An important contribution to springtime Arctic aerosol from biomass burning in Russia

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[1] Using aircraft observations and transport model calculations we determine the total amounts of various gas-phase and aerosol species in the Arctic due to distant biomass burning (BB) emissions. We find that for many climate-relevant species, including black carbon (BC) and organic aerosols, fires in Russia that typically occur during the critical springtime snowmelt can more than double the high seasonal Arctic atmospheric background that has built up in the winter months (commonly called "Arctic haze"). Decision makers have targeted BC, because it is expected to cause strong positive forcing over snow-covered surfaces yet is significantly shorter lived than greenhouse gases. These results demonstrate that BB is more important for the Arctic than previously believed and should be considered in any attempt to mitigate impacts. **Citation:** Warneke, C., et al. (2010), An important contribution to springtime Arctic aerosol from biomass burning in Russia, *Geophys. Res. Lett.*, *37*, L01801, doi:10.1029/2009GL041816.

1. Introduction

[2] Concentrations of aerosol particles and trace gases are enhanced within the Arctic troposphere in winter and spring due to transport of anthropogenic emissions and slow removal processes leading to long lifetimes for many species [*Quinn et al.*, 2007]. Anthropogenic emissions from northern Eurasia are identified by modeling, surface, airborne and remote sensing measurements as the principal source for this seasonal atmospheric background [*Law and Stohl*, 2007], which is commonly called "Arctic haze".

[3] Arctic surface temperature has increased by about 1.5°C over the past three decades, warming much faster than the global average [*Intergovernmental Panel on Climate Change*, 2001]. *Shindell and Faluvegi* [2009] attributed as much as 1.1°C of this increase to changes in regional aerosol composition, surpassing warming from greenhouse gases. They found that improvements in air quality in Europe and the United States have caused a decrease in sulfate and black carbon (BC) aerosol while anthropogenic emissions in Asia have increased BC in the Arctic.

[4] Several studies indicate that during the summer fire season BB can also be a significant source for aerosols and trace gases in the Arctic [*Koch and Hansen*, 2005; *Stohl et al.*, 2006]. Only recently it was found that BB was responsible for a large fraction of deposited BC in Arctic snow samples [*Hegg et al.*, 2009]. This finding is significant to climate forcing because light-absorbing BC deposited on snow surfaces accelerates snowmelt [*Flanner et al.*, 2007].

[5] Agricultural burning and wildfires occur seasonally in southern Russia after the local snowmelt each spring [*Soja et al.*, 2004] and are usually started by human influence. In 2008 the fire season started unusually early and was rather intense. Due to efficient northward transport in spring [*Stohl*, 2006], BB plumes from these regions were carried to the Arctic, where gas-phase and aerosol species were characterized in detail using aircraft measurements conducted during the International Polar Year (IPY) [*Warneke et al.*, 2009].

[6] In this paper we show, using a combination of measurements and model calculations, that BB was a particularly important source for aerosols in the Arctic during springtime of 2008, and may be significant springtime source in more typical years. The springtime is the most critical time for aerosol induced Arctic warming [*Flanner et al.*, 2009].

2. Methods

[7] During IPY, airborne field experiments, the Aerosol, Radiation, and Cloud Processes affecting Arctic Climate (ARCPAC) study and the NASA Arctic Research of the Composition of the Troposphere from Aircraft and Satellites (ARCTAS) mission [*Jacob et al.*, 2009], were conducted in Alaska in April 2008 (study area is shown in Figure 3a). We use data from the NOAA WP-3D and NASA B-200 aircraft, which carried extensive suites of instruments for measuring gas, aerosol and radiation properties [*Jacob et al.*, 2009; *Warneke et al.*, 2009]. The instruments that are used most extensively in this work are: a vacuum ultraviolet fluorescence measurement for carbon monoxide (CO), a Proton Transfer Reaction-Mass Spectrometer (PTR-MS) for VOCs (volatile organic compounds), a flask sampling system for hydrocarbons and halocarbons, multiple condensation and optical particle counters for the aerosol volume distribution, a compact time-of-flight Aerodyne Aerosol Mass Spectrometer (AMS) for size-resolved particle composition, a Single-Particle Soot Photometer (SP-2) for BC mass and a Particle Analysis by Laser Mass Spectrometry (PALMS) instrument for size-resolved single-particle composition, and the High

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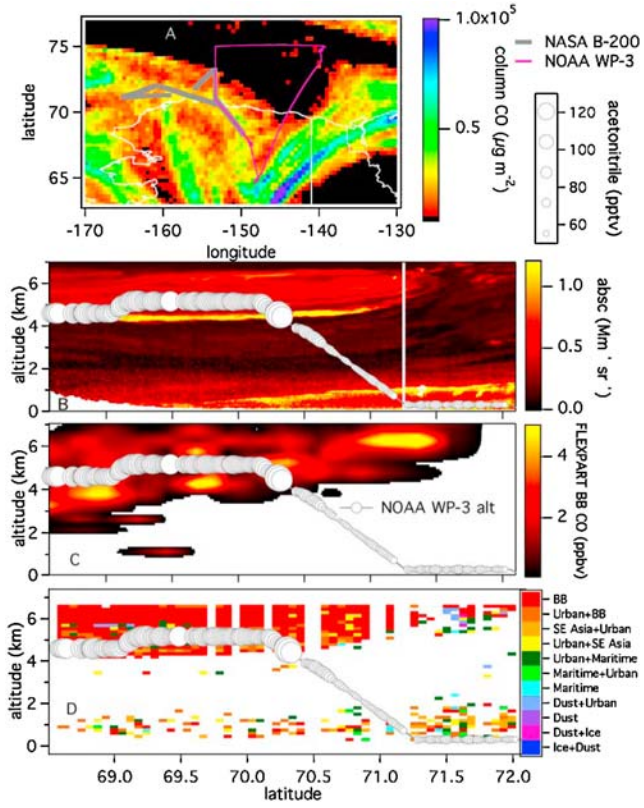

Figure 1. (a) FLEXPART total column BB CO on April, 12 2008 in the Alaskan Arctic with the flight tracks of the NOAA WP-3D and the NASA B-200. (b) NASA B-200 lidar aerosol backscatter vertical curtain along the NOAA WP-3 flight track. The altitude of the NOAA WP-3 aircraft is size coded with the measured acetonitrile mixing ratio. (c) FLEXPART BB CO vertical curtain along the NOAA WP-3 flight track. (d) NASA B-200 LIDAR aerosol classification along the NOAA WP-3 flight track.

Spectral Resolution Lidar (HSRL), which measured aerosol backscatter and extinction profiles at 532 nm and aerosol backscatter profiles at 1064 nm.

[8] The Lagrangian particle dispersion model FLEXPART was used to characterize the transport of pollution from surface emissions into the Alaskan Arctic region [*Warneke et al.*, 2009]. From each location along the flight tracks, 20,000 air parcels ("particles") were released in the model and tracked for 20 days backward in time. EDGAR was used as the anthropogenic emissions inventory [*Olivier and Berdowski*, 2001]. For BB, the inventory was based on fire locations detected by the moderate-resolution imaging spectrometer MODIS and a land-cover classification [*Stohl et al.*, 2007]. The model output is anthropogenic and BB CO mixing ratios including only emissions within the previous 20 days.

[9] Total CO emissions from the Russian fires in April 2008 were 17 Tg, which is very similar to a-posteriori values calculated from the GEOS-Chem model (15 Tg) using ARCPAC measurements [*Fisher et al.*, 2009].

[10] *Warneke et al.* [2009] demonstrated that many gas and aerosol species were highly correlated with CO originating from Russian BB. We also showed that the FLEXPART model effectively simulated the sources, transport, and enhancements in BB CO during ARCPAC in 2008. Here we use FLEXPART to estimate the CO enhancements throughout the Arctic from 2003–2008. We use this modeled BB CO enhancement and the measured ratio with CO to estimate the BB enhancements of a number of gas and aerosol species.

3. Results

[11] On April 12, 2008, the NOAA WP-3D and NASA B-200 aircraft operated over Alaska and the Arctic sea ice in an area where the FLEXPART transport model predicted large BB CO enhancements from fires in the Chita/Amur region east of Lake Baikal [*Warneke et al.*, 2009] (Figure 1a). The HSRL observed a layer of scattering aerosol at 4–6 km (Figure 1b) in good agreement with FLEXPART (Figure 1c). The layer was attributed to mostly BB sources based on the backscattering, extinction, and depolarization data from the lidar using a cluster analysis procedure grouping the data into major categories (Figure 1d). Measurements of acetonitrile, a tracer of biomass combustion [*de Gouw et al.*, 2003], and other species onboard the NOAA WP-3D, such as low gas-phase alkanes and organic-rich aerosols containing potassium, unambiguously attributed the layer at 4–6 km to BB emissions. This is one example of the more than 50 BB plumes encountered during April 12–23, 2008 by the NOAA WP-3D. The CO enhancements calculated by FLEXPART in the observed fire plumes usually agreed within a factor of two with the measurements, and within 30% averaged over the entire study (using 160 ppbv CO atmospheric background) [*Warneke et al.*, 2009].

[12] A more detailed look at the aerosol classification over the springtime Alaskan Arctic is given in Figure 2, which shows aerosol volume distributions together with particle classification distributions from single-particle mass spectrometry measurements [*Froyd et al.*, 2009]. Aerosol particles are classified on a particle-by-particle basis into broad aerosol types using a variety of compositional markers in PALMS mass spectra. Aerosol particles identified as biomass burning are either directly emitted from fires or result from coagulation or condensation of direct emissions with non-fire particles. To generate the composition-resolved volume distributions in Figure 2, the number fraction of each aerosol type in a given size bin is multiplied by the total aerosol volume for that size bin. In the mid-latitude and tropical background troposphere, 20–30% of accumulation mode particles show biomass burning influence [*Froyd et al.*, 2009]. During ARCPAC, particles containing BB material dominated the distribution inside (Figure 2a) and frequently outside (Figure 2b) intense fire plumes. Only in the boundary layer most particles consistently had no BB component (Figure 2c).

[13] Figures 1 and 2 summarize some of the evidence for the strong influence of BB on the Alaskan Arctic in April 2008. How representative are these results for other years and regions of the Arctic? We can use FLEXPART to determine the total amount of BB and anthropogenic CO enhancements within the ARCPAC region (Figure 3c) and within the Arctic as a whole (Figure 3d), defined as north of 70° (Figure 3a). This was done for March, April and May of 2003–2008. Using the measurement model comparison, we

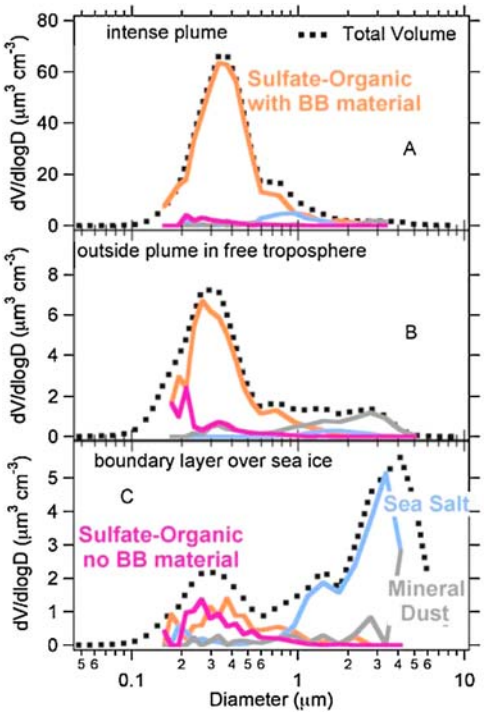

Figure 2. Composition-resolved aerosol volume distribution for three representative types of air encountered in the Alaskan Arctic. (a) Inside an intense BB plume (April 20, 2008 0:03–0:19 between 1.1–3 km altitude). (b) Typical free troposphere values measured on April 15, 2008. CO values for this flight segment were between 160–170 ppbv. (c) Aged boundary layer air that showed minimal influence of recent BB emissions.

estimate about 30% error on the calculated BB CO in the Arctic.

[14] During two intense fire years in Russia (2003 and 2008 [*Huang et al.*, 2009; *Warneke et al.*, 2009]), the BB CO enhancements dominated over the anthropogenic CO enhancements. The transport of the Siberian fire emissions to the Arctic usually goes through the Alaskan ARCPAC region [*Stohl*, 2006] causing the larger BB CO enhancements relative to anthropogenic CO in this area. Figures 3c and 3d show that about half of the BB CO emissions transported into the Arctic were located in the Alaskan Arctic during April 2008.

[15] The total amount of BB CO is the only model result used in the following analysis, everything else will be based on measurements.

[16] Aircraft measurements of different gas-phase and aerosol species were used to determine enhancement ratios relative to CO ($\Delta X/\Delta CO$) within BB plumes [*Warneke et al.*, 2009]. Using these measured enhancement ratios and the BB CO calculated according to FLEXPART, we calculated the total amount of all the measured species from BB present in the Arctic and the ARCPAC region. This method relies on the accurate modeling of one species, CO, in fire plumes and utilizes in-situ measurements of enhancements relative to CO in the Arctic for all other species of interest. Most models do not reproduce Arctic aerosols well due to difficulties in the accurate representation of physical and chemical processing (including removal), where especially BC is clearly underestimated in the Arctic [*Koch et al.*, 2009]. The analysis used here assumes that the observed $\Delta X/\Delta CO$ values are the same every year, which adds to the uncertainty in years other than 2008.

[17] Gas-phase and aerosol species in the BB plumes are superimposed upon background concentrations. Atmospheric background values of all species of interest are determined from the frequency distributions of the observations. The NOAA WP-3 aircraft was flying most of the time outside plumes in background conditions and therefore the concentrations that were observed most often are here defined as the background values. As an example, we determined background values of 160 ppbv CO, 90 pptv benzene, 1.65 μg m^{-3} aerosol mass, 29 ng m^{-3} BC, 0.9 μg m^{-3} aerosol sulfate and 0.42 μg m^{-3} aerosol organics, which are consistent with long-term surface-based measurements [*Quinn et al.*, 2007; *Shindell et al.*, 2008]. Here we do not determine the sources causing the background, but instead compare the background to the large perturbations from BB.

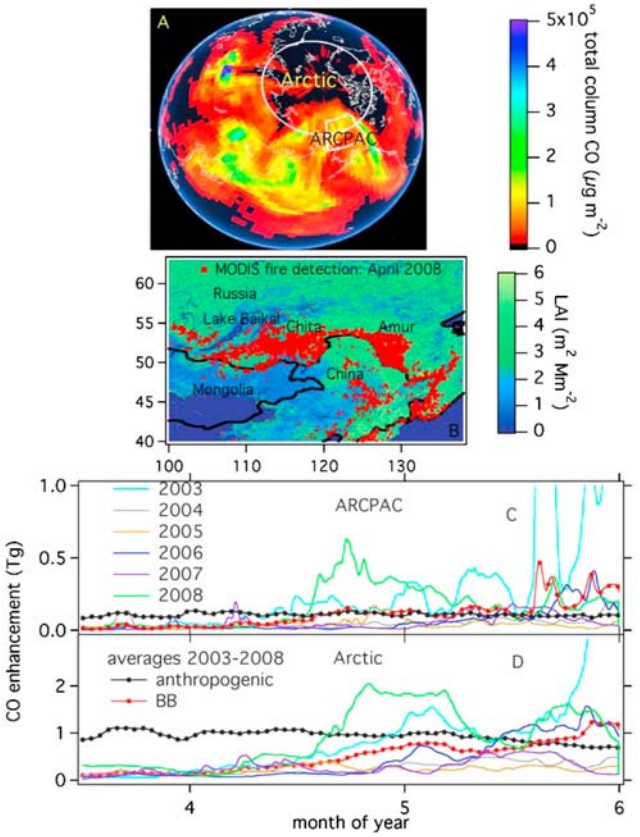

Figure 3. (a) Total column BB CO enhancement from FLEXPART for April 19, 2008. The ARCPAC and Arctic regions are outlined. (b) MODIS fire detections for April 2008 in the Chita/Amur region on top of the leaf area index (LAI) map as an indication for the amount of biomass and land use. (c) Time series of total BB CO enhancement from FLEXPART inside the ARCPAC area for March, April and May between 2003–2008. Also shown are the 2003 to 2008 average anthropogenic and BB CO enhancement from FLEXPART. (d) Same as Figure 3c, but for the entire Arctic (north of 70°). FLEXPART CO enhancements in Figure 3 are from emissions within the previous 20 days.

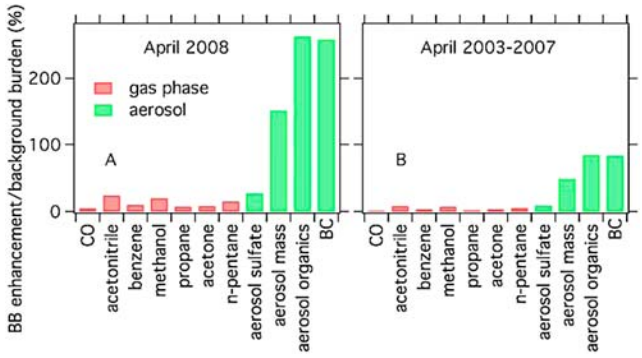

Figure 4. The average trace gas and aerosol enhancements due to BB from emissions within the previous 20 days compared with the determined atmospheric background burden in the Arctic (north of 70°). (a) April 2008 average. (b) April 2003–2007 average.

[18] Below we determine the contribution from the BB plumes relative to the background burden. Using the volume of the Arctic atmosphere and measured background values the total Arctic burden of all the measured compounds can be determined. For example, the April background CO of 160 ppbv is equivalent to about 18 Tg of CO in the Arctic troposphere. According to FLEXPART, fire plumes added on average about 0.8 Tg CO or 4% to this measured background in April 2008 (Figure 3c). Ratios of the BB enhancement, calculated using measured enhancement ratios and modeled CO, relative to the Arctic tropospheric background burden, determined from our measurements, were small for all measured gas-phase species in April (Figure 4). This finding is consistent with model results that attribute most of the CO measured during ARCPAC and ARCTAS to Asian and, to smaller extent, European anthropogenic emissions and not to BB [*Fisher et al.*, 2009]. The gas-phase species most influenced by recent BB emissions was the BB marker acetonitrile (about 12% of the background burden), which has negligible anthropogenic/industrial emissions and a surface sink [*de Gouw et al.*, 2003].

[19] In contrast with the gas-phase species, the Arctic burdens of all aerosol species were strongly impacted by recent BB emissions (Figure 4a). Aerosol BC and organic matter from BB were larger by 260% than the Arctic background burden. The mass of submicron particles from BB was 150% and aerosol sulfate 30% of the background burden. Why are the enhancements due to BB larger for aerosol than for gas-phase species? First, the main sinks for aerosols, wet and dry deposition, although slow in the winter Arctic troposphere, are faster than the photochemical sink for many gas-phase species [*Shindell et al.*, 2008]. Second, fires are copious producers of aerosol mass compared with anthropogenic sources, such as industrial and vehicular emissions. As a consequence of these differences in sources and sinks, the background aerosol concentrations do not build up over the winter as much as the gas-phase species, so that the overall impact of the episodic springtime BB plume events is substantially larger for aerosol species.

[20] Long-term surface measurements indicate that background aerosol consists mainly of sulfate and to a lesser extent of organics, ammonium, nitrate and BC [*Law and Stohl*, 2007; *Quinn et al.*, 2007]. BB plume aerosols are rich in BC and organic carbon and contain relatively little sulfate [*Warneke et al.*, 2009]. This explains why BB had a larger impact on BC and organic carbon in the Arctic than on aerosol sulfate (Figure 4a).

[21] Figure 4b shows that for the April averages from 2003-2007 the influence of BB was smaller than in 2008. BC and organic aerosol from BB was about 80% of the total atmospheric background burden. The year 2008 was anomalous with about twice as many fires in April in Russia as the average [*Fisher et al.*, 2009], but it is also clear that fires in this region perturb the Arctic atmosphere annually.

[22] The BB influence determined here is likely a lower limit: (1) FLEXPART traces particles for 20 days only and all the BB CO that remains in the Arctic longer is not accounted for. (2) A small part of the background may also have come from older fires such as European biofuel burning co-emitted with anthropogenic pollution during the winter. On the other hand, anthropogenic emissions within the previous 20 days will increase the total atmospheric burden.

4. Implications

[23] Our results, based on in-situ measurements in the Arctic and a transport model of CO, suggest that BB plumes transported to the Arctic in spring in 2008 more than double the Arctic atmospheric background burden, which is believed to be of mainly anthropogenic origin. BC is especially important for the springtime Arctic: in the free troposphere, even with mass loadings observed here, BC likely has a only small warming effect at the surface [*Flanner et al.*, 2009], but small amounts deposited on the snow-covered surface from BB, as observed by *Hegg et al.* [2009], could cause strong warming and snowmelt [*Flanner et al.*, 2009]. Determining the relative importance of both processes and determining the fraction of BC deposited on the surface requires further study. Due to a lack of long-term data and strong inter-annual variability, it is not clear if a trend in fire emissions in Russia exists and it is unclear how much of the above average Arctic warming can be attributed to BB emissions. In a future warmer climate a feedback loop is possible. Snowmelt and therefore the fire season in boreal forest regions might start earlier and possibly be more severe so that fire years such as 2003 and 2008 become more common [*Stocks et al.*, 1998].

[24] Strong gradients in MODIS fire counts across political borders despite the apparent lack of differences in land use, as in the Chinese/Russian border region in 2008 (Figure 3b) and previous years [*Soja et al.*, 2004], suggest that fire suppression during the springtime snowmelt could have a very substantial effect on Arctic climate.

[25] As a historical note, one may conclude that our results also indicate that some of the haze layers originally observed by aircraft and termed "Arctic haze" [*Mitchell*, 1957] may have been biomass burning layers. Arctic haze was chemically characterized based mainly on surface observations [e.g., *Quinn et al.*, 2007]) but this might often not be representative of the haze layers aloft.

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