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Biomass burning in Siberia and Kazakhstan as an important source for haze over the Alaskan Arctic in April 2008

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[1]   During the ARCPAC (Aerosol, Radiation, and Cloud Processes affecting Arctic Climate) airborne field experiment in April 2008 in northern Alaska, about 50 plumes were encountered with the NOAA WP-3 aircraft between the surface and 6.5 km. Onboard measurements and the transport model FLEXPART showed that most of the plumes were emitted by forest fires in southern Siberia-Lake Baikal area and by agricultural burning in Kazakhstan-southern Russia. Unexpectedly, these biomass burning plumes were the dominant aerosol and gas-phase features encountered in this area during April. The influence on the plumes from sources other than burning was small. The chemical characteristics of plumes from the two source regions were different, with higher enhancements relative to CO for most gas and aerosol species from the agricultural fires. In 2008, the fire season started earlier than usual in Siberia, which may have resulted in unusually efficient transport of biomass burning emissions into the Arctic. **Citation:** Warneke, C., et al. (2009), Biomass burning in Siberia and Kazakhstan as an important source for haze over the Alaskan Arctic in April 2008, *Geophys. Res. Lett.*, *36*, L02813, doi:10.1029/2008GL036194.

1. Introduction

[2]   Arctic haze and the gas-phase air pollutants in it have been regularly observed in the Arctic since the 1950s and are thought to be of mostly anthropogenic origin due to emissions from Europe and Asia that are transported to and trapped in the Arctic air mass during the winter and early spring [*Stohl*, 2006]. Slow removal processes result in very long lifetimes in the wintertime Arctic so that the Arctic haze can persist.

[3]   In addition to greenhouse gas induced warming and feedbacks, Arctic warming may also be caused by shorter-lived climate forcing agents that are part of the Arctic haze. Climate relevant processes include 1) direct warming of the lower troposphere by the absorption of solar radiation and IR emission by aerosol particles from anthropogenic and biomass burning (BB) sources [*Ritter et al.*, 2005], 2) acceleration of snow melt due to absorption by soot (light-absorbing carbon) deposited to the surface in springtime

[*Flanner et al.*, 2007], and 3) increases in IR emissivity of wintertime and springtime clouds in the Arctic due to the effects of anthropogenic and BB aerosol particles on cloud properties [*Lubin and Vogelmann*, 2006].

[4]   BB is a large source of aerosols usually to the late spring- and summer-time Arctic. Boreal forest fires caused increases in pollution levels in different parts of the Arctic in summer 2004 [*Stohl et al.*, 2006]. Record high pollution levels were found in the European Arctic in May 2006 as a result of agricultural fires in Eastern Europe [*Stohl et al.*, 2007]. Recent increases in total BB and earlier starts to the fire season, possibly because of warming in the boreal regions, have led to the speculation that the contribution of black carbon (BC) deposition from boreal forest fires to Arctic ice melt has been increasing [*Kim et al.*, 2005].

[5]   In this paper we present airborne observations of about 50 pollution plumes transported to the Alaskan Arctic at various altitudes already in April 2008. The plumes were caused by fires that were burning unusually early in the season in boreal forests in southern Siberia and by agricultural fires in northern Kazakhstan.

2. Measurements and Model Description

[6]   An airborne field experiment, the Aerosol, Radiation, and Cloud Processes affecting Arctic Climate (ARCPAC) was conducted in Fairbanks, Alaska in April 2008 using the NOAA WP-3D aircraft. A major focus of this campaign was to investigate the chemical, optical, and microphysical characteristics of aerosols and gas phase species in the Arctic springtime and to determine the source types (industrial, urban, biomass/biofuel, dust, sea-salt) of those components. The aircraft performed 6 research flights from Fairbanks, Alaska, all of them over northern Alaska and the Arctic sea ice.

[7]   The aircraft carried an extensive suite of instruments for measuring gas and aerosol components and solar and IR radiation. The instruments that are used most extensively in this work are: a vacuum ultraviolet fluorescence measurement for carbon monoxide (CO), a PTR-MS instrument for VOCs (volatile organic compounds) [*de Gouw and Warneke*, 2007], a glass flask sampling system with 12 flasks per flight for hydrocarbons and halocarbons [*Montzka et al.*, 2005], a compact time-of-flight Aerodyne aerosol mass spectrometer (AMS) for size-resolved particle composition [*Drewnick et al.*, 2005], an SP-2 for BC mass [*Schwarz et al.*, 2006] and a PALMS instrument for size-resolved single-particle composition [*Murphy et al.*, 2006].

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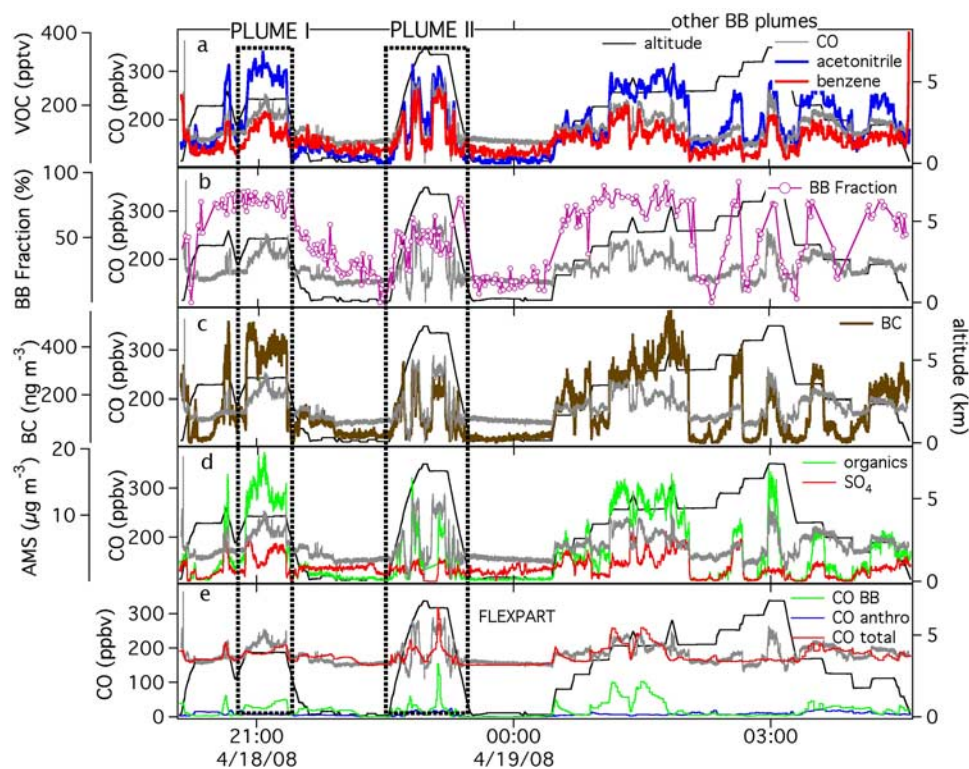
Figure 1. Time series of various species during the NOAA WP-3 flight on April 18, 2008 over the Alaskan Arctic. Fire plumes were observed multiple times at various altitudes. Two plumes (out of 9) are marked and their sources are discussed in Figure 2.

[8] The Lagrangian particle dispersion model FLEXPART was used to characterize the transport of pollution into the Alaskan Arctic region from surface emissions [*Stohl et al.*, 2005]. From each location along the flight tracks, 20,000 particles were released in the model and tracked for 15 days backward in time. The model output consists of a response function, which is proportional to the residence time of the particles in a given volume. When convolved with the gridded emission fluxes from an emission inventory and integrated over the volume of the atmosphere, a model-calculated mixing ratio of the emitted species at the aircraft is obtained. In the present study, EDGAR [*Olivier and Berdowski*, 2001] was used as the anthropogenic emissions inventory and for BB an inventory as described by *Stohl et al.* [2007] that uses fire locations detected by the moderate-resolution imaging spectrometer MODIS and the land cover classification.

3. Observations of Pollution Plumes in the Arctic

[9] The time series of various gas and aerosol species from a flight on April 18, 2008 north of Fairbanks, Alaska are shown in Figure 1. The area where the ARCPAC flights took place is shown by the rectangle in Figure 2a. Plumes are identified as strong enhancements of CO (above typical background levels of about 160 ppbv) and of most other tracers. Plumes were observed during large parts of the flights at various altitudes. Some plumes were targeted, but others encountered on route to different objectives. Figure 1a shows acetonitrile and benzene measured by the PTR-MS together

with CO. Acetonitrile is a good indicator for BB emissions and benzene is emitted in large quantities from BB, as well as from anthropogenic sources [*de Gouw et al.*, 2003]. Both tracers follow CO very well in all plumes observed during ARCPAC indicating a BB source for the observed plumes. Figure 1b shows the fraction of particles measured by PALMS (Dp > 200 nm) that are classified as BB particles [*Hudson et al.*, 2004]. Inside the plumes most particles were classified as BB particles. Figure 1c shows black carbon measured by the SP-2. In all observed plumes strong enhancements of BC were found correlated with CO. Figure 1d shows the organics and sulfate aerosol mass as measured by the AMS; organics in particular are enhanced in the plumes, although sulfate, nitrate, and ammonium are also often enhanced. Figure 1e shows the result of the FLEXPART model for a transport time of 15 days. FLEXPART CO from recent anthropogenic and BB emissions is shown together with total modeled CO, including a 160 ppbv background typical of the Arctic spring. In agreement with the measurements, FLEXPART indicates that BB emissions are the dominant source of the CO enhancements for this flight. In general FLEXPART reproduced the measured CO quantitatively and qualitatively quite well. For this flight a linear fit of measurements versus model forced through zero yielded a slope of 0.97 with a correlation coefficient of R = 0.63.

[10] The FLEXPART-derived BB CO source contribution (SC) maps for plumes I and II from Figure 1 are shown in Figure 2. The maps show the location and the amount of BB emissions that resulted in CO concentrations (ppbv) at
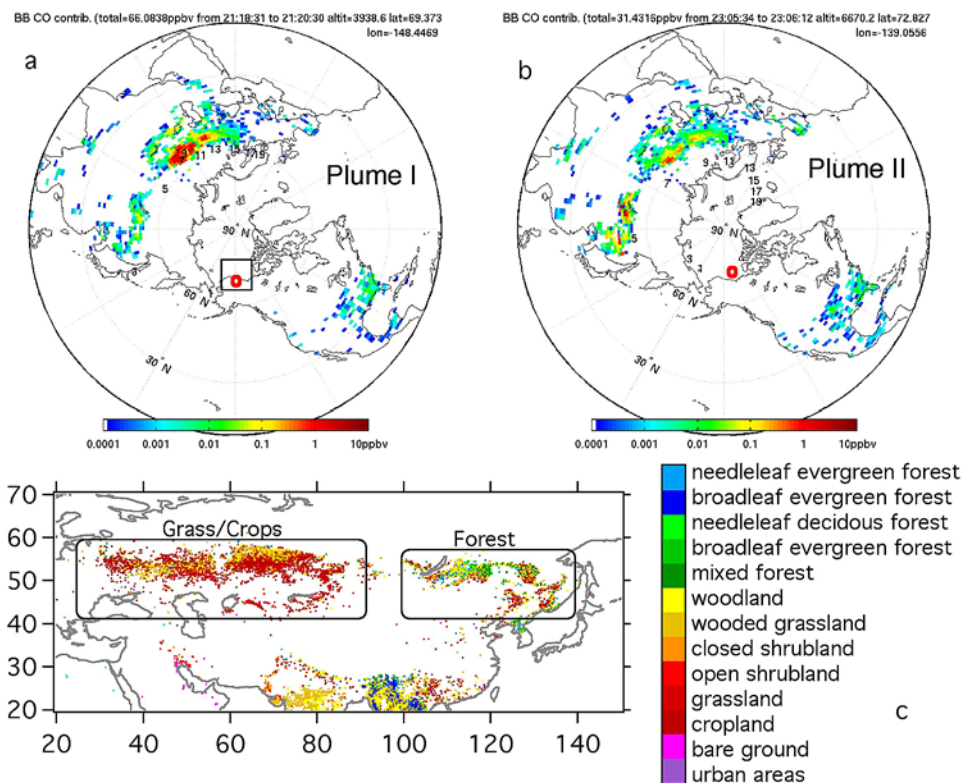
Figure 2. (a) and (b) FLEXPART BB CO source contribution for plume I and II in Figure 1 on a logarithmic scale. The square in Figure 2a indicates the area where the ARCPAC flights took place and all the BB plumes were observed. (c) MODIS fire detection between April 1, 2008 and April 20, 2008. The color code indicates the dominant land cover classes.

the location of the aircraft (marked with a small red circle in Figure 2). Summing up all the grid boxes yields the total amount of BB CO predicted at the aircraft location. FLEXPART attributes plume I mainly to fires burning in northern Kazakhstan and southern Russia, and plume II mainly to fires burning between Lake Baikal and the Amur River area in southern Siberia. Figure 2c shows the MODIS fire locations during the first half of April color-coded by the land cover classification. The fires burning in Kazakhstan and southern Russia were agricultural fires, likely started by farmers clearing the fields for spring planting, whereas the fires burning between Lake Baikal and the Amur River area in southern Siberia were mostly boreal forest fires.

[11] Almost all observed plumes over the Alaskan Arctic in April 2008 were identified as BB plumes. However, the chemical composition differed from plume to plume. As an example, plumes I and II in Figure 1 are both identified as BB, but the gas and aerosol phase composition were markedly different. The differences in chemical composition are evident in scatter plots versus CO of acetonitrile, benzene, BC and aerosol organics (Figure 3a–3d). For all species in Figure 3 except benzene, the enhancement ratios (slope versus CO) were larger in the agricultural fire plumes than in the forest fire plumes. High emission ratios are expected from residual smoldering combustion during pasture maintenance fires as has been reported from Amazonian agricultural fires [*Christian et al.*, 2007]. Only small enhancements of propane and C_2Cl_4 were observed in the BB plumes

(Figure 3), indicating a nearly negligible contribution from anthropogenic pollution to the plumes [*de Gouw et al.*, 2004].

[12] Table 1 summarizes the enhancement ratios of various gas phase species from all the fire plumes observed over the Alaskan Arctic during ARCPAC. Based on FLEXPART, 10 plumes were attributed predominantly to the Kazakhstan agricultural fires and 39 to the Lake Baikal forest fires. The enhancement ratios for CO_2, BC and VOCs were calculated for each plume from the slope of an orthogonal distance regression to the data in and just around the plume. For most compounds in Table 1, the enhancement ratios are larger in the agricultural fire than in the forest fire plumes. Enhancement ratios in forest fire plumes from Alaska and Canada observed in 2004 are also shown in Table 1 [*de Gouw et al.*, 2006; *Warneke et al.*, 2006] and are similar to the ones from the Lake Baikal fires indicating similar fire types. The 2004 Alaskan fires show no enhancements for propane and the pentanes. The agricultural fires and not the forest fires might have therefore caused the enhancements observed here. Note that enhancement ratios will change with plume age and for shorter-lived compounds the measured ratios are much smaller than the emission ratios at the source. According to FLEXPART, the transport time for the Lake Baikal fires was about 4–5 days and for the Kazakhstan fires 7–9 days.

[13] Light absorbing aerosols from boreal forest fires in Siberia may be deposited on the Arctic sea ice and snow cover thereby reducing surface albedo [*Generoso et al.*, 2007]. Therefore, the presence of fire plumes in the
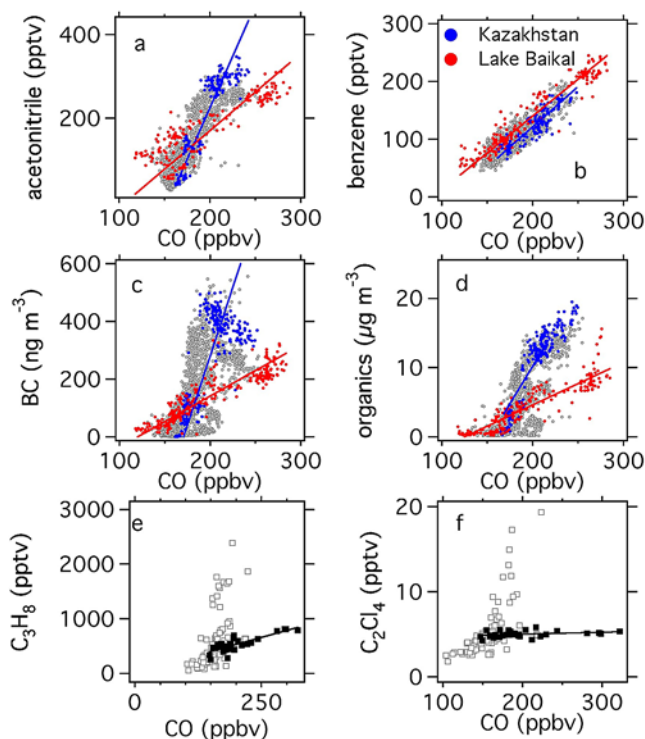
Figure 3. (a)–(d) Scatter plots with CO for the NOAA WP-3 flight on April 18, 2008 (gray points). Plumes, marked in Figure 1, from agricultural fires in Kazakhstan (plume I) are in blue and from forest fires in the Lake Baikal area (plume II) are in red. (e)–(f) C_3H_8 and C_2Cl_4 versus CO for multiple flights in the Alaskan Arctic. Flasks sampled in the fire plumes are in black all others in gray. Multiple flights are shown for Figures 3e and 3f because of the small number of flask samples (12) taken on any individual flight.

boundary layer over the sea ice and snow cover is of particular importance, since BC may be deposited to the surface due to dry and wet deposition. Figure 4a shows the altitude profile of acetonitrile and CO for a flight profile off the coast over the sea ice close to Barrow, Alaska. The location of the profile is marked with a red circle on the FLEXPART BB CO source contribution plot (Figure 4b). Even at the lowest flight altitude of <40 m, elevated concentrations of all measured gas- and aerosol phase species were observed. The measured enhancement ratios were similar to the ones from the Lake Baikal forest fires, and the FLEXPART source contribution plot (Figure 4b) also indicates the same source. The exchange between the stable surface layer over the sea ice to the free troposphere is generally very slow. However, if this BB plume stays within the near-surface layer over sea ice for sufficient time for dry and wet deposition processes, it would contribute significant BC amounts to the snow and ice surface. It has been speculated that blowing and drifting snow might cause increased wet deposition to the surface, which might accelerate the interaction between the surface and the atmosphere [*Stohl et al.*, 2007].

4. Conclusions and Implications

[14] During the ARCPAC campaign a large number of dense BB plumes, originating from agricultural fires in Kazakhstan and southern Russia and from forest fires in the Lake Baikal-Amur River area, were encountered over the Alaskan Arctic. During six flights in northern Alaska and the adjacent sea ice, more than 50 BB plumes were encountered. Both the measurements and the particle dispersion model FLEXPART indicate that these BB emissions were significant in controlling the gas-phase and aerosol characteristics in the Alaskan Arctic as early as April in 2008; the

Table 1. Enhancement Ratios (Slope With CO): Averages From 10 Different Fire Plumes From Mainly Agricultural Burning in Kazakhstan and 39 Different Fire Plumes From Mainly Forest Fires in the Lake Baikal Area Observed Over the Alaskan Arctic[a]

	Agricultural Fires Kazakhstan (pptv ppbv^{-1})	Forest Fire Lake Baikal (pptv ppbv^{-1})	Alaskan and Canadian Forest[b] (pptv ppbv^{-1})
CO/CO_2	50 ± 25	42 ± 19	
methanol	31 ± 11	20 ± 8	11 ± 5
acetonitrile	3.1 ± 1	2.1 ± 0.8	2.4 ± 0.7
acetone	11.7 ± 4.5	7.7 ± 3.6	6 ± 5
acetic acid	12.0 ± 5.2	9.6 ± 3.9	6 ± 3
MEK	1.7 ± 0.6	1.1 ± 0.5	0.7 ± 0.5
benzene	1.3 ± 0.3	1.2 ± 0.3	1.1 ± 0.2
toluene	0.15 ± 0.06	0.2 ± 0.1	0.2 ± 0.1
C8-aromatics	0.14 ± 0.03	0.08 ± 0.08	0.12 ± 0.1
C9-aromatics	0.23 ± 0.08	0.13 ± 0.10	0.09 ± 0.02
C_2H_2[c]		4.02 (0.97)	4.2 (0.93)
C_3H_8[c]		3.61 (0.83)	0.8 (0.95)
i-pentane[c]		0.13 (0.72)	-0.02 (-0.50)
n-pentane[c]		0.17 (0.84)	-0.01 (-0.02)
C_2Cl_4[c]		0.002 (0.31)	0.004 (0.66)
$CHCl_3$[b]		0.003 (0.21)	0.001 (0.10)
BC[d]	10 ± 5	7 ± 4	N/A

[a]Values are given in pptv/ppbv together with their standard deviations. Enhancement ratios from forest fires in Alaska and Canada observed in 2004 over the New England area are also included.

[b]*de Gouw et al.* [2006] and *Warneke et al.* [2006].

[c]All flask samples collected inside all fire plumes observed during ARCPAC flights were used to determine the enhancement ratios. The correlation coefficient is shown in the brackets.

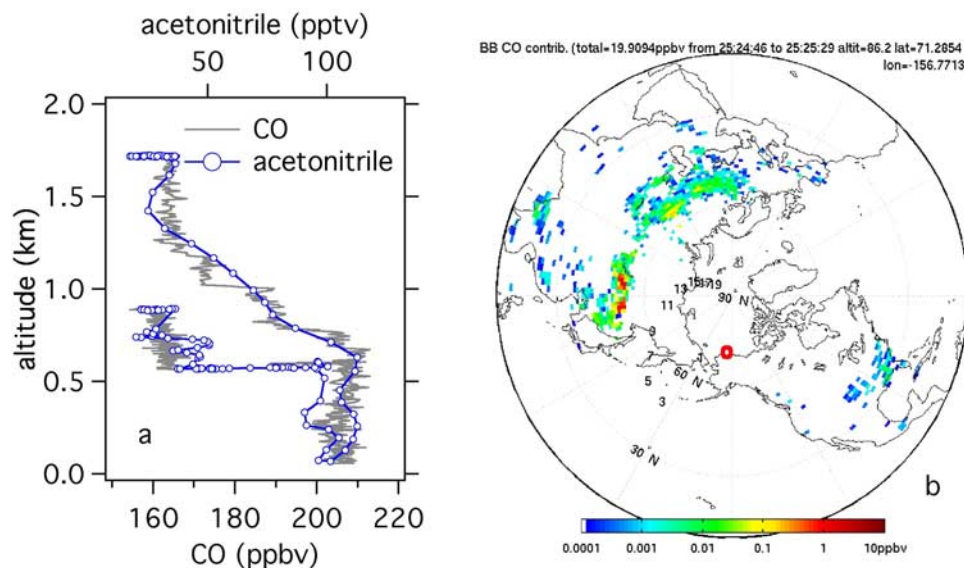[d]Units are in ng m^{-3} ppbv^{-1} at 1013 hPa and 273K.

Figure 4. (a) Altitude profile for CO and acetonitrile near Barrow, Alaska on April 19, 2008, showing significant BB influence in the surface layer. (b) FLEXPART BB CO source contribution for the lowest point during the altitude profile, which is shown with the red circle.

Lake Baikal-Amur River fires were the dominant contributor. The chemical measurements clearly showed that the sources of both plumes are BB, but that the chemical composition was very different; the agricultural fires had larger enhancement ratios for most measured species in comparison with the forest fires. The BB plumes were found at altitudes from the surface layer to the highest flight levels of about 6.5 km. The presence of the BB smoke in the surface layer over Arctic sea ice and snow-covered land suggests the possibility of relatively efficient deposition of light absorbing aerosols onto the snow and ice and potential reduction in surface albedo.

[15] Forest fires in Siberia and agricultural burning in Kazakhstan, southern Russia and Eastern Europe are a seasonal occurrence and usually start at the end of April. The agricultural burning usually lasts for a few weeks [*Stohl et al.*, 2007], whereas the forest fires last all summer [*Generoso et al.*, 2007]. In 2008 the fire season started unusually early because of the low snow amount in Siberia and Russia that year (see http://rapidfire.sci.gsfc.nasa.gov/firemaps/). Early in spring, when the temperatures are still cold, quasi-isentropic transport of cold air from high latitude Asian source regions is a possible pathway into the lower troposphere of the Arctic [*Stohl*, 2006]. This means that the earlier the fire season in Siberia starts, the easier it is to transport the emissions into the Arctic. Long-term light absorption measurements from Barrow, Alaska [*Quinn et al.*, 2007] decrease through the 90s and are constant since 2000, but show an increase during March 2003, a year that had an early and intense fire season.

[16] High latitude warming might cause a possible increasing trend in area burnt each year and could extend and intensify the burning season [*Kasischke et al.*, 2005]. This may give rise to a feedback loop [*Stohl*, 2006]: higher forest fire emissions, especially early in the fire season, cause a more efficient transport and deposition of BC in the Arctic and enhance the snow and ice melting thereby causing a stronger climate forcing at high latitudes.

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