2 increase scenario. Kharuk et al. [2008] found a one-third reduction in fire return interval time for larch dominated forests in Siberia from the 19th to the 20th century, related to warming temperatures in northeast Siberia. Severe forest fires that also burn insulating organic and peat soil layers result in deeper soil thawing [Kharuk et al., 2008] and thereby may release large amounts of stored carbon into the atmosphere.

Concurrent with a significant warming trend, Kasischke and Turetsky [2006] found that the annual boreal forest area burned in Alaska and Canada doubled from the 1960s/70s to the 80s/90s and the proportion of burning in the early and late growing seasons increased. In Alaska seven of the eleven largest fires in a 56 year interval (1950-2005) have burned since 1988, with the largest area burned on record occurring in 2004 and the third largest in 2005 [Soja et al., 2007]. Across the entire circumboreal zone the frequency of extreme fire years has increased. Kasischke et al. [2002] have found that high fire years in Alaska consist of larger fires occurring later in the growing season. In low precipitation years peat burning is expected to increase as the summer advances due
strong boreal burning years they attribute ~80% of the global BC/snow forcing to be from anthropogenic fossil fuel and biofuel sources.

Only recently have studies investigated the optical properties of boreal region biomass burning aerosols in intensive burning years [Stohl et al., 2006b; Myhre et al., 2007]. Pfister et al. [2008] discuss the need for better characterization of the optical properties of boreal region biomass burning smoke particles, especially for the case of significant peat burning. In this investigation we present an analysis of a long time series of aerosol optical depth measurements from 1994 through 2008 at an AERONET sun-sky radiometer site located in the boreal forest zone of central Alaska. In the extreme burning years of 2004 and 2005 the AOD was very high, allowing for accurate characterization of the spectral imaginary refractive index (absorption) and single scattering albedo (ω₀) from almucantar retrievals. Additionally, we compare the particle size distributions and ω₀ of these fine mode dominated smoke aerosols to the size distributions and absorption of smoke aerosols from other major biomass burning regions. We also present AOD data from a site on the Arctic Ocean coast in Alaska to compare the seasonality and frequency of smoke transport to the Arctic (primarily in summer) to the springtime Arctic haze impacts on optical depth. The Arctic haze phenomenon has been attributed primarily to the long-distance transport of aerosols from industrial source regions (Shaw, 1995).

2. Instrumentation, Study Sites and Techniques

2.1 Study Region and Sites

The principal AERONET site analyzed in this study is the Bonanza Creek, Alaska site located in the boreal forest biome of central Alaska (Figure 1). This is a National Science
every 15 minutes at 340, 380, 440, 500, 675, 870, 940, and 1020 nm (nominal wavelengths). The direct sun measurements take ~8 seconds to scan all 8 wavelengths, with a motor driven filter wheel positioning each filter in front of the detector. These solar extinction measurements are then used to compute aerosol optical depth (AOD, $\tau_a$) at each wavelength except for the 940 nm channel, which is used to retrieve total columnar (or precipitable) water vapor in centimeters. The filters utilized in these instruments were ion assisted deposition interference filters with bandpass (full width at half maximum) of 10 nm, except for the 340 and 380 nm channels at 2 nm. The estimated uncertainty in computed $\tau_a$, due primarily to calibration uncertainty, is ~0.010-0.021 for field instruments (which is spectrally dependent with the higher errors in the UV; Eck et al. [1999]). Schmid et al. [1999] compared $\tau_a$ values derived from 4 different solar radiometers (including an AERONET sun-sky radiometer) operating simultaneously together in a field experiment and found that the $\tau_a$ values from 380 to 1020 nm agreed to within 0.015 (rms), which is similar to our estimated level of uncertainty in $\tau_a$ retrieval for field instruments. The spectral aerosol optical depth data have been screened for clouds following the methodology of Smirnov et al. [2000], which relies on the greater temporal variance of cloud optical depth versus aerosol optical depth. The sky radiances measured by the sun/sky radiometers are calibrated versus the 2-meter integrating sphere at the NASA Goddard Space Flight Center, to an absolute accuracy of ~5% or better.

2.3 Inversion Methodology
to demonstrate successful retrievals of mode radii and the relative magnitude of modes for various types of bimodal size distributions such as those dominated by a sub-micron accumulation mode or distributions dominated by super-micron coarse mode aerosols. To ensure sufficient sensitivity to aerosol absorption, only almucantar scans where AOD(440nm)>0.4 [Dubovik et al., 2000] were analyzed for the investigation of the characteristics of spectral refractive indices and single scattering albedo.

3. Results and Discussion

3.1 Temporal and Spectral Variability of AOD in central Alaska

3.1.1 Monthly and inter-annual variation in AOD and Angstrom exponent

The monthly climatology of 500 nm AOD and Angstrom Exponent (440-870 nm) at Bonanza Creek, Alaska showing monthly means from multiple years of observations is shown in Figure 2. The 440-870 nm Angstrom is computed from linear regression of In AOD versus In λ scale at 440, 500, 670 and 870 nm. The multi-year monthly means are computed as a mean of the individual monthly averages. The seasonal trend in monthly average AOD shows a steady increase of AOD from near background levels in March (~0.06) to values exceeding ~0.14 for June, July and August and then a rapid 2-month decline down to background again (~0.05) in October. It should be noted that the inter-annual variability in AOD is extremely large (see Figure 3, discussed below) due to the episodic nature of boreal fires with severe and numerous fires occurring in drought years, compared to few fires in wet years. Fine mode biomass burning aerosols dominate this seasonality as the Angstrom exponent increases in summer (June-August) when most burning occurs.
and 2008. However, in 2004 there were similar number of fires in July as August while
the AOD at Bonanza Creek was >2.5 times higher in August. Daily area burned estimates
for Alaska and Canada combined as shown by Stohl et al. [2006] reach a maximum from
late June through July, and much less area burned in August. This suggests the possibility
that factors other than area burned and numbers of fires are important in determining the
total atmospheric column AOD. Other factors in addition to area burned that would
influence the AOD are meteorological conditions (including wind speed and direction,
atmospheric stability, and precipitation), intensity of burning since intense fires enhance
convection that may loft smoke to higher altitudes where winds are higher, and the types
of fuel burned and the phase of combustion [Reid et al., 2005b]. It is noted that AOD
measurements at a point location (Bonanza Creek) are also strongly affected by the
location of the fires in the entire state of Alaska relative to the wind direction and other
meteorological factors. Satellite hot spot remote sensing robustly detects fires in the high
temperature flaming phase of combustion but it is much more difficult to detect lower
temperature smoldering phase fires. However the flaming phase is typically of relatively
short duration, while the larger diameter woody fuels in a forest and the organic soils and
peatlands may burn for several days in the smoldering phase. As previously mentioned,
Turquety et al. [2007] estimated that the proportion of carbon monoxide emissions was
double the proportion of peat area burned (~37% of the carbon monoxide emissions from
only 17% of the area burned) in 2004 for fires in Alaska and Canada. This suggests that
peat burning may possibly also have resulted in a disproportionate amount of total
aerosol emissions, relative to the total area burned.
Based on the assumption that aerosol size distributions are bimodal, O'Neil et al. (2001, 2003) have developed a spectral deconvolution algorithm (SDA) that utilizes spectral total extinction AOD data to infer the component fine and coarse mode optical depths. An additional fundamental assumption of the algorithm is that the coarse mode Angstrom exponent and its derivative are both close to zero. The Angstrom exponent $\alpha$ and the spectral variation of $\alpha$ (as parameterized by $\alpha' = d\alpha/d\ln \lambda$) are the measurement inputs to the SDA. These continuous-function derivatives (usually computed at a reference wavelength of 500 nm) are derived from a second order fit of $\ln \tau_\lambda$ versus $\ln \lambda$ (Eck et al., 1999). The spectral AODs employed as input to the SDA were limited to the six CIMEL wavelengths ranging from 380 to 1020 nm. Figure 6 shows the time series of fine and coarse mode daily average AOD at 500 nm from the SDA algorithm for June 1 through September 12, 2004 at Bonanza Creek. These are level 2 cloud screened data that have had the SDA algorithm applied to the AOD spectra. It is noted that the coarse mode is typically very low and nearly constant, while the fine mode AOD from biomass burning exhibits very large day-to-day variability. Only 2 days show the coarse mode AOD to be significantly higher than the fine mode, possibly due to dust transported from Asia or from residual cloud contamination. We assume that the fine mode aerosols at Bonanza Creek are dominated by biomass burning smoke since there are numerous fire hot spots observed (Figure 1), combined with a lack of any other significant regional sources of fine mode particles.
The SDA algorithm computed daily average fine mode fractions (FMF) for the Bonanza Creek site are plotted versus $\alpha_{380-500}$ in Figure 8, and compared with measurements made in Brazil and Zambia (from the same observations as shown in Figure 7). For the same range of FMF, 0.8 – 1.0, the $\alpha_{380-500}$ is typically significantly lower for the Alaska site than the Brazil and Zambia biomass burning sites, again suggesting much larger accumulation mode particles in Alaska. In addition to the greater percentage of smoldering combustion for the forest regions with woody fuels (Alaska and Brazil) other possible reasons for larger accumulation mode particles include higher AOD levels (Figure 7) in Alaska and Brazil that increase the coagulation rate since aerosol concentrations are higher. Another possible factor in creating large accumulation particles might be the characteristics of smoke from peat burning in Alaska [Reid et al., 2005a], since extensive areas of peat lands (in addition for forests) were observed to burn during the summer of 2004.

### 3.2 Retrievals of aerosol size distribution and single scattering albedo in central Alaska

#### 3.2.1 Volume size distributions

The almucantar retrievals of aerosol volume size distributions for the Bonanza Creek site in 2004 and 2005 for scans where AOD(440nm)>0.4 are shown in Figure 9a. These averages are plotted as a function of AOD(440nm) with AOD bins from 0.4-0.8, >0.8-1.0,...>2.4-2.8, >2.8 resulting in averages of from 9 to 34 almucantars per bin. At all AOD levels the retrievals show the dominance of fine mode aerosols, and the $\alpha_{440-870}$ for the bins range from 1.74 at the lowest AOD(440) average of 0.53 to 1.36 at the highest
exhibiting $r_v \sim 0.21 \text{ \mu m}$ and transported smoke from a Russian peat/forest fire showing retrieved $r_v$ of 0.28 $\text{\mu m}$ (Version 2 retrievals as compared to somewhat smaller radius values reported from Version 1 in Eck et al. [2003a]). These however were aged aerosol events with smoke age > 2 days due to long distance transport from distinct source regions. However, AERONET measurements made in Moscow on September 7, 2002 (AOD(440) ~2.5) near to a fire that was predominately burning peat had large accumulation mode particles of ~0.22 $\text{\mu m}$ radius despite relatively short transport distance and therefore likely little aging. Therefore, the large radius of the smoke aerosol measured in late summer in Alaska in 2004 and 2005 possibly resulted in part from the smoldering combustion of peat fuels in addition to the high aerosol concentrations that would result in greater coagulation, condensation and secondary production rates.

In comparison to the Alaska smoke we show the aerosol volume size distribution retrievals for biomass burning aerosols from the Mongu, Zambia AERONET site in Figure 11a. These retrievals are from September (peak burning month) data only for the years 1997-2005, and shown for two AOD levels, ~0.5 and ~1.3 at 440 nm. Both the mean sizes and the shift of fine mode radius as AOD increases in Zambia are relatively small (radius ~0.14 to 0.16 $\text{\mu m}$) as compared to the boreal smoke measured in Alaska. Smoke in Brazil (southern Amazonia; not shown) exhibited slightly larger fine mode radius (~0.15 to 0.17 $\text{\mu m}$; Schafer et al., 2008) at these AOD levels than Zambia, possibly due to more smoldering combustion of woody fuels and higher aerosol concentrations that may have lead to greater coagulation rates. In addition to the relatively small change in volume median radius in Mongu, Zambia, the width of the fine mode size distribution is narrower for the Zambia smoke than for the Alaska smoke. The geometric standard
The average single scattering albedo in 2004 and 2005 at Bonanza Creek is high (weak absorption) for biomass burning aerosols, with most values ranging from ~0.96 to 0.97 (Figure 9b). Only four of the individual retrievals out of a total of 124 almucantars with AOD(440nm)>0.4 in 2004 and 2005 had \( \omega_0 \) less than 0.935 at 440 nm, with the lowest at 0.909. The highest retrieved value of \( \omega_0 \) at 440 nm was 0.996. For all but the lowest AOD level the average \( \omega_0 \) at 440 nm is slightly lower (by ~0.01) than that at 675 nm, and the \( \omega_0 \) from 675 nm through 1020 nm are relatively constant (Figure 9b). This spectral dependence of \( \omega_0 \) appears to be anomalous for biomass burning aerosols, as the single scattering albedo typically decreases with increasing wavelength in both measurements and retrievals [Reid et al., 2005b]. For example, Dubovik et al. [2002] show this typical wavelength dependence for biomass burning aerosols from four major regions: Amazonian forest, S. American cerrado (savanna-like), African savanna in Zambia, and boreal forest (primarily Canada). However, the boreal forest data set in Dubovik et al. [2002] does not include events with as high AOD as occurred in Alaska in 2004 and 2005, nor does it include observations with significant peat burning. For all other years from 1994-2007 (excluding 2004 and 2005) there were a total of only 14 almucantar retrievals at Bonanza Creek with AOD(440nm)>0.4. Two of these had \( \omega_0 \) at 400 nm of ~0.86 which suggests flaming phase crown fires, while the other 12 had values ranging from 0.92 to 0.98. The mean of these 14 almucantars was 0.94 at 440 nm and within less than 0.005 of the values given by Dubovik et al. [2002] at all 4 wavelengths (Dubovik's values were a mean for boreal forest biomass burning aerosols).

Similar wavelength dependence of \( \omega_0 \) to the Alaskan smoke of 2004 and 2005 was observed however for the previous mentioned case of peat burning smoke in Moscow on
higher and relatively constant (~0.72). For the burning of lignite fuel Bond et al. [1999] also measured greater fine particle absorption at shorter wavelengths (450 nm), implying larger imaginary refractive index at shorter wavelengths, possibly due to absorption by organic carbon. It is noted that their laboratory combustion of this lignite fuel occurred partially in the smoldering phase. However it is noted that the absorption properties of organic carbon are not well known and therefore are a topic of much recent research.

Another factor that contributes to the relatively constant spectral single scattering albedo of the Alaskan smoke is the much larger size and wider distribution of the fine mode particle radius, as compared to particles from most other biomass burning regions. Because the scattering cross section (or hence scattering optical depth) increases more rapidly than absorption cross section with increasing particle size the SSA also increases (in the absence of variations in refractive index) with increasing particle size. This trend is the optical equivalent of stating (as above) that SSA typically decreases with increasing wavelength. However this trend is less extreme at larger particle sizes because the SSA approaches unity at a lesser rate with respect to increasing particle size (or decreasing wavelength).

In situ measurements from nephelometer and particle soot absorption photometer data at the surface in Barrow, Alaska on July 3-4, 2004 yielded an aerosol single scattering albedo of 0.96 at 550 nm [Stohl et al., 2006; Stone et al., 2008], for a case of very high smoke AOD advected from fires in central Alaska and the Yukon. This is essentially equal to the mean AERONET retrievals of $\omega_0$ interpolated to 550 nm for the total column aerosol in central Alaska at Bonanza Creek (Figure 9a). For agricultural smoke originating in Europe and subsequently advected to Svalbard in the Arctic, Myhre et al.
conditions, Lewis et al. [2008] measured AAE as high as 2.5 (532 to 870 nm) at high \( \omega_0 \)
(near unity) while AAE values approached 1.0 for \( \omega_0 < 0.8 \) at 532 nm. The measured
organic carbon fraction to total carbon was highest for the smoke with the highest AAE
and \( \omega_0 \), thereby suggesting that the enhanced absorption AAE results from light
absorbing organic carbon.

3.3 Seasonal variation of AOD in the coastal Arctic at Barrow, Alaska

In this section we present data from the Arctic AERONET site located at Barrow, Alaska on the Beaufort Sea coast (Figure 1). The data collection at this site has many
more gaps than for the Bonanza Creek site in central Alaska due to its more severe
weather, which sometimes resulted in instrument electronic or mechanical problems. This
occurred especially in earlier years before a modified version of the CIMEL that
incorporated heating elements was deployed. Additionally there is more persistent cloud
cover at this site than at Bonanza Creek, resulting in fewer observations of the sun,
therefore less AOD measurements and very few almucantar scans of sky radiance
however measurements were only made during the peak Arctic haze month of April in
three years, 2002, 2005 and 2008, when data collection began in late March or early
April. All years had monitoring from July through September, and four of the six years
had data from May through September, thus covering the biomass burning season.
Therefore due to the numerous gaps in data acquisition, the AERONET data presented
here for Barrow cannot be considered a fully representative monitoring record, especially
for the spring arctic haze season. These data gaps therefore preclude any analysis of
solar zenith angle. Stohl et al. [2006a] have shown from transport modeling that this
smoke event originated from the fires located in central Alaska and the Canadian Yukon,
and that the smoke continued to be transported beyond Barrow and deep into the arctic.
Their simulations suggested that smoke from this event reached the North Pole on July 8,
2004 although cloud cover precluded verification from satellite images. The AERONET
site located at Resolute Bay, Canada (74° 44’ N, 94° 54’ W; ~1950 km ENE from
Barrow) measured smoke AOD on July 5, 2004 as high as 2.3 at 500 nm ($\alpha_{440,870} > 1.3$
from this same arctic transport event. A Terra MODIS image (Figure 7 in Stohl et al.
(2006)) shows widespread smoke from Alaska and the Yukon through the Arctic islands
Figure 12b is the same as 12a but with the extreme AOD event of July 3, 2004
excluded. Individual daily averages of AOD are shown as well as 20 day interval means
(means computed with the one extreme day removed also). Seasonality of AOD is
evident, with the highest 20-day averages of AOD occurring during the arctic haze season
from late March through mid-May. These higher average AOD values result partly from
the lack of measured low background AOD in the spring, when values are rarely lower
than 0.07. In contrast the daily average AOD during summer months is often <0.04 and
as low as 0.02. However daily average AOD on some summer days, as high as or higher
than the spring Arctic haze AOD, from biomass burning aerosols result in mean values of
AOD during the summer that are elevated significantly above background levels. Again it
is emphasized that due to a much less extensive data record at Barrow (as compared to
Bonanza Creek), and since the extreme Alaska burning years of 2004 and 2005 are
monthly average Angstrom Exponent (440-870 nm) at both sites, 1.25 at Barrow and 1.10 at Bonanza Creek.

Shaw [1982] measured AOD from sunphotometer at Barrow during the mid to late 1970s and computed a March-April mean of 0.135 at 500 nm. Therefore the mean AOD measured by AERONET for the spring arctic haze (and smoke) at Barrow for 2002, 2005 and 2008 were somewhat higher than that measured ~25 years earlier. Bodhaine and Dutton [1993] presented measurements of AOD at Barrow computed from broadband (300-690 nm) pyrheliometer measurements for the years 1977-1992, with estimates of volcanic aerosol optical depth removed. They show relatively low AOD in 1980 and 1981 and a significant downward trend from 1982 (peak year) to 1992 that they suggest may have resulted from the reduction of emissions in the Soviet Union and Europe during that era. Continued monitoring of AOD at Barrow is important for understanding arctic haze magnitude and trends. Ground based photometric measurements of AOD at 532 nm at the arctic island of Spitsbergen (~79N, 12E) by Herber et al. [2002] from 1991 through 1999 showed a gradual increase in AOD of ~9% over the 9 year interval. The relative sparseness of long-term records of AOD at Arctic locations coupled with the possibility of different regional influences (sources and meteorology) make it very difficult to assess trends of aerosol loading across the entire arctic region.

4. Summary and Conclusions

Aerosol optical properties data acquired from monitoring at two AERONET sites in Alaska were investigated. Data from long-term monitoring at a central Alaska boreal
3. Absorption by the smoke aerosol in Bonanza Creek in 2004 and 2005 was very weak, with retrieved single scattering albedo ranging from ~0.96 to 0.97, along with relatively flat spectral dependence. These high single scattering albedos result from small values of the imaginary index of refraction, implying low black carbon fraction probably due to predominately smoldering combustion, coupled with large fine mode particle radius which results in greater scattering efficiency (increased $\omega$, amplitude) and reduced wavelength dependence of the $\omega$. Additionally, the single scattering albedo at 440 nm was ~0.01 lower than at the longer wavelengths due to a somewhat larger imaginary refractive index at 440 nm, which is possibly due to enhanced short wavelength absorption by organic carbon aerosols. This also suggests the possibility that smoke from peat burning (smoldering combustion) had a significant influence on aerosol emissions.

4. Although AERONET monitoring at the Arctic coastal site of Barrow from 1999 to 2008 was often interrupted and not complete enough to be considered a representative climatology, some seasonal characteristics of AOD were nonetheless evident. The average AOD in the spring (late March through late May) is higher than the average AOD in the summer. Even though several individual daily mean values in summer are significantly higher (from transported biomass burning smoke) than most daily means in spring, the lack of very low background AOD levels in spring, due primarily to persistent industrial arctic haze, resulted in higher mean AOD in spring.

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Figure 5. Remotely sensed fire counts for the Alaska region from the MODIS sensor on the Terra satellite for the years 2002 through 2008.

Figure 6. Spectral Deconvolution Algorithm (SDA) computed daily average fine and coarse mode AOD versus day of the year from June 1, 2004 through September 12, 2004 at Bonanza Creek, Alaska.

Figure 7. A comparison of the daily average 380-500 nm Angstrom exponents as a function of 500 nm AOD for the Bonanza Creek data from 2004 versus the AERONET data from the major tropical region biomass burning sites of ABRACOS Hill, Brazil (2002 data; southern Amazonia) and Mongu, Zambia (2004; southern Africa savanna burning region). For all three sites the data for the June through October biomass burning seasons are shown.

Figure 8. Spectral Deconvolution Algorithm (SDA) computed daily average fine mode fraction (FMF) for the Bonanza Creek site plotted versus $\alpha_{380-500}$ and compared with measurements made in Brazil and Zambia (from the same observations as shown in Figure 6).

Figure 9. Almucantar retrievals of a) aerosol volume size distributions b) single scattering albedo, and c) imaginary part of the refractive index from the Bonanza Creek site in 2004 and 2005 for scans where AOD(440nm)>0.4. These averages are plotted as a function of
Figure 3. Time series of a) monthly average AOD at 500 nm and and b) Angstrom exponent (440-870 nm), by year for the summer and early fall months (June-September) at Bonanza Creek.
Figure 6. Spectral Deconvolution Algorithm (SDA) computed daily average fine and coarse mode AOD versus day of the year from June 1, 2004 through September 12, 2004 at Bonanza Creek, Alaska.
Figure 9. Almucantar retrievals of a) aerosol volume size distributions b) single scattering albedo, and c) imaginary part of the refractive index from the Bonanza Creek site in 2004 and 2005 for scans where AOD(440 nm)>0.4. These averages are plotted as a function of AOD(440 nm) for AOD bins from 0.4-0.8, >0.8-1.0,...,>2.4-2.8, >2.8 resulting in averages of from 9 to 35 almucantar scans per bin.
Figure 12. a) The daily average 500 nm AOD measured at the Barrow AERONET site as a function of the day of the year for all monitoring during the 1999 through 2008 time interval. b) The same as in a) but with the single outlier point of AOD (500 nm) of 3.4 from July 3, 2004 removed, and with 20 day averages computed.