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Impacts of enhanced biomass burning in the boreal forests in 1998 on tropospheric chemistry and the sensitivity of model results to the injection height of emissions

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[1] Carbon monoxide reached record high levels in the northern extratropics in the late summer and fall of 1998 as a result of anomalously large boreal fires in eastern Russia and North America. We investigated the effects of these fires on CO and tropospheric oxidants using a global chemical transport model (GEOS-Chem) and two independently derived inventories for the fire emissions that differ by a factor of two. We find that it is essential to use both surface and column observations of CO to constrain the magnitude of the fire emissions and their injection altitude. Our results show that the larger of the two inventories appears more reliable and that about half of the emissions were injected above the boundary layer. The boreal fire emissions cause a much larger enhancement in ozone when about half the emissions are released above the boundary layer than when they are released exclusively in the boundary layer, as a consequence of the role of PAN as a source of NO_x as air descends in regions far from the fires.

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1. Motivation and Introduction

[2] Surface measurements of CO in the late summer of 1998 were the highest on record for that season at northern mid and high latitudes since 1988, when widespread measurements began [*Novelli et al.*, 2003]. Anomalously high CO columns were also observed [*Yurganov et al.*, 2004]. The elevated CO has been attributed to emissions from enormous boreal fires in Siberia and North America, possibly linked to El Nino conditions in late 1997/1998 [*van der Werf et al.*, 2004; *Yurganov et al.*, 2004].

[3] The magnitude of emissions from boreal fires is relatively low compared to those from tropical fires [*Lobert et al.*, 1999]. The record is punctuated however by years of intense fire activity, during which emissions from boreal forests may be enhanced by as much as an order of magnitude [*Amiro et al.*, 2001a, 2001b; *Stocks et al.*, 2002; *Duncan et al.*, 2003b; *French et al.*, 2002]. Boreal fires have been posited to be the cause of the interannual variability of CO in the northern extratropics [*Wotawa et al.*, 2001; *Novelli et al.*, 2003; *Yurganov et al.*, 2004; *Kasischke et al.*, 2005] and to contribute to the variability in CH_4 and CO_2 [*van der Werf et al.*, 2004].

[4] Emissions from boreal fires have affected U.S. air quality in recent years. Smoke from large wildfires in Quebec in July 2002 was transported rapidly down the east coast of the United States as far as Virginia, reducing visibility and dramatically affecting air quality, with CO enhancements exceeding 500 ppb [*Colarco et al.*, 2004; *DeBell et al.*, 2004]. Ozone is formed in plumes from forest fires, but boreal fires have relatively low emissions of nitrogen oxides (NO_x) [e.g., *Wofsy et al.*, 1992]. *Wotawa and Trainer* [2000] found that transport of emissions from large fires in western Canada caused increases in surface CO of up to 200 ppb, and in ozone of 10–20 ppb over the southeast United States. A model analysis of effects of these fires showed that ozone was enhanced by 10–30 ppb over much of the central and eastern United States; about half the increase was associated with emissions from the fires, and the remainder with ozone production from anthropogenic sources of NO_x downwind from the fires [*McKeen et al.*, 2002].

[5] There were major boreal fires in Russia in 2002 and 2003. Observations of CO by the MOPITT (Measurement of Pollution in the Troposphere) instrument showed hemispheric-scale pollution from these fires [*Edwards et al.*, 2004], as did the surface and ground-based column observations [*Yurganov et al.*, 2005]. The Russian fires influenced air quality in the northwest United States. For example, surface ozone was 5–9 ppb higher in May in 2003 than in years without major

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fires [*Jaffe et al.*, 2004]; model simulations suggested that the fires could have increased ozone by 2–6 ppb.

[6] In this study we investigate the effects of the boreal fires in 1998 on tropospheric composition using a three-dimensional (3-D) chemical tracer model, GEOS-Chem. Our principal focus is on CO, but we also investigate the effects of enhanced emissions on ozone and OH. There is considerable uncertainty in estimates of emissions from the boreal fires of 1998, and as part of this work we assess the reliability of two independent emission inventories for the fires in Russia.

[7] We use two gridded inventories for boreal fire emissions, developed independently by *Kasischke et al.* [2005] and *Kajii et al.* [2002] (referred to as KAS05 and KAJ02 respectively), and evaluate model results using CO measurements from surface sites and ground-based column measurements [*Novelli et al.*, 2003; *Yurganov et al.*, 2004]. The two inventories rely on burned areas derived from satellite observations of hot spots (thermal anomalies), but their estimates of CO emissions from Siberian fires differ by a factor of two (105 Tg CO for KAS05, 43 Tg for KAJ02) because of different assumptions about fuel consumption and emission factors. A third inventory, also derived from satellite data, gives an estimate of 74 Tg for CO emissions from these fires, with an estimate of 119 Tg for an ''extreme'' scenario [*Soja et al.*, 2004a]; this inventory is not available as a monthly gridded product. Several studies provide assessments of CO emissions from fires in the entire boreal region in 1998: 131 Tg for the moderate scenario of KAS05, with 69 Tg and 163 Tg for their low-severity and high-severity scenarios; 69 Tg, derived by scaling a climatological inventory with satellite-derived aerosol data [*Duncan et al.*, 2003b]; 148 Tg derived from the enhancement in column measurements of CO and a simple box model [*Yurganov et al.*, 2004]; and 44 Tg (north of 38°N) derived from a simple inversion analysis that assumed that interannual variability in CO was caused only by variability in emissions from fires [*van der Werf et al.*, 2004]. Using the CASA model in conjunction with satellite data, *van der Werf et al.* [2006] estimate that boreal fires burned about 70% more carbon in 1998 than the moderate scenario of KAS05, mainly because they derived larger areas burned.

[8] There is evidence that emissions from boreal forests are injected above the boundary layer, as major fires can produce enough energy to create a convective column. Plumes from boreal fires were observed between about 2 and 7 km during aircraft campaigns over Alaska and central Canada [e.g., *Blake et al.*, 1992, 1994; *Wofsy et al.*, 1992; *Shipham et al.*, 1992, 1994]. *Lavoué et al.* [2000] showed that the injection height of emissions is related to the energy released along the flame front of the fires; injection heights were 2.5 and 5 km for two experimental boreal fires, and 13 km for a wildfire in Canada. In extreme cases, the convection associated with boreal fires reaches into the lower stratosphere, but such events are relatively infrequent [*Fromm et al.*, 2000, 2005; *Fromm and Servranckx*, 2003; *Livesey et al.*, 2004; M. Fromm, personal communication, 2005].

[9] Measurements of aerosols by the Multiangle Imaging SpectroRadiometer (MISR) on board the Terra spacecraft were used to determine plume heights for fires in Alaska and northwest Canada in 2004 [*Mazzoni et al.*, 2007]. The median height of the young plumes was 2.2 km, with a range of 0.8–5.2 km, at 1030 local time. The injection height of emissions from boreal fires has also been deduced from model studies. Simulations of long-range transport of emissions from boreal fires from Russia [*Bertschi et al.*, 2004] and Canada [*Colarco et al.*, 2004] imply that the emissions must have been injected between 3 km and 6 km. In this study, we investigate the sensitivity of our results to the injection height of the emissions from boreal fires.

[10] The GEOS-Chem model is described in section 2, where we also discuss the boreal fire inventories, the simulation of carbonaceous aerosols, and the scenarios used for injection heights of the fire emissions. The CO observations are described in section 3, and model results are evaluated with observations in section 4. The effects of the fires on ozone and OH are shown in section 5, and our conclusions are discussed in section 6.

2. Model Description

[11] The GEOS-Chem model was originally described by *Bey et al.* [2001]. The model is driven by assimilated meteorological observations from the Global Modeling and Assimilation Office (GMAO) from the Goddard Space Flight Center. In this study, we use GEOS-3 meteorological fields resolved over 48 vertical sigma levels. The meteorological information is provided with six hour resolution (three hourly for surface variables and mixing depths) on a $1^\circ \times 1^\circ$ grid, but we use resolution of 4° (latitude) $\times$ 5° (longitude) for computational expediency. The model time step is 30 minutes. We use version 5.05.03 of the GEOS-Chem model (http://www.as.harvard.edu/chemistry/trop/geos/). Emissions from fossil fuels and industry are those for 1997 [*Bey et al.*, 2001].

[12] The model is run with 31 transported tracers, including CO and the nitrogen oxide and hydrocarbon species needed to simulate the major cycles important to tropospheric oxidant chemistry. Updates to the model are described by *Martin et al.* [2003]. Gas phase chemical reaction rates and photolysis cross sections are taken from *Sander et al.* [2000]. The newer photolysis cross sections of ozone and of reaction rates involving $O^1(D)$ result in a lowering of tropospheric OH from earlier model versions which used the 1997 kinetic data [*Fiore et al.*, 2005]. Photolytic reaction rates are calculated using the Fast-J algorithm, which takes into account scattering by aerosols [*Wild et al.*, 2000]. In addition, sulfate and ammonium aerosols are modeled explicitly [*Park et al.*, 2004]. Carbonaceous aerosols are simulated in a separate experiment and are used offline, as described below.

[13] Stratosphere to troposphere transport of ozone is parameterized using the Synoz method developed by *McLinden et al.* [2000], and implemented as described by *Bey et al.* [2001] with a stratospheric source of 475 Tg O_3 y^{-1}. The cross-tropopause transport of NO_y is treated in a similar manner.

[14] We used results from a simulation for 2001, a normal year meteorologically and one without anomalous biomass burning emissions, as initial conditions; concentration fields for the end of December were used for 1 January 1998. The GMAO changed from the GEOS-STRAT assimilation system

Table 1. Emission Factors[a]

	Forest			Grassland	
KAJ02	Crown and Superficial On-Ground[b]	Steady On-Ground[b]	Peat	Superficial On-Ground[b]	Steady On-Ground[b]
CO	81	163	189	72	146
NO	2.0	3.8	4.4	1.8	3.8
KAS05	Flaming	Smoldering			
CO	97	230			
Duncan et al. [2003b] in GEOS-Chem	All Fire Types				
CO	120				
NO	0.44				

[a]Unit is g/kg dry mass burned.
[b]Categories defined by KAJ02.

to GEOS-3 at the end of 1997, causing a discontinuity in the meteorological fields, so we preferred to use GEOS-3 data for initialization. Consequently we did not investigate the effects of the anomalous tropical fires in late 1997 on CO in the first part of 1998. The model simulates the general seasonal and latitudinal distribution of surface CO, but consistently underestimates the amplitude of the seasonal cycle at northern high latitudes [*Heald et al.*, 2003].

2.1. Emissions From Biomass Burning in GEOS-Chem

[15] The preexisting global inventory for biomass burning in the GEOS-Chem model is summarized by *Lobert et al.* [1999] and [*Duncan et al.*, 2003b]. In that inventory, emissions for Canada were based on provincial statistics for area burned for 1980–1990 provided by the Canadian Forest Service, and on biomass fuel for different types of forest provided by B. Stocks. Since reliable statistics for forest fires in the former Soviet Union were not available when the inventory was developed, the area burned was calculated by assuming that the same mean fraction of the forests burned as in Canada, 0.25% yr^{-1} in the 1980s. The fire emissions were distributed over the forests in each country/province using the vegetation map of *Matthews* [1983].

[16] The GEOS-Chem model can be run with either mean emissions or year specific emissions. For the mean emissions, the seasonal variation in each grid box is based on an analysis of satellite hot spot data; for year-specific emissions, TOMS aerosol index data are used to scale the emissions in various regions [*Duncan et al.*, 2003b]. After 1997, when ATSR hot spot data are available, they are used to locate the fires within Canada and Russia, in addition to the interannual scaling of the base emissions. For the baseline simulation in this study we used the mean seasonal biomass burning emissions.

2.2. Emissions From Boreal Forest Fires in 1998

[17] Emissions from biomass burning are typically quantified using estimates of area burned, amounts of fuel consumed per unit area, and emission factors. Different assumptions about these quantities contribute to significant differences in the emission inventories for the same region, as described below. For 1998 we use the inventories of KAJ02 and KAS05. KAJ02 provide estimates only for Russian boreal fires (40–69°N and 79–153°E) from April to October, so we adopt emissions specific to 1998 from *Duncan et al.* [2003b] for other months and for all other regions. E. Hyer and E. Kasischke (personal communication, 2004) provided their inventory for all boreal fires; it is very similar to the moderate scenario they published in KAS05, and is referred to as such. Both inventories were provided with monthly resolution on a $1° \times 1°$ grid and regridded to $4° \times 5°$ for model simulations.

2.2.1. Area Burned and Timing of Fires

[18] KAJ02 used AVHRR fire count data and applied a three-threshold algorithm to AVHRR images to determine the location of hot spots with resolution of 1 km by 1 km. KAS05 used estimates for fire sizes and locations in Russia that were determined using a combination of AVHRR hot spots and burn scar data [*Soja et al.*, 2004a, 2004b; *Sukhinin et al.*, 2004]. The location and areas of fires in Canada were taken from a large fire database up to 2000 and from provincial data thereafter [*Stocks et al.*, 2002], and estimates for Alaskan boreal fires were made using a database kept by the Alaskan Fire Service and data from the National Interagency Coordination Center [*Kasischke and Brulwiler*, 2003; *Kasischke et al.*, 2005]. The timing of both Alaskan and Canadian boreal fires was determined from AVHRR hot spots [*Kasischke et al.*, 2005]. The area burned in Russia in both studies differ by about 20%: 8.8×10^{10} m^2 for KAJ02 and 10.8×10^{10} m^2 for KAS05.

2.2.2. Fuel Consumption

[19] One of the major difficulties in making reliable estimates for emissions from boreal fires is the uncertainty in the amount and type of fuel that is burned, as discussed by KAS05. Key uncertainties include the ratio of crown fires to surface fires and the amount of duff that is burned. Duff is the organic material covering the forest floor. As reviewed in KAS05, it varies in depth from 4 cm in well-drained forests to over 50 cm in boreal forest underlain with permafrost, and the amount of carbon burned can vary from 2 to >10 t C ha^{-1}. Burning of duff can therefore be as important as burning of aboveground biomass, fuel consumption for which vary from <10 t C ha^{-1} to >20 t C ha^{-1}. Emissions from duff burning may be especially high during intense fires, when the deeper, more carbon rich, duff layers are burned. Compared to KAJ02, KAS05 assumes both more widespread crown fires, which consume a much higher fraction of the available carbon than do surface fires [*French et al.*, 2002], and higher consumption of duff.

KAS05 assumes that 90% of the fires in Russia were crown fires in August, on the basis of satellite observations, while KAJ02 assumes that only 19–25% of fires in taiga were crown fires. KAS05 assume that the depth of burning of the duff was 50% greater in crown fires than in surface fires, while KAJ02 do not discuss their assumptions with regard to the importance of below ground fuels. The net result is that for August 1998, less than half the carbon burned was from crown fires in KAJ02, compared to over 90% in KAS05. In addition to these uncertainties, it has been suggested that emissions from the burning of duff may persist for weeks after hot spots are evident in satellite images (E. Kasischke, personal communication, 2005).

2.2.3. Emission Factors

[20] Table 1 shows the emissions factors used by KAJ02 and KAS05 as well as those adopted in the standard version of GEOS-Chem. KAS05 and *Duncan et al.* [2003b] relied on the work of *Cofer et al.* [1998], who reported the results from 2 controlled burn experiments (in the North West Territories (NWT) of Canada and the Bor Island Experiment in Siberia) as well as typical results from fires associated with logging. The mean emission factor (EF) for CO from smoldering fires for the Bor Island study was larger than that from the NWT by nearly a factor of 2. For crown fires, the differences were only 20%. KAS05 used an average of the 3 studies of *Cofer et al.* [1998] while *Duncan et al.* [2003b] relied on the high values from Siberia. Emission factors for KAJ02 were constructed using the lowest EFs from literature sources for crown and ''superficial on-ground fires,'' and the highest EFs for peat fires, and interpolated values for intermediate fire types.

[21] KAJ02 provided emissions of NO derived with the EFs given in Table 1. For consistency, we applied the same EFs to the inventory of carbon burned provided by E. Hyer and E. Kasischke. There have been very few measurements of EFs for NO from boreal fires. *Wofsy et al.* [1992] reported molar enhancements ratios, $\Delta NO/\Delta CO$, of 0.0056 in plumes measured 200–1200 km downwind of Alaskan fires, while *McKeen et al.* [2002] report a ratio of 0.007 for $\Delta NO_y/\Delta CO$ measured 2 days downwind from fires. The latter argue that their reported value represents a lower limit for the emission ratio, because of removal of NO_y. Measurements in the vicinity of three different Alaskan fires give higher enhancement ratios, 0.012 for $\Delta NO_x/\Delta CO$ [*Nance et al.*, 1993], and 0.014 and 0.018 for $\Delta NO/\Delta CO$ for two different fires [*Goode et al.*, 2000]. The data from these studies are more likely to be representative of emission ratios, as there has been less time for loss of NO_x. The average of the measurements in the vicinity of the fires corresponds to an emission factor of 1.4 g NO kg^{-1} dry matter (DM), 30% lower than the value adopted by KAJ02 for boreal forest fires, and significantly higher than the value of 0.44 g NO kg^{-1} DM that was used in previous versions of GEOS-Chem [*Staudt et al.*, 2003], based on data given by *Wofsy et al.* [1992].

2.2.4. Comparison of Estimates for CO Emissions in 1998

[22] The spatial distribution of emissions from KAJ02 and KAS05 is shown in Figure 1 and the seasonality of emissions in Siberia is shown in Figure 2. The timing of the emissions from both studies is similar, with a maximum in August. The CO emissions for the Siberian fires given by KAS05 (104.6 Tg y^{-1}) are nearly a factor of 2 greater than those from KAJ02 (42.6 Tg y^{-1}) for the same region (79–153°E and north of 40°N) because of higher fuel consumption, although the estimated area burned differs only by about 20%. Emissions from Canadian fires given by KAS05 (19.6 Tg CO y^{-1}) are similar to those in the GEOS-Chem inventory for 1998 (25.6 Tg CO y^{-1}) The average CO emissions from all boreal fires used in the baseline simulation are 14 Tg y^{-1} [*Duncan et al.*, 2003b], about an order of magnitude lower than those of KAS05. KAJ02 derived emissions of 50 Tg CO and a 1.2 Tg NO for Russia, which corresponds to an overall molar emission ratio of 0.022 for NO:CO. Application of the emission factors used by KAJ02 to the inventory for carbon burned of KAS05 gives emissions of 3.54 Tg NO, which corresponds to an overall molar emission ratio of 0.032 for NO:CO. The difference in the mean ratio for NO:CO stems from differences in the assumptions about fuel consumption throughout the burning season.

2.3. Modeling of Carbonaceous Aerosols

[23] In this study organic carbon (OC) and black carbon (BC) aerosols were calculated in an offline GEOS-Chem simulation. Carbonaceous aerosols affect tropospheric photochemistry, and therefore aerosol emissions consistent with gaseous emissions are necessary in model simulations. For simulations with the KAS05 emissions, the OC and BC aerosol emissions were calculated using the gridded inventory of carbon burned from KAS05. For the simulations using the KAJ02 emissions, the underlying carbon inventory was not available, so OC and BC aerosol emissions from fires were calculated using the mean biomass burning emissions inventory in GEOS-Chem [*Duncan et al.*, 2003b]. To calculate aerosol emissions for the rest of the world, we used the 1998 specific emissions from *Duncan et al.* [2003b]. For the baseline simulations, the mean emissions of *Duncan et al.* [2003b] were used everywhere. Emission factors were specified as 2 g EC/kg DM and 14 g OC/kg DM following *Park et al.* [2004]. The global anthropogenic emissions of black carbon followed the inventory of *Bond et al.* [2004].

2.4. Modeling of Injection Heights

[24] There is evidence from satellite aerosol data that convection can carry biomass burning emissions plumes above the planetary boundary layer, and on occasion, above the tropopause, as discussed in the Introduction. Satellite instruments recorded higher than average aerosol concentrations 3 to 5 km above the tropopause at high latitudes between May and October 1998, concurrent with hot spots, suggesting that there were widespread and strong convective events produced by boreal forest fires [*Fromm et al.*, 2000]. On the basis of AVHRR images and radiosonde data, *Fromm and Servranckx* [2003] showed an instance in which strong convection carried smoke from an intense boreal forest fire through the tropopause. However, the global impact of explosive convection on the transport and fate of atmospheric trace gases is not well quantified, and the first systematic study of plumes from boreal fires (in Alaska) showed heights that were usually less than 5 km [*Mazzoni et al.*, 2007],

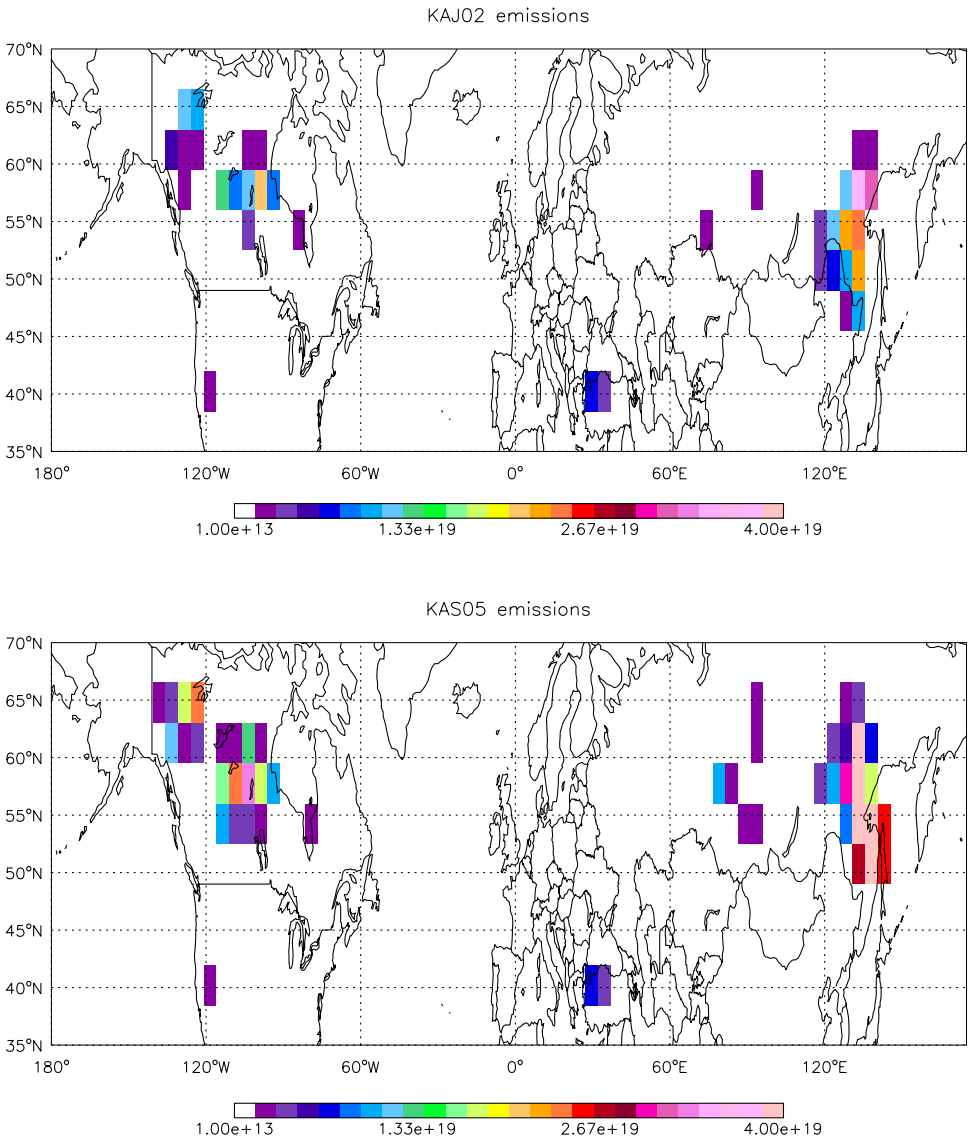

Figure 1. Biomass burning emissions for August 1998 (the peak month) for the two data sets used in this study, (top) from *Kajii et al.* [2002] for Siberia and (bottom) from *Kasischke et al.* [2005] for all boreal regions. Values are in molecules/cm^2.

although these were for young plumes measured at mid morning.

[25] In the standard version of GEOS-Chem, biomass-burning emissions are distributed throughout the boundary layer. More realistically, emissions should be injected into the atmosphere at various heights depending on a variety of factors including type of fire, fuel consumption, and local meteorological conditions. Detailed models of case studies are currently under development (M. Fromm, personal communication, 2005). We chose here to perform sensitivity studies to examine the effect of the injection height of fire emissions on atmospheric composition.

[26] We performed three different simulations to probe the sensitivity of model results to the injection heights of the emissions (Table 2), using emissions from KAS05. Emissions from crown fires have been shown to reach an altitude of ~5 km [*Cofer et al.*, 1998]. We assume in scenario KAS05.D1 that emissions are injected uniformly throughout the troposphere; in scenario KAS05.D2, 40% of the gas phase emissions enter the boundary layer initially, while 60% of emissions are emitted between 3 and 5 km. The fractions in KAS05.D2 were chosen by assuming that the majority of fires in late summer are crown fires, that a large fraction of these emissions are injected above the boundary layer, and that emissions from surface fuels are emitted in the boundary layer only.

2.5. Model Runs

[27] We performed simulations using boreal fire emissions specific to 1998 from KAJ02 and from KAS05, and a baseline simulation using mean biomass burning emissions, with the emissions injected into the boundary layer only (KAJ02.BL, KAS05.BL, Baseline). All simulations were performed using meteorological data from 1998. The model simulations were started on 1 January 1998, and our focus is on the results from May onward, when there were large

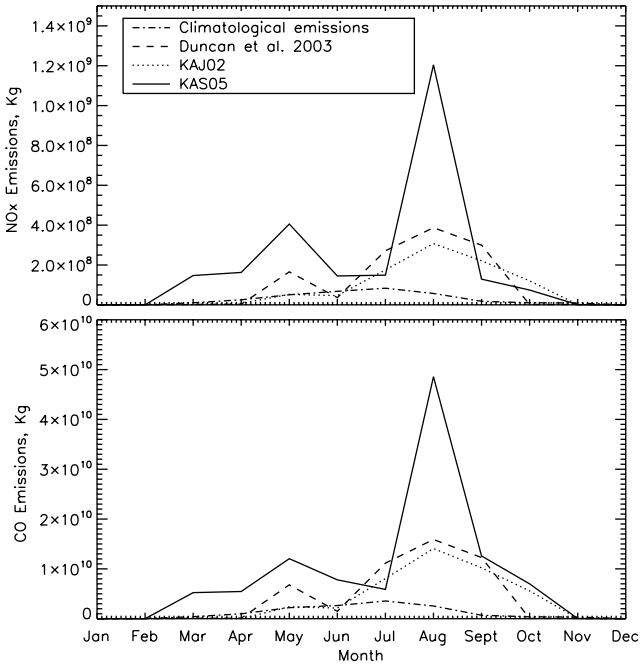

Figure 2. Monthly mean emissions of from boreal fires in Siberia, for (top) NOx and (bottom) CO. Estimates are shown for 1998 for KAS05 (solid), KAJ02 (dotted), *Duncan et al.* [2003b] (dashed) and mean fire emissions (dot-dashed).

emissions from the boreal fires (Figure 2). Two simulations were conducted with emissions injected above the boundary layer, KAS05.D1 and KAS05.D2 as described above. Table 2 summarizes the details of the simulations.

3. Observations

[28] Carbon monoxide is measured weekly at a network of surface stations operated by the NOAA Earth System Research Laboratory, Global Monitoring Division (GMD) (formerly known as the Climate Monitoring and Diagnostics Laboratory, CMDL) [*Novelli et al.*, 2003]. Flask samples are collected in pairs, and as part of the GMD data analysis, flask pairs are flagged as nonbackground if they deviate from a smooth curve fit to the data by more than 3 sigma. We include these data points, as long as they are duplicated, because we do not screen for nonbackground air in our model. Monthly means are calculated as the average of the weekly means. The error bars are calculated as the standard deviation of all values for the month. The mean values for 1992–1997, used for evaluating the Baseline simulation, are the means of the individual monthly means for each station. Some stations started in 1993 and 1994, and for these the data prior to 1998 are used for evaluation of the baseline simulations.

[29] Column measurements for CO, made using infrared spectroscopy, were taken from *Yurganov et al.* [2004]. Monthly mean values were provided for 1998 and for the mean of 1996–2001, excluding 1998. We applied averaging kernels to model results in order to compare them properly to the column observations. In this study, averaging kernels provided by C. Rinsland (personal communication, 2005) were used for all locations except for Zvenigorod, for which we used an averaging kernel provided by L. Yurganov (personal communication, 2003). For sites above sea level, the model results for the sigma layers above the altitude of the station were used.

4. Evaluation of Model Simulations With Surface and Column Observations

4.1. Boreal Fire Emissions Released in the Boundary Layer

[30] Model results were evaluated with CO measurements for 20 GMD sites from 28°N to 82°N (Tables 3 and 4). The 8 sites shown in Figure 3 were selected to illustrate the effects of emissions from the Russian fires at sites closest to the most intense fires (the near-field sites, Barrow, Shemya Island, and Cold Bay in the Pacific sector) and those far removed from Russia (the far-field sites, Alert, Spitsbergen, Ocean Station M, Iceland, and Mace Head, in the Atlantic sector). The observations show that there are pronounced enhancements in CO in the late summer of 1998 compared to the mean for 1992–1997, with peak concentrations between September and November. The enhancements are about 100 ppbv (~90% of baseline values) in September at the high-latitude near-field sites, and 50–80 ppb at the far-field sites.

[31] Figure 3 shows that the simulation using the KAJ02 emissions for 1998 released in the boundary layer (KAJ02.BL) matches the observations for 1998 much better than does the corresponding simulation using the KAS05 emissions; the latter overestimates CO from August onward, particularly at the near-field sites. The baseline simulation tends to overestimate CO for 1992–1997 in late summer at the time of the seasonal minimum, a feature seen in an earlier study using GEOS-Chem [*Heald et al.*, 2003]. To compensate for this bias, we compare the observed anomaly in CO in 1998 (the difference between CO in 1998 and in 1992–1997) to the model anomaly (the difference between the simulations with 1998 emissions and the baseline simulation) in Figure 4.

[32] The late summer anomaly in surface CO in 1998 is captured by both cases KAJ02.BL and KAS05.BL, but the latter greatly overestimates the anomaly at the near-field sites. Furthermore, the peak anomaly occurs a month early at the sites closest to the fires, Shemya and Cold Bay. The two simulations generally bracket the observed anomaly at

Table 2. GEOS-Chem Simulations

Simulation Name	Biomass Burning Emissions	Aerosol Emissions From Biomass Burning	Vertical Distribution of Emissions
Baseline	climatological	climatological	100% BL[a]
KAJ02.BL	1998 from KAJ02	climatological	100% BL
KAS05.BL	1998 from KAS05	1998 from KAS05	100% BL
KAS05.D1	1998 from KAS05	1998 emissions from KAS05	entire troposphere
KAS05.D2	1998 from KAS05	1998 emissions from KAS05	60% at 3–5 km, 40% in BL

[a]Boundary layer.

Table 3a. GMD Surface Stations

Station	Latitude	Longitude	Altitude, m
Alert	82°N	62°W	210
Spitsbergen (Ny-Alesund)	78°N	11°E	475
Barrow, AK	71°N	156°W	11
Ocean Station M	66°N	2°E	5
Iceland	63°N	20°W	100
Cold Bay, AK	55°N	162°W	25
Mace Head	53°N	9°W	42
Shemya Island	52°N	174°E	40
Hegyhatsal	46°N	16°E	116
Park Falls, WI	45°N	90°W	868
Black Sea	44°N	28°E	3
Niwot Ridge, CO	40°N	105°W	3013
Wendover, UT	39°N	113°W	1320
Terceira Island	38°N	27°W	40
Qinghai	36°N	100°E	3810
Gozo Is., Malta	36°N	14°E	30
Grifton, NC	35°N	77°W	505
St. David, Bermuda	32°N	64°W	8
Canary Islands	28°N	16°W	2360
Midway Island	28°N	177°W	8

the far-field sites, with the KAS05.BL case generally too high and the KAJ02.BL case slightly low. The mean bias for the CO anomaly is 22 ppb for KAS05.BL and −4 ppb for KAJ02.BL in August to October (Table 4). If we considered the surface sites alone, with emissions released exclusively in the boundary layer, we would conclude that the inventory of KAS05 significantly overestimates the emissions from the boreal fires in 1998.

[33] Evaluation of model simulations for 1998 with ground-based column measurements of CO leads to the opposite conclusion to that drawn above. Both the column abundance of CO in 1998 and the anomaly in the column are better predicted in simulation KAS05.BL than in simulation KAJ02.BL (Figures 5 and 6 and Table 4). As with the surface data, pronounced enhancements in the CO column are observed in the late summer of 1998, with maximum enhancements occurring in August (Hokkaido, Japan) and September–October (European stations). The simulation KAJ02.BL consistently underestimates the anomaly in the CO column, and the maximum anomaly occurs about a month later than in the data. Simulation KAS05.BL reproduces the anomalies in 1998 better than does KAJ02.BL, in terms of both magnitude and timing, with much smaller biases for August to October (Table 4). One notable exception is Hokkaido, the site closest to the fires, where the maximum column abundance in KAS05.BL occurs a month late and the seasonal evolution of the model anomaly is different from that observed.

[34] The anomaly in the CO column in 1998 is much smaller at the high-altitude European stations, Jungfraujoch (3.6 km, 47°N) and nearby Zugspitze (3.0 km) than at other sites close to sea level, and it is present all year. The anomaly is smaller relative to the baseline in early 1998 (∼15%) than in September–October (20–27%). *Yurganov et al.* [2004] suggested that the anomaly in early 1998 over central Europe was caused by emissions from huge tropical fires in late 1997, but a model simulation using GEOS-Chem of the effects of these fires gives an anomaly of only 1×10^{17} molecules/cm^2 in December 1997 [*Duncan et al.*, 2003a], smaller than the observed anomaly in January 1998 by about a factor of two (Figure 6). There is also a large anomaly in the CO column in May 1998 at Zvenigorod whose cause is unknown; its magnitude relative to the baseline is ∼20%, much smaller than that in October, ∼40%.

[35] The baseline simulation underestimates the amplitude of the spring maximum for the column data from 1996 to 2001 (excluding 1998). It is also consistently lower than the observed CO column throughout the year at Jungfraujoch and Hokkaido, a discrepancy that is not observed in the surface measurements at similar latitudes.

[36] There is good evidence that emissions from boreal fires are injected above the boundary layer, as discussed in the Introduction. We show next that the contradictory results provided by evaluation of the two emissions scenarios using the surface and column data may be resolved by allowing a significant fraction of emissions to be injected above the boundary later.

4.2. Effect of Vertical Distribution of Emissions on Surface and Column CO Levels

[37] We show the sensitivity of surface CO in 1998 to the injection altitude of the boreal fire emissions using the KAS05 inventory in Figure 7. We find that CO at the near-field sites is especially sensitive to the injection height of the emissions, and that model CO decreases as a smaller fraction of the emissions is injected in the boundary layer (100% for KAS05.BL, 40% for KAS05.D2, and ∼25% for KAS05.D1). Simulation KAS05.D2, with 40% of the emissions in the boundary layer and 60% between ∼3 and 5 km, performs best with the smallest bias in the CO anomaly for near-field and far-field sites, as well as for the four GMD sites in the United States, and for the ensemble of extratropical sites (Table 4).

[38] The anomaly in column CO is smaller when a large fraction of the emissions are released above the boundary layer, with the exception of results for Hokkaido, which is relatively close to the location of the fires in eastern Russia (Figure 8). Simulations KAS05.D1 and KAS05.D2 both give a large anomaly in August at Hokkaido, similar in magnitude to that observed, and mimic the smaller anomalies in September–December, including the shoulder in October. We find that emissions from the fires in eastern Siberia that are injected above the boundary layer are transported southward toward Japan, causing the more pronounced and earlier maximum in August. Our results are consistent with those of both *Zhao et al.* [2002] and *Tanimoto et al.* [2000] who observed enhanced CO levels in Northern Japan in the summer of 1998 and concluded that these episodes were caused by the influx of air carrying enhanced biomass burning emissions from Siberia. The bias in the mean column anomaly for August–October is smaller for simulation KAS05.D2 than for KAS05.D1, and is similar to that for KAS05.BL, but the latter does not

Table 3b. Column Observation Stations

Station	Latitude	Longitude	Altitude, m
Spitsbergen	78.92°N	11.94°E	20
Kiruna	67.84°N	20.41°E	419
Zvenigorod	55.70°N	36.80°E	200
Zugspitze	47.42°N	10.98°E	2964
Jungfraujoch	46.55°N	8.00°E	3580
Hokkaido	44.00°N	143.10°E	280
Kitt Peak	31.90°N	68.60°W	2090

Table 4. Mean Bias of the CO Anomaly for August-October 1998[a]

GMD Sites	KAJ02.BL	KAS05.BL	KAS05.D1	KAS05.D2
20 sites, 82–28°N	−5.1	42.2	−20.0	3.9
NF	−6.3	75.4	−23.0	9.1
FF	−4.4	22.4	−18.2	0.7
U.S. sites	−5.8	−7.3	−10.8	−1.2
Column	-2.7×10^{17}	-0.7×10^{17}	-2.3×10^{17}	-0.8×10^{17}

[a]Unit is ppb for the GMD sites and molecules cm^{-2} for the column stations. The table gives the difference (model anomaly − observed anomaly), averaged for the 3 months, August–October. NF, near-field sites (Barrow, Shemya Island, Cold Bay); FF, far-field sites (Alert, Spitsbergen, Ocean Station, Iceland, Mace Head (Ireland)). Continental U.S. sites: Wendover, Park Falls, Niwot Ridge, and Grifton. Column data are over all seven sites.

reproduce seasonality of CO anomaly at Hokkaido, and fails to capture the August maximum.

[39] On the basis of the results of the simulations shown here, we conclude that the KAS05 inventory with 40% of the emissions released in the boundary layer and 60% at 3–5 km best reproduces the CO anomalies seen in the surface and column observations for 1998. The surface data alone would not allow us to constrain both the magnitude of the emissions and the injection altitude. The column data are essential to show that the KAJ02 inventory underestimates significantly the observed anomaly in CO. Clearly, there is considerable uncertainty in the typical fraction of boreal fire emissions injected above the boundary layer, and it is likely to be highly variable, as discussed in section 6 below.

[40] The pervasive effect of the boreal fire emissions on surface CO in the northern extratropics in September 1998 is shown in Figure 9. The enhancement in CO exceeds 200 ppb near the fires in eastern Siberia, and is about 100 ppb downwind of these fires in western Alaska. It is 50–75 ppb over much of eastern Canada, ~40 ppb over the northeast United States, and 20–40 ppb over the northern United States, with similar enhancements over much of

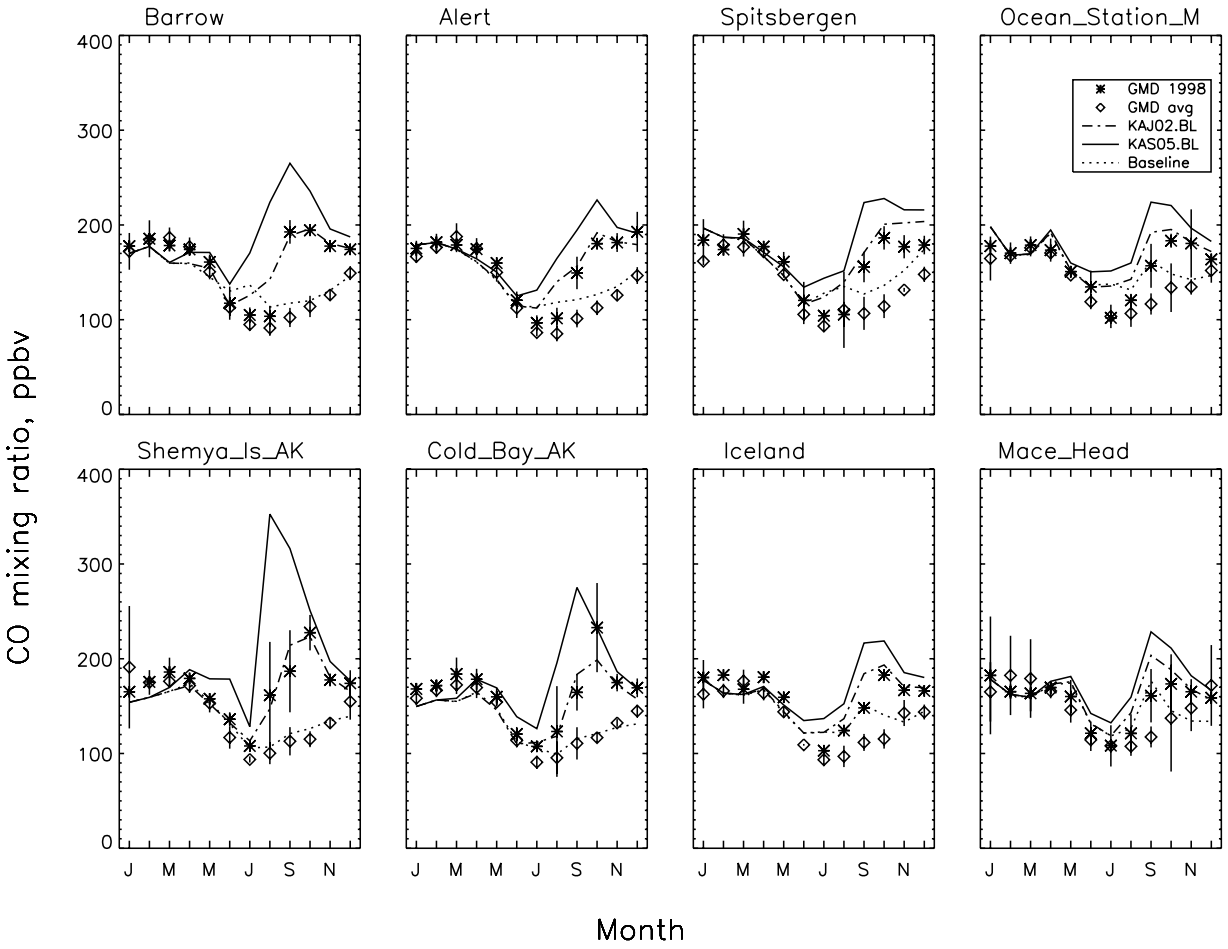

Figure 3. Comparison of observed and simulated CO mixing ratios with boreal fire emissions released in the boundary layer only. GMD data for 1998 (asterisks) are compared to simulations for 1998 for cases KAS05.BL (solid) and KAJ02.BL (dot-dashed). GMD data for 1992–1997 (diamonds) are compared to the baseline simulation with mean fire emissions (short dashes).

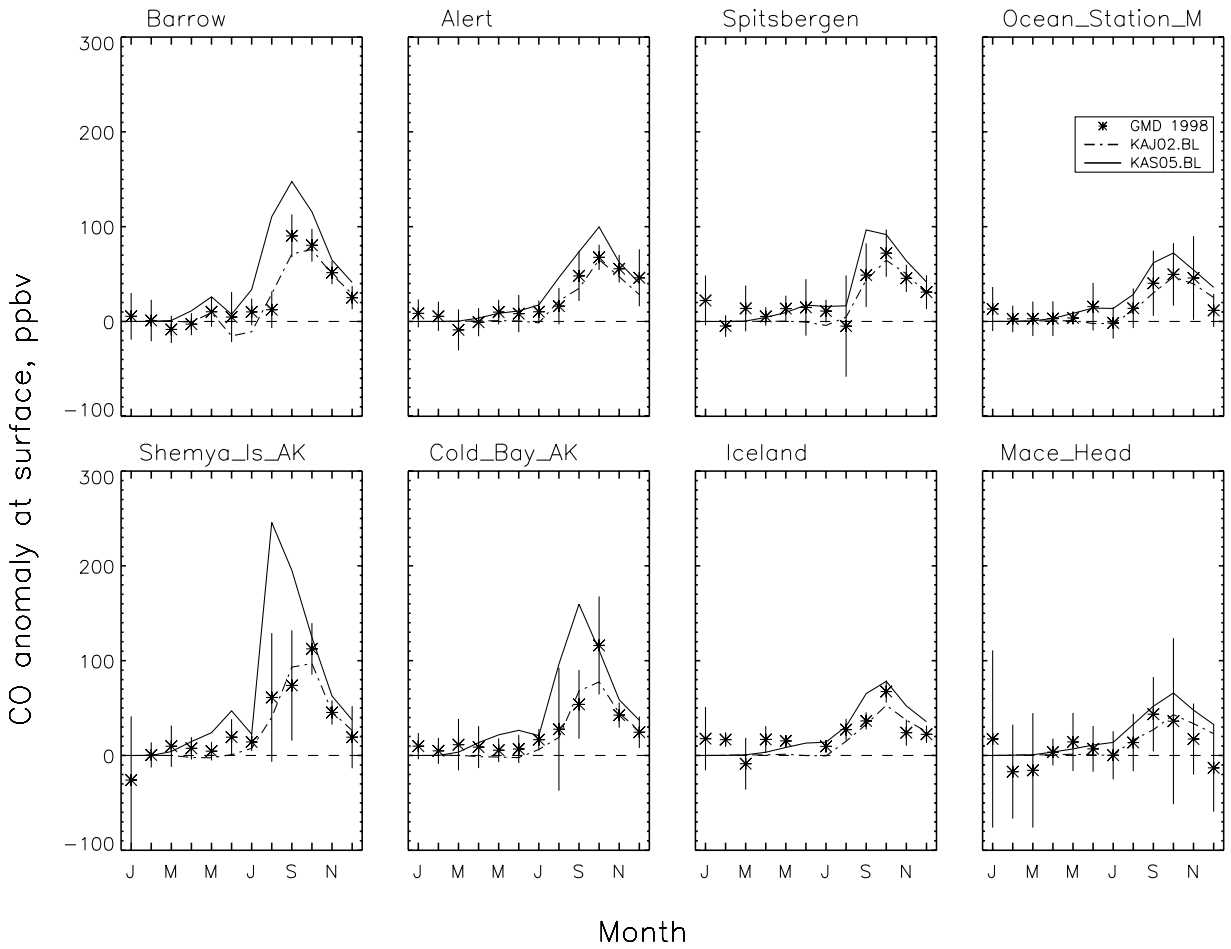

Figure 4. Comparison of the observed and simulated anomaly in CO mixing ratios for 1998, with boreal fire emissions released in the boundary layer only. The symbols (asterisks) are the difference between monthly CO in 1998 and the mean CO for the same month for 1992–1997. The lines are the difference between the simulation with 1998 emissions and the baseline simulation with mean fire emissions, for case KAS05.BL (solid) and KAJ02.BL (dot-dashed).

Europe and western Russia. The enhancement exceeds 30 ppb everywhere north of 30°N over the Atlantic and most of the Pacific. The enhancement in CO is larger in September than in August (when boreal fire emissions are largest) over most of the NH, except in the vicinity of and downwind of the fires, as shown by results for the GMD sites in Figure 7. The fires are still active in September, and the CO lifetime increases from late summer into autumn, leading to a larger anomaly one month after the peak in fire activity.

5. Effects of Boreal Fire Emissions on Tropospheric Ozone and OH

[41] The anomaly in ozone in September 1998 at the surface and at ~500 hPa is shown for the optimal scenario KAS05.D2 in Figure 10. Ozone is enhanced at the surface by 3–6 ppb over Alaska and Canada, by 1–3 ppb over the continental United States, and by 2–4 ppb over the north Atlantic, Europe, and Eurasia upwind of the fires. The largest enhancement, 5–6 ppb, is in the vicinity of the fires (see Figure 1) and over Greenland. In the middle troposphere, the ozone anomaly is 6–10 ppb north of 50°N, except immediately downwind of the Siberian fires where it exceeds 20 ppb. When the emissions are released in the boundary layer only, the enhancements are <2 ppb at the surface and <3 ppb at 500 hPa except in the vicinity of the fires, where they reach 4 ppb at the surface and 500 hPa.

[42] The anomaly in ozone at 500 hPa is spatially correlated with that in PAN (Figure 11). Emissions of NO_x are rapidly converted to PAN in biomass burning plumes [*Mauzerall et al.*, 1998; *Real et al.*, 2007]. PAN is thermally stable in the middle troposphere, but undergoes thermal decomposition as air masses descend, releasing NO_x [*Singh and Hanst*, 1981]. The larger anomaly in surface ozone for case KAS05.D2 compared to KAS05.BL results in part from role of PAN as a source of NO_x, the limiting precursor for ozone formation in the remote atmosphere, as air descends in regions far from the fires; subsidence also transports enhanced ozone from the middle troposphere to the lower troposphere.

[43] We find that hydroxyl concentrations in September are suppressed by over 80% in the vicinity of the fires in the boundary layer, where the CO anomaly is largest, but by about 20% upwind of the fires (not shown). The converse is true at 500 hPa, where OH increases in a small region near the fires, but with little effect elsewhere. Over most of the

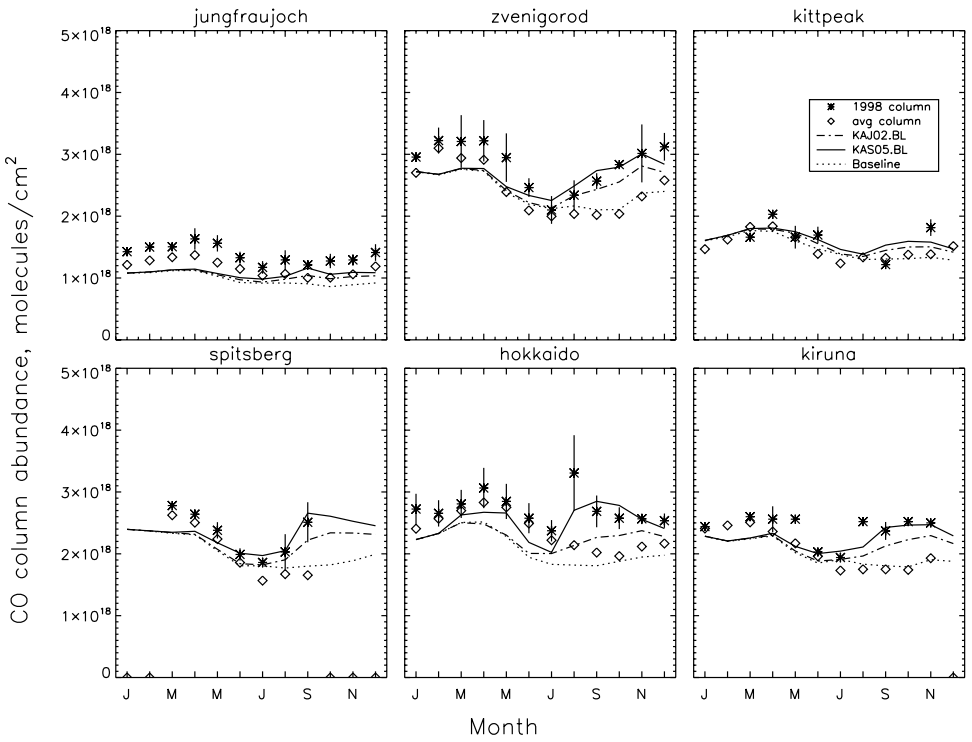

Figure 5. Comparison of the observed and simulated CO column with boreal fire emissions released in the boundary layer only. Column data for 1998 (asterisks) are compared to simulations for 1998 for cases KAS05.BL (solid) and KAJ02.BL (dot-dashed). Column data for 1992–1997 (diamonds) are compared to the baseline simulation with mean fire emissions (short dashes).

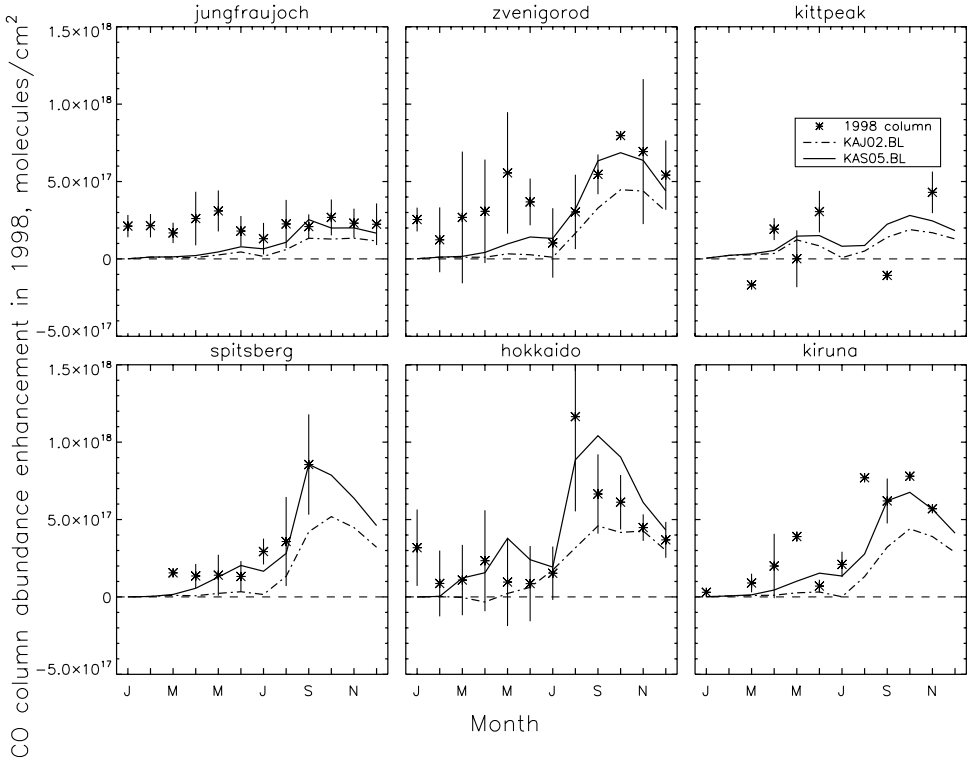

Figure 6. Comparison of the observed and simulated anomaly in the CO column for 1998, with boreal fire emissions released in the boundary layer only. The symbols (asterisks) are the difference between monthly CO in 1998 and the mean CO for the same month for 1996–2001 (excluding 1998). The lines are the difference between the simulation with 1998 emissions and the baseline simulation with mean fire emissions, for case KAS05.BL (solid) and KAJ02.BL (dot-dashed).

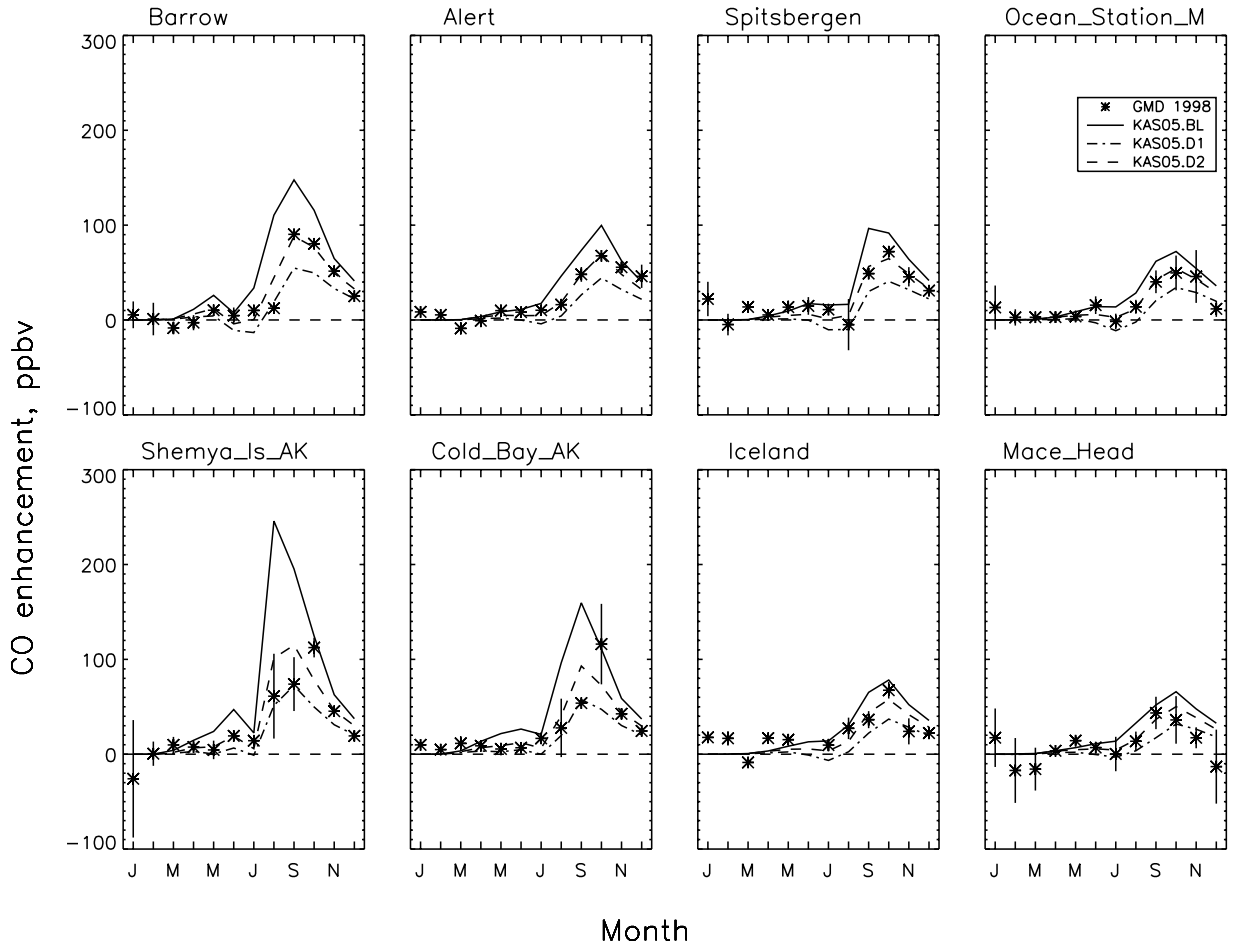

Figure 7. Comparison of the observed and simulated anomaly in CO mixing ratios for 1998, with three scenarios for the injection height and emissions from KAS05. Emissions are injected in the boundary layer only (KAS05.BL, solid), distributed throughout the troposphere (KAS05.D1, dot-dashed) and with 40% in the boundary layer and 60% at 3–5 km (KAS05.D2, dashed). The anomalies are calculated as described in Figure 4.

extratropics however, OH is slightly lower for case KAS05.D2 compared to the base case.

[44] The overall effect of the enhanced boreal fire emissions on global mean OH is low, causing an increase in the lifetime of methylchloroform and methane of less than 0.1% in the simulations for 1998 compared to the baseline. Our results, which show a decrease in methane consumption of 0.17 Tg in 1998, differ from those of *Butler et al.* [2005], who report decrease in mean OH of 2.2% between July 1997 and December 1998, and a decrease in the removal rate of methane of 16 Tg. The difference arises because we do not simulate the effects of the major tropical fires in late 1997 on OH.

6. Summary and Discussion

[45] We investigated the effects of the boreal fires in 1998 on tropospheric composition using the GEOS-Chem model and two gridded inventories for emissions from these fires [*Kajii et al.*, 2002; *Kasischke et al.*, 2005]. The two inventories differ in their estimates of CO emissions from the Siberian fires by a factor of two (43 Tg for KAJ05 and 105 Tg for KAS05) because of different assumptions about fuel consumption and emission factors; they are based on burned areas which differ by only 20%. Surface and column observations of CO were used to evaluate the model results.

[46] We found that the surface data alone would not allow us to constrain both the magnitude of the fire emissions and their injection altitude. The KAJ02 inventory underestimates significantly the observed anomaly in column CO from August to December (Figure 6), even though it reproduces the anomaly in surface CO rather well (Figure 4). The KAS05 inventory reproduces both the surface and column anomalies at most stations in scenario KAS05.D2 with 40% of the emissions released in the boundary layer and 60% at 3–5 km (Figures 8 and 9 and Table 4). In particular, the model matches the large CO anomaly in August in northern Japan (Hokkaido) only when emissions are released above the boundary layer.

[47] We explored only a few scenarios for injection altitude in this study, and clearly the distribution of injection altitudes for boreal fires not well known. Our assumption of a 40:60 split between the boundary layer and 3–5 km was based on evidence that fire plumes are often seen in the middle troposphere, and on the assumptions that the majority of fires in August were crown fires [*Kasischke et al.*, 2005] and that a large fraction of the emissions from such fires are

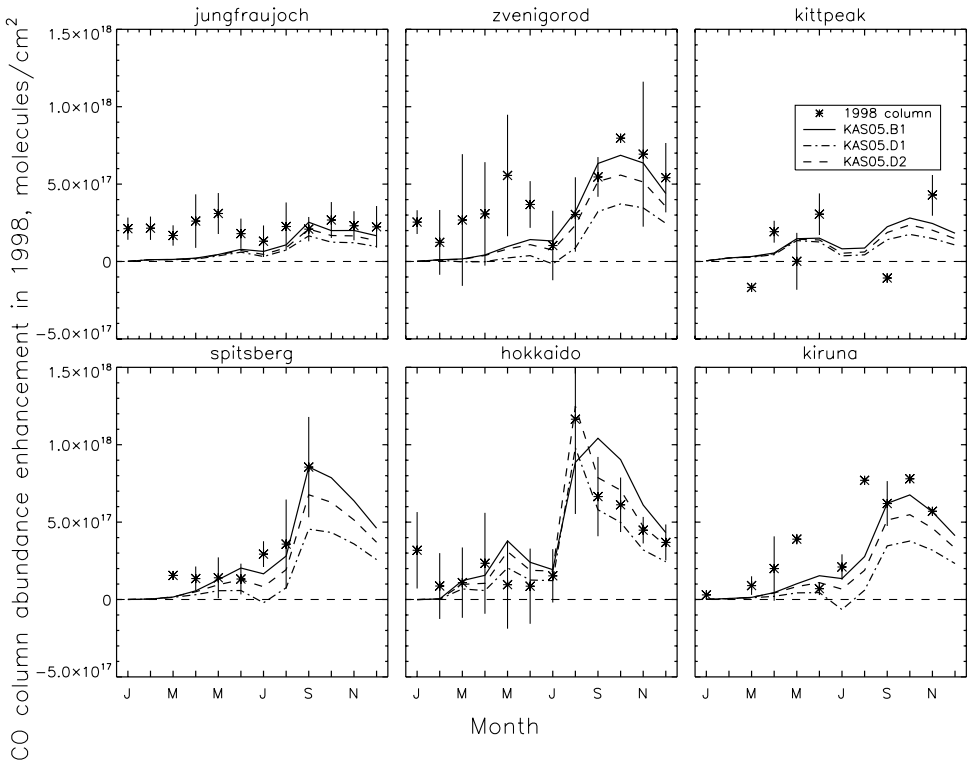

Figure 8. Comparison of the observed and simulated anomaly in the CO column for 1998, with three scenarios for the injection height and emissions from KAS05. Emissions are injected in the boundary layer only (KAS05.BL, solid), distributed throughout the troposphere (KAS05.D1, dot-dashed) and with 40% in the boundary layer and 60% at 3–5 km (KAS05.D2, dashed). The anomalies are calculated as described in Figure 6.

lofted above the boundary layer. A recent study of emissions from the large fires in Alaska and the Yukon in 2004 showed that results were not sensitive to a 40:60 verses a 60:40 split, but that about half of the emissions needed to be released above the boundary layer to match MOPITT CO data; that study used the GEOS-Chem model with resolution of 2° × 2.5° [*Turquety et al.*, 2007].

[48] There is strong evidence that some of the emissions from boreal fires are lofted above the boundary layer, as discussed in the Introduction: observations from aircraft campaigns in the Arctic [*Blake et al.*, 1992; *Shipham et al.*, 1992; *Wofsy et al.*, 1992], experimental fires [*Lavoué et al.*, 2000], model simulations of transport events [*Bertschi et al.*, 2004; *Colarco et al.*, 2004; *Damoah et al.*, 2006], and MISR data for the heights of fresh plumes [*Mazzoni et al.*, 2007]. For 1998 in particular, *Fromm et al.* [2000] showed that aerosols from fires in eastern Russia and Canada were lofted into the lower stratosphere in several episodes between

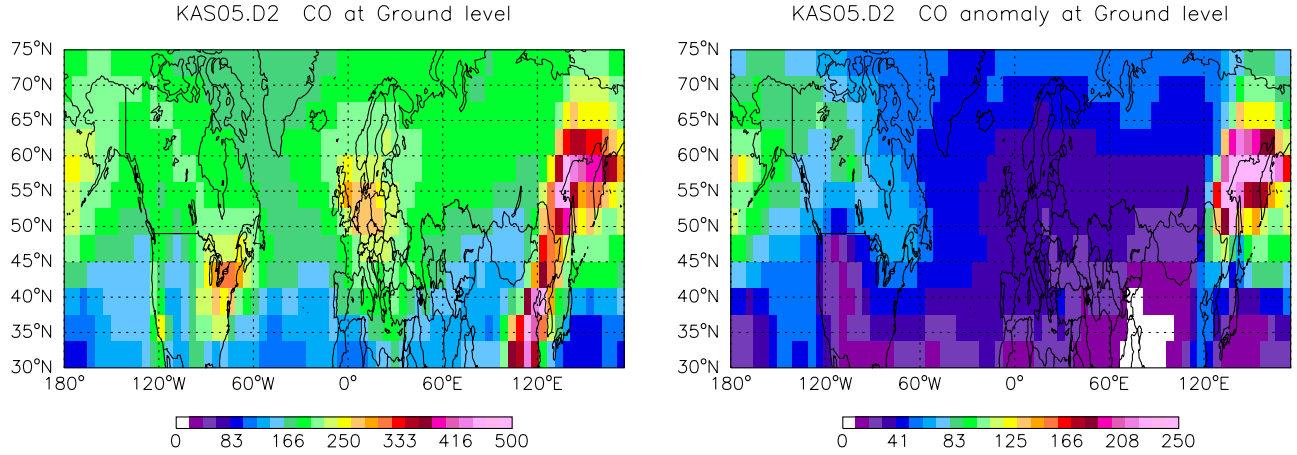

Figure 9. (left) Surface CO and (right) the surface CO anomaly in September 1998 for simulation KAS05.D2, in ppb. The anomaly is the difference between values in simulation KAS05.D2 (which used 1998 emissions) and the baseline simulation (which used mean fire emissions).

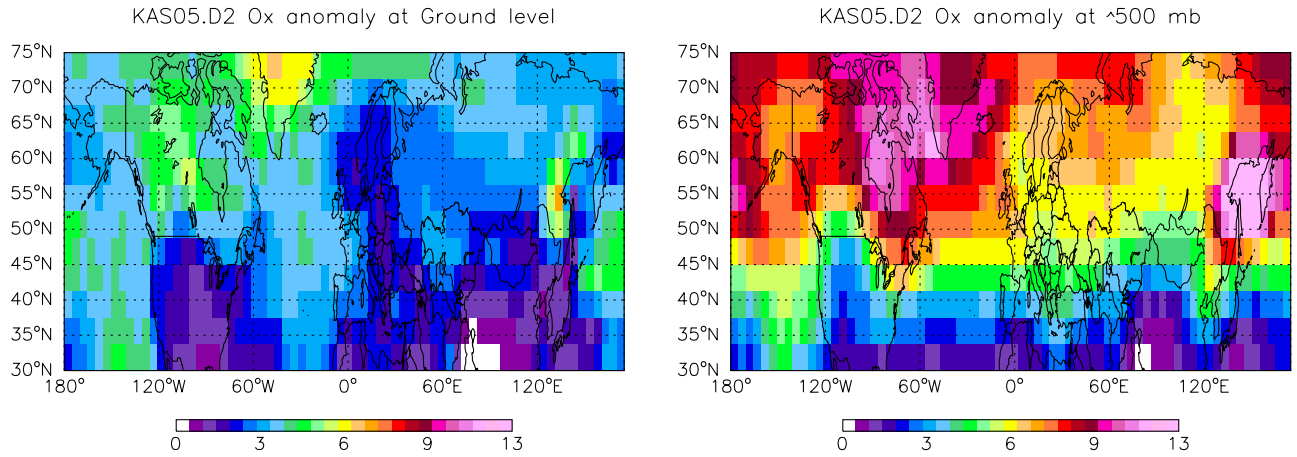

Figure 10. O_x anomaly (primarily ozone) in September 1998 for simulation KAS05.D2 (left) at the surface and (right) at ~500 hPa in ppb. The anomaly is calculated as described in Figure 9.

May to September; *Forster et al.* [2001] showed that large-scale haze layers in August at 3–6 km over Germany were caused by transport of emissions from fires in Canada, and showed that ozone was also enhanced in the haze layers.

[49] Extensive smoke from the boreal fires in 1998 is evident in the aerosol index (AI) product from the total ozone mapping spectrometer (TOMS) on the Earth Probe spacecraft (http://toms.gsfc.nasa.gov), with examples shown in *Zhao et al.* [2002]. *Massie et al.* [2004] showed that the mean optical depth derived from TOMS data was about 3 over eastern Siberia in July to September, as result of the smoke from the fires. The value of the TOMS AI increases with increasing optical depth and increasing altitude of the aerosol layer [*Hsu et al.*, 1999]. Values for the AI greater than 10 are used by M. Fromm to identify aerosols that may be in the stratosphere, and he found such high values on 9 days over Russia in 1998 (M. Fromm, personal communication, 2006). Values of 8 are likely indicative of aerosol well above the boundary layer [*Hsu et al.*, 1999], and we found such values of the TOMS AI on over 40% of days from mid-July to the end of August downwind of the Siberian fires, with several more occurrences in late September.

[50] Our model simulations demonstrate that the effect of the boreal fires on CO was pervasive in the northern extratropics, and in the United States was largest in the north and east (Figure 9). Carbon monoxide was elevated by ~70 ppb at the GMD site in Wisconsin in August and September, 60% above typical values; it was ~100 ppb higher than normal at high-latitude sites in the north Pacific, and 50–80 ppb higher at other sites north of 50°N in September 1998. The anomaly in CO began in late summer and persisted over the winter because of the long lifetime of CO, but its magnitude decreased. This trend was seen in both the surface and column measurements and in the model simulations.

[51] Surface ozone in the model is enhanced by 5–6 ppb in September close to the biomass fires, with relatively high enhancements also in northern Canada and Greenland, but the effect on ozone is smaller elsewhere at midlatitudes. The effect of the fires on ozone is larger at 500 hPa than at the surface for the optimal case KAS05.D2, with an enhancement of 6–10 ppb north of 45°N, and as much as 20 ppb over northeast Canada and eastern Russia. The anomaly in ozone is correlated with that in PAN.

[52] The anomaly in surface ozone is larger when about half the emissions are released above the boundary layer

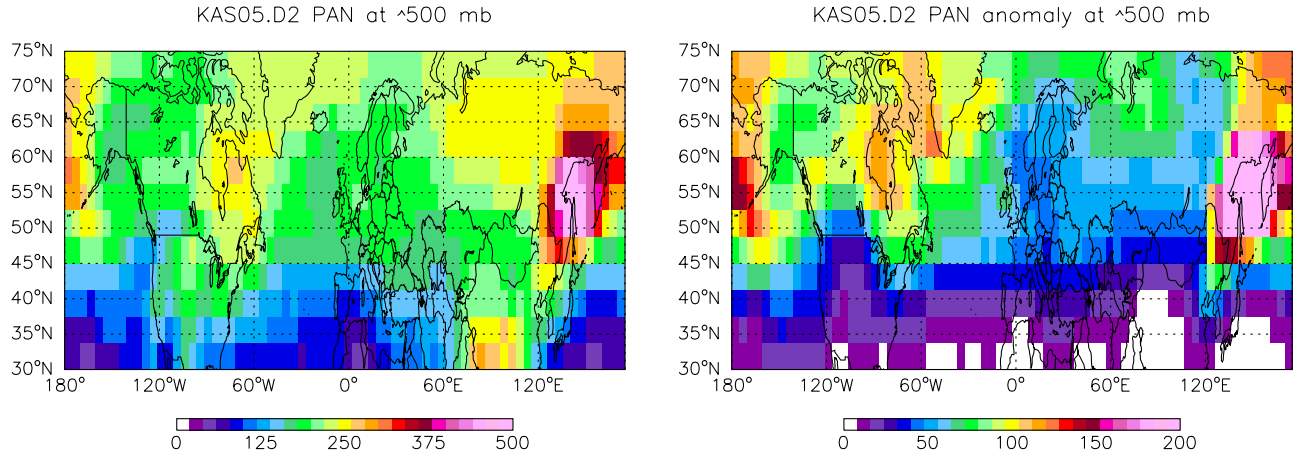

Figure 11. (left) PAN and (right) the PAN anomaly at ~500 hPa in September 1998 for simulation KAS05.D2, in ppt. The anomaly is calculated as described in Figure 9.

than that when emissions are released exclusively in the boundary layer. This is a consequence of the role of PAN (produced from the NO_x emitted by the fires) as a source of NO_x as air descends in regions far from the fires.

[53] The model anomaly in ozone at 500 hPa is 5–10% of mean ozone values at midlatitudes in September, except for northeast Canada, where it is about 15% of mean values. We examined ozonesonde profiles and found plumes with ozone maxima greater than 80 ppb at 3–7 km in two of the weekly profiles in September at Churchill (59°N, 94°W) and Edmonton (54°N, 114°W) and one plume in August at both sites. It is unclear at present if these ozone plumes are a consequence of emissions from the boreal fires, or of anthropogenic emissions in Asia, or if the plumes are stratospheric intrusions, as backward trajectories with the HYSPLIT model (http://www.arl.noaa.gov/) give ambiguous results. One case, 23 September at Churchill, appears to result from boreal fire emissions as the ozone plume, exceeding 120 ppb, is coincident with a massive aerosol cloud that has traveled from Siberia over the preceding few days. Model simulations with daily or weekly fire emissions are needed for a detailed investigation of the causes of the ozone plumes.

[54] The effect of the fires on OH was a relatively small decrease, except in the immediate vicinity of the fires, and we found that the emissions have minimal effects on the global lifetimes for global methylchloroform and methane, because the OH is perturbed at relatively high latitudes and at the end of summer and in early autumn.

[55] Our model simulations included organic carbon and black carbon emissions from the boreal fires to allow for their effects on photochemistry. The TOMS aerosol products (AI and optical depth) show a large perturbation in the extratropics from the boreal fires in 1998 [*Massie et al.*, 2004]. In future work we will use the model aerosol distributions and resulting aerosol optical depths to calculate the instantaneous radiation forcing (direct effect) from these fires, and compare it to that from ozone. *Duncan et al.* [2003a] found that the radiative forcing from aerosols from the major Indonesian fires in 1997 was much larger than that from the perturbation to ozone.

[56] The results shown in this study demonstrate that a significant fraction of boreal fire emissions are injected above the boundary layer. The sensitivity of model results to injection height should be taken into account in future inverse studies of CO emissions from fires. We are presently using MISR data to investigate the height distribution of aerosols emitted from boreal fires, and to explore the meteorological conditions that associated with these observations, so that parameterizations of injection heights from fires can be developed for use in future studies.

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