Transport of boreal forest fire emissions from Canada to Europe

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Abstract. In August 1998, severe forest fires occurred in many parts of Canada, especially in the Northwest Territories. In the week from August 5 to 11, more than 1000 different fires burned >1 x 10⁹ ha of boreal forest, the highest 1-week sum ever reported throughout the 1990s. In this study we can unambiguously show for the first time that these fires caused pronounced large-scale haze layers above Europe and that they influenced concentrations of carbon monoxide and other trace gases at the surface station Mace Head in Ireland over a period of weeks. Transport took place across several thousands of kilometers. An example of such an event, in which a pronounced aerosol layer was observed at an altitude of 3-6 km over Germany during August 1998, is investigated in detail. Backward trajectories ending at the measured aerosol layer are calculated and shown to have their origin in the forest fire region. Simulations with a particle dispersion model reveal how a substantial amount of forest fire emissions was transported across the Atlantic. The resulting aerosol lamina over Europe is captured well by the model. In addition, the model demonstrates that the forest fire emissions polluted large regions over Europe during the second half of August 1998. Surface measurements at Mace Head are compared to the model results for an anthropogenic and a forest fire carbon monoxide tracer, respectively. While wet deposition removed considerable amounts of aerosol during its transport, forest fire carbon monoxide reached Europe in copious amounts. It is estimated that during August 1998, 32%, 10%, and 58% of the carbon monoxide enhancement over the background level at Mace Head were caused by European and North American anthropogenic emissions and forest fire emissions, respectively.

1. Introduction

Boreal forest fires are a frequent and intense disturbance factor in temperate and high northern latitudes.

Typically, about 30,000 to 40,000 wildfires burn in Russia [Kasischke et al., 1999], Alaska, Canada, and Scandinavia per year [Fromm et al., 2000], with a great year-to-year variability [Lavoué et al., 2000]. In some years, more than 12 x 10⁹ ha are burned [Stocks et al., 2000]. Forest fires can produce large amounts of haze and smoke with substantial horizontal and vertical transport [e.g., Westphal and Toon, 1991; Stocks and Flannigan, 1987]. During 1998, substantial amounts of aerosols from Canadian wildfires were observed over Greenland [Hsu et al., 1999] and even transported to the stratosphere [Fromm et al., 2000; Siebert et al., 2000]. Trace gases like carbon monoxide (CO) are affected by wildfires as well. A number of studies in the framework of the NASA Arctic Boundary Layer Expeditions [Harriss et al., 1992, 1994] investigated the influence of forest fires on trace gases in high northern latitudes and showed that forest fires, together with stratospheric intrusions are a potential source for trace gases in the Arctic and sub-Arctic regions. Waibel et al. [2000] presented measurements of highly elevated CO concentrations in the tropopause region over Europe during 1994, which were a result of boreal fires in Canada. The CO concentrations in the lowermost stratosphere varied between typical background values of around 40 parts per
billion by volume (ppbv) and 300 ppbv within the forest fire plume.

Carbon monoxide and aerosols play an important role for atmospheric chemistry and the radiative properties of the atmosphere. Carbon monoxide strongly influences the abundance of the OH radical and initiates various important chemical reactions [e.g., Seinfeld and Pandis, 1998]. For instance, CO is directly involved in ozone chemistry [Crutzen, 1973], and aerosols have an influence on stratospheric ozone concentrations through catalytic chemical reactions [Fromm et al., 2000]. Therefore, a change in aerosol amounts and CO concentrations affect trace gases, which play critical roles for the global climate [Daniel and Solomon, 1998; Logan et al., 1981]. Moreover, large amounts of aerosols can directly influence the shortwave reflectivity of the atmosphere [Christopher et al., 2000; Hsu et al., 1999] and can modify the radiative properties of clouds in an indirect way by acting as cloud condensations nuclei [Christopher et al., 2000].

In order to quantify the chemical and radiative impact of forest fire emissions properly, it is necessary to understand more fully the long-range transport of smoke and trace gases from wildfires. In the tropics the intercontinental transport and effect of biomass burning emissions was investigated by the NASA Transport and Atmospheric Chemistry near the Equator-Atlantic campaign [Fishman et al., 1996]. Recently, Wotawa and Trainer [2000] investigated episodes of high CO concentrations in the southeastern United States during summer 1995. They performed simulations with a Lagrangian transport model taking into account emissions from anthropogenic sources and from wildfires in Canada. The results showed that the Canadian forest fires are episodically the dominant source of the highly elevated CO concentrations in eastern and southeastern United States, regions with the highest anthropogenic emissions worldwide.

In August 1998, severe forest fires occurred in many parts of Canada, especially in the Northwest Territories. In the week from August 5 to 11, more than 1000 different fires burned >10^6 ha of boreal forest, the highest 1-week sum ever reported throughout the 1990s [Canadian Forest Service, http://www.nrcan.gc.ca/cfs, 2000]. In this paper we investigate the origin of an aerosol layer, which was observed at around 3–6 km altitude at different sites in Germany in August 1998. Using a dispersion model, we show that these aerosols were indeed smoke particles released from wildfires in northern Canada and traveled to Europe within ~1 week. Ozone concentrations within the aerosol layer were also elevated, indicating that the photochemical processes within the air mass have been affected substantially. We furthermore show that surface concentrations of carbon monoxide at Mace Head, Ireland, were influenced strongly by transport from the biomass burning regions during the second half of August 1998. With this study a tropospheric long-range transport of forest fire smoke particles and carbon monoxide from continent to continent over an extended time period is fully documented.

2. Models and Measurement Data
2.1. Modeling Tools

Two transport models, both driven with model level data from the European Centre for Medium-Range Weather Forecasts (ECMWF) [1995] (T213 L31 model), were used to interpret the measurement data. The ECMWF data as available for this study have a horizontal resolution of 1° and a time resolution of 3 hours (analyses at 0000, 0600, 1200, 1800 UTC; 3 hour forecasts at 0300, 0900, 1500, 2100 UTC) and cover the Northern Hemisphere.

The first tool used to calculate three-dimensional trajectories is the Lagrangian trajectory model FLEXTA (version 3.2d) [Stohl et al., 1995]. FLEXTA uses bicubic horizontal, quadratic vertical, and linear time interpolation to determine the three wind components at a trajectory position and employs the numerical method of Petterssen [1940] for the trajectory calculations. The model has been validated with constant level balloon flights [Stohl and Koff, 1998], massed gas balloon flights [Baumann and Stohl, 1997] and meteorological tracers [Stohl and Seibert, 1998] and recently underwent an intercomparison exercise with other trajectory models [Stohl et al., 2000].

The second tool is the Lagrangian particle dispersion model FLEXPART (version 3.2) [Stohl et al., 1998; Stohl and Thomson, 1999]. FLEXPART was developed to calculate the transport and dispersion of non-reactive tracers. It has recently been validated with data from three large-scale tracer experiments in North America and Europe [Stohl et al., 1998] and performed very well in comparison with other models. FLEXPART was also used by Wotawa and Trainer [2000] to simulate the transport of pollutant plumes from Canadian forest fires. FLEXPART treats advection and turbulent diffusion by calculating the trajectories of a multitude of particles. Stochastic fluctuations of the three wind components, obtained by solving Langevin equations [Stohl and Thomson, 1999], are superimposed on the grid-scale winds interpolated from ECMWF data to simulate transport by turbulent eddies. In the boundary layer, the magnitude and the Lagrangian decorrelation times of the turbulent wind components are obtained from a parameterization by Hanna [1982]. Above the boundary layer, the turbulent components are set to small values in dependence of the wind shear. Tracer concentrations on a three-dimensional grid are determined applying a kernel method, which is equivalent to, but more accurate than summing up the masses of all particles contained in a grid cell and dividing by its volume. (Documentation manuals and the source codes of both FLEXTA and FLEXPART can be obtained via the Web at the address http://www.fw.tum.de/EXT/LST/METEO/stohl/.)
We carried out four FLEXPART model runs, simulating the transport of CO originating from European and North American anthropogenic emissions and from Canadian forest fires. The results of these simulations will assist us in section 3 in attributing measured CO concentrations in Europe to the different CO sources. Particles were released at the locations of the CO sources, and all particles carried an equal fraction of the total mass of CO emitted from each source category. The chemical lifetime of CO varies from 1 month in the tropics to 4 months at midlatitudes in spring and fall to much longer lifetimes in winter [Seinfeld and Pandis, 1998]. Since chemical destruction of CO was not explicitly accounted for in the simulations, we assumed that CO decays exponentially with an e-folding lifetime of 50 days. This has only a moderate impact on the model results, since we consider transport times much shorter than the CO lifetime. Sensitivity experiments using different CO lifetimes confirmed this.

The first model calculation simulated the transport of anthropogenic CO emissions from Europe. According to the emission inventory of the cooperative programme for monitoring and evaluation of the long-range transmission of air pollutants in Europe (European Monitoring and Evaluation Programme, http://www.emea.int, 2000) for the year 1997, 5.6 x 10^8 particles were released from a grid with a resolution of 50 km. Constant emissions of 1/12 of the annual sum were assumed throughout August 1998.

The second model simulation considered anthropogenic CO emissions from the United States, taken from an inventory on a county basis for the year 1996 [Environmental Protection Agency (EPA), 1997], and from Canada using an inventory with base year 1985 [EPA, 1989]. The emission inventories were transformed to a 2° latitude/longitude grid, from which 4.5 x 10^8 particles were emitted during the simulation period without imposing temporal variations. Both European and North American anthropogenic emissions were put into a layer up to 150 m above ground level.

The third model simulation used 6.6 x 10^8 particles to calculate the transport of CO emissions from forest fires in Canada. The forest fire CO emissions during August 1998 were assumed to be proportional to the area burned, with 4500 kg CO released per hectare of burned forest. This estimate is consistent with emission measurements from crown fires performed in the Canadian Northwest Territories (NWT) [Cofer et al., 1998] and was confirmed by long-range transport model calculations [Wotawa and Trainer, 2000]. Since ~50% of the area burnt during the investigation period was destroyed in the Northwest Territories and since most of the fires there were crown fires, we can presume that this emission estimate is a valid approximation. We have to acknowledge, though that different phases of the fires (e.g., crown fires, smoldering fires) and different types of fuel may cause different CO emissions per hectare.

The weekly total area burned in Canada was taken from reports of the Canadian Forest Service (CFS) [Canadian Forest Service, 2000]. A first disaggregation to specific provinces was performed according to the weekly CFS situation reports. In a second step the locations of the forest fires within the provinces were determined, on the basis of two sources of information: First, there is a detailed fire inventory available for the NWT, which reports coordinates, date of detection, date of extinction, and areas burned of each detected wildfire [Government of the Northwest Territories, 2000]. All of the large fires (>20,000 ha) burned through the whole simulation period but with varying intensity. According to this, we selected three release boxes (see Figure 1), which represent a number of fire hot spots located in each box. For all other provinces, no site-specific information was available. Therefore we selected five release areas of 3°-5° resolution according to the daily satellite-based hot spot maps created by the Canadian Center for Remote Sensing [Natural Resources Canada, 2000] (see Figure 1) following the approach adopted by Wotawa and Trainer [2000]. The coordinates, areas burned during August 1998, and the resulting CO emissions are summarized in Table 1.

In the FLEXPART simulation, particles were released within the boxes shown in Figure 1, with weekly rates according to the CFS reports. The particles were started randomly distributed between the surface and 2500 m above ground level. During well-developed

Figure 1. Release areas as defined for the FLEXPART simulation. One area is situated in the Yukon Territory (YKT), three in the Northwest Territories (NWT1, NWT2 and NWT3), and one in each of the provinces Alberta (ALB), Saskatchewan (SASK), Manitoba (MANI) and Quebec (QUEB).
Table 1. Selected Forest Fire CO Release Locations in Canada, Area Burnt in August 1998, and Resulting CO Emissions.

<table>
<thead>
<tr>
<th>Location</th>
<th>Area, 10^3 ha</th>
<th>CO Emission, kt</th>
</tr>
</thead>
<tbody>
<tr>
<td>NWT1</td>
<td>205.6</td>
<td>925.3</td>
</tr>
<tr>
<td>NWT2</td>
<td>123.5</td>
<td>555.6</td>
</tr>
<tr>
<td>NWT3</td>
<td>492.4</td>
<td>2216.0</td>
</tr>
<tr>
<td>QUEB</td>
<td>250.7</td>
<td>1128.2</td>
</tr>
<tr>
<td>SASK</td>
<td>302.3</td>
<td>1360.3</td>
</tr>
<tr>
<td>MANI</td>
<td>227.8</td>
<td>1025.3</td>
</tr>
<tr>
<td>ALB</td>
<td>104.6</td>
<td>470.7</td>
</tr>
<tr>
<td>YKT</td>
<td>139.3</td>
<td>626.7</td>
</tr>
<tr>
<td>SUM</td>
<td>1846.3</td>
<td>8308.2</td>
</tr>
</tbody>
</table>

Nearly 60% of this area was destroyed during the week from August 5 to 11. Please note that the CO emissions of the United States (base year 1996) are ~6600 kt per month, those of Germany and France are 1200 kt.

2.2. Measurement Data

This study uses lidar measurement data from stations in Germany and surface in situ measurement data from Mace Head, Ireland. The lidar measurements were taken within the framework of the German Aerosol Lidar Network and the Lindenberg Aerosol Characterization Experiment (LACE 98). Since 1997, regular measurements of the vertical distribution of aerosols in the atmosphere are performed with lidar instruments at five stations in Germany, which are the Max Planck Institute for Meteorology, Hamburg (MPI); the Leibniz Institute for Atmospheric Physics, Kühlungsborn (IAP); the Institute for Tropospheric Research, Leipzig (IFT); the Meteorological Institute, University of Munich (MIM); and the Fraunhofer Institute for Atmospheric Environmental Research, Garmisch-Partenkirchen (IFU). In 2000, this lidar network was extended to the European Aerosol Research Lidar Network (EARLINET), which consists of 21 stations distributed over most of Europe [Schneider et al., 2000]. During LACE 98 in July/August 1998 (A. Ansmann et al., Lindenberg Aerosol Characterization Experiment LACE 98: Overview, submitted to Journal of Geophysical Research, 2001), the three mobile instruments of MPI, MIM, and IFT of the German lidar network were stationed at Lindenberg (52°N, 14°E, 100 m), whereas the systems in Kühlungsborn (54°N, 12°E, 70 m), Leipzig (51°N, 12°E, 92 m), and Garmisch-Partenkirchen (47°N, 11°E, 730 m) continued their routine observations. In section 3.1 we present measurements performed at these four locations (marked in Plates 2, 4, and Figure 4) during an episode of high aerosol load in the free troposphere on August 9 and 10, 1998.

The systems of the aerosol lidar network typically operate at wavelengths of 355, 532, and 1064 nm. Standard output products are vertical profiles of the particle backscatter coefficient at these wavelengths. The systems of IFT, MPI, and IAP are equipped with nitrogen Raman channels and allow an independent determination of particle extinction-coefficient profiles. A detailed description of the lidar data evaluation for the measurements taken in Lindenberg on August 9 and 10, 1998, as well as results of measurements at multiple wavelengths and of the inversion to microphysical particle properties in the free-troposphere aerosol layer are given by U. Wandinger et al. (Characterization of optical and microphysical particle properties from multiwavelength lidar and airborne in situ measurements in biomass-burning and industrial-pollution aerosols, submitted to Journal of Geophysical Research, 2001). Airborne in situ measurements in the aerosol layer are described by M. Fiebig et al. (manuscript in preparation, 2001).

Mace Head is located at the west coast of Ireland (53°N, 10°W; marked in Plates 2, 4, and Figure 4). Car-
bon monoxide measurements were made using a model RGA3 reduction gas analyzer (Trace Analytical, Inc., California) on a 20-min cycle with the sequence calibration standard, ambient air, calibration standard, etc. To determine any detector nonlinearity, a suite of calibration standards covering the concentration range 50-500 ppbv has also been analyzed. The calibration standards used at Mace Head to determine ambient CO concentrations are linked closely to the scale developed at NOAA Climate Monitoring and Diagnostics Laboratory (CMDL) [Novelli et al., 1991]. This scale was propagated to Commonwealth Scientific and Industrial Research Organization (CSIRO) in the period 1991-1992 by a suite of standards in high-pressure aluminum cylinders. An intercomparison experiment conducted during 1994 showed very close agreement between the two laboratories at that time [Novelli et al., 1998].

A commercial instrument known as an ethalometer (Magee Scientific, Model AE 9) was used to measure the attenuation of visible light from which the absorption coefficient or mass concentration of black carbon can be inferred, assuming an attenuation cross-section value of 19 m² g⁻¹. Condensation nuclei number concentration was measured using a TSI model 3025 condensation particle counter with a 50% cut size at 3 nm particle diameter.

Ozone measurements were obtained with an UV spectrometer (Model 8810, Monitor Labs). Measurements were acquired every 10 s and reported as hourly averages. Calibration was performed every 3 months against a primary UV photometer [Sweeney and Stacey, 1992]. CFC-11 measurements are recorded routinely as part of the Advanced Global Atmospheric Gases Experiment (AGAGE). A detailed description of the instrumentation and calibration is given by Prinn et al. [2000].

3. Long-Range Transport of Forest Fire Haze

3.1. Case Study August 9-10, 1998

Measurements obtained with lidars of the German aerosol lidar network (see section 2.2) revealed an aerosol layer at around 3-6 km altitude on August 9 and 10, 1998. As an example, Plate 1 shows the particle backscatter coefficient for this period taken with the IFT multiwavelength lidar at Lindenberg. The aerosol layer is clearly elevated above the atmospheric boundary layer, where strong backscatter is also seen. The lamina with the highest optical thickness has a vertical extent of ~500 m, is initially located at around 4.5 km altitude and descends to ~3 km altitude during August 10. On August 9, relatively high particle concentrations were also detected up to 6 km, above the lamina with the densest haze.

It is interesting to note that on both days, observers at many meteorological stations in northern Germany reported the presence of cirrus clouds [Institut der Freien Univ. Berlin, 1998; Deutscher Wetterdienst, 1998]. The lidars, however, only detected clouds during short periods on August 9, but not on August 10. Furthermore, a more detailed analysis of German weather charts [Institut der Freien Univ. Berlin, 1998; Deutscher Wetterdienst, 1998] suggests that the synoptic situation was not particularly conducive to cirrus cloud formation (see also Plate 2c). Thus it appears that the reported cirrus clouds was in fact the haze layer.

Chemical and microphysical analysis of in situ measurements taken aboard a research aircraft revealed that soot contributed ~35% of the aerosol mass and made up the largest part of the light absorbing component of the aerosol (M. Fiebig et al., manuscript in preparation, 2001). This led to the hypothesis that the aerosols originated from biomass burning. Routine back trajectories calculated by the German Weather Service for three levels at the site of Lindenberg suggested that the haze layer was transported from Greenland. These routine back trajectories did not go back far enough, but from an analysis of the synoptic situation it seemed that extending them backwards in time would bring them close to the forest fires burning in Canada at that time.

We will now analyze the transport processes producing the haze layer over Germany in detail. Plate 2 shows the geopotential height at 500 hPa on August 4, 6, and 9 at 1200 UTC. On all these days, there is a mainly westerly flow with embedded troughs and ridges from Canada over Greenland and the Atlantic to northern Germany. Total Ozone Mapping Spectrometer (TOMS) aerosol index data confirm that smoke generated by Canadian forest fires drifted eastward (Plate 2) and passed over Greenland, as already reported by Hsu et al. [1999], a few days before it reached southern Scandinavia and northern Germany.

In order to investigate the origin of the aerosol lamina, we calculated 192-hour back trajectories terminating at the lidar sites every 250 m from the surface to 10 km above ground level. As an example we show the results for Lindenberg on August 9 at 0600 UTC (Plate 3). For clarity, not all trajectories are plotted, but only those terminating at the altitude of the lamina and a few typical ones below and above. The trajectories ending at the altitude of the haze lamina had their origin in the NWT, almost exactly where the largest forest fires burned (compare Plate 3 with Figure 1 and Table 1). The polluted air mass was first transported offshore at low levels and was then lifted up to ~7 km in the southeastern part of the low located to the northwest of Greenland on August 6 (see Plate 2b). Finally, the air mass descended again in the ridge over the eastern Atlantic and western Europe (Plate 2c) and arrived at around 4-5 km above Lindenberg. This transport pattern remained basically unchanged throughout the
Plate 1. Time-height contour of the 532-nm particle backscatter coefficient at Lindenberg on August 9 and 10, 1998. The measurements were taken with the IFI multiwavelength lidar during LACE 98 with a time resolution of 30 s and a range resolution of 15 m. The data were smoothed with a running average of 2.5 min in time and 75 m in height.

2-day measurement period, although the lifting was less strong at later times.

The FLEXPART simulation of the forest fire CO tracer transport reveals how the air mass polluted by the forest fires traveled across the Atlantic Ocean. Plate 4 shows three snapshots of the total CO tracer column obtained for the same dates as shown in Plate 2. On August 4 (Plate 4a) a filament of CO tracer already stretches across the Atlantic Ocean toward Europe, but the CO columns within this filament are relatively low. Comparing the FLEXPART CO tracer column with the TOMS aerosol index plotted in Plate 2a shows qualitative agreement between the shapes of the plumes close to the sources. However, the FLEXPART filament over the Atlantic Ocean is missing in the TOMS data. This can be explained by wet removal of the aerosols, as the filament is also not present in plots of the total aerosol tracer columns (not shown).

Two days later, on August 6, the FLEXPART results show a large plume of forest fire CO travelling north-eastward over Greenland (Plate 4b). Both the shape and the position of this plume are in good agreement with the TOMS aerosol index data (Plate 2b). Again, the tracer filaments over the eastern Atlantic Ocean and over Europe are missing in both the TOMS data and the aerosol tracer data.

On August 9, both the FLEXPART results (Plate 4c) and the TOMS data (Plate 2c) show that parts of the plume have crossed the Atlantic Ocean and are located over Europe. The shape of the filament that stretches from the east coast of Greenland to eastern Europe as seen in the TOMS data agrees well with the centerline of the FLEXPART plume. Also the orientation and shape of the various other maxima in the TOMS aerosol index agree well with the FLEXPART simulation, but it is obvious that the TOMS data capture only a small part of the true forest fire plume, which by that time covers large parts of the North Atlantic region. In particular, the forest fire plume has now also reached the locations of the lidar measurement sites.

Plate 5 shows time-altitude sections of the CO tracer (Plate 5a) and the aerosol tracer (Plate 5b) for the location of Lindenberg. Because of wet (and dry) deposition, the concentrations of the aerosol tracer are lower than those of the CO tracer by ~2 orders of magnitude. Neither of the tracers can be compared quantitatively with the aerosol lidar data, but the measured lidar profiles superimposed on the model results in Plate 5b nevertheless demonstrate that FLEXPART is able to reproduce the structure of the observed lamina (Plate 1). The simulated lamina for both tracers is much coarser than the observed one, but both its altitude and its
Plate 2. Geopotential height in geopotential decameters (gpdm) on the 500-hPa level on (a) August 4, (b) 6 and (c) 9 at 1200 UTC. The geopotential is contoured every 8 gpdm. Superimposed on the geopotential charts as color shadings are the aerosol index data from the Total Ozone Mapping Spectrometer (TOMS). The locations of the measurement sites Garmisch (G), Kühlungsborn (K), Leipzig (Le), Lindenber (Li) and Mace Head (M) are marked.
Plate 3. The 192 hour three-dimensional backward trajectories terminating at Lindenberg for various altitudes on August 9 at 6000 UTC: (a) a horizontal projection of the trajectories, with the color-coding according to the label bar referring to the actual heights (in km asl); positions are marked with asterisks every 24 hours. The dots at the trajectory starting points indicate the heights where the trajectories terminated along the lidar profile, with the color code of the dots corresponding to the colors in Plate 3(b). The same information is also provided by the line widths (thinner lines representing trajectories terminating at higher levels). (b) Time-height profiles of the trajectories with both color and line width indicating their ending height at $T=0$.

descent to lower levels at the end of the measurement period are quite well captured by the FLEXPART simulation. The major difference between the two tracers is that the secondary maximum at $\sim$2 km altitude on August 10 is much weaker for the aerosol tracer than for the CO tracer. Also the descent of the lamina on August 10 is more emphasized in the aerosol tracer data. Both facts are in good agreement with the lidar data and demonstrate the importance of the washout for the aerosols.

Figure 2 shows the measured backscatter coefficient and the corresponding aerosol tracer profiles at different times on August 9 for Kühlungsborn and Lindenberg and on August 10 for all stations within the German aerosol lidar network. Figure 2 demonstrates how the aerosol layer descended to lower levels at all sites during August 10 and finally disappears in the evening on August 10. Although the model results do not show the detailed structure of the measured profiles, they are qualitatively in good agreement with the measurements. A clear link has thus been established between the forest fires in Canada and the haze layer observed over Europe.

The peak in the backscatter coefficient profile at around 6 km altitude on August 9 (Figure 2a, solid curve) was caused by a thin cloud layer, which was present during the measurement and is thus missing in the model profiles. Both the lidar data and the model results for Garmisch are difficult to interpret. Boundary layer heights at Garmisch, located in an Alpine valley, are greater than at the other stations and thus the haze layer at $\sim$4 km altitude is not as clearly separated from
Plate 4. Total columns of the CO tracer modeled with FLEXPART for (a) August 4, (b) 6 and (c) 9 at 1200 UTC. Please note the logarithmic scale. The locations of the measurement sites Garmisch (G), Kühlungborn (K), Leipzig (Le), Lindenberg (Li) and Mace Head (M) are marked.
Plate 5. Altitude-time section of the FLEXPART (a) forest fire CO tracer and (b) aerosol tracer over Lindenberg on August 9 and 10. Superimposed on the aerosol tracer in Plate 5(b) are profiles of the lidar measurements. For the scale of the lidar data, see Figure 2.
the boundary layer as at the other sites. The altitudes of the model profiles may be shifted to lower altitudes because the model topography is below the real topography.

The FLEXPART simulation suggests that up to 80 ppbv of forest fire CO, on top of the CO background, were present over Lindenberg (see Plate 5a). This doubling of the normal CO background should
have a strong effect on ozone production. Fortunately, there is an ozone sounding available for Lindenberg on August 10 at 1100 UTC. The ozone profile (Figure 3) shows enhanced ozone values of almost 80 ppbv within the haze layer at 3–3.5 km over a tropospheric background of ~55 ppbv.

3.2. Forest Fire and Anthropogenic CO Over Europe During August 1998

The FLEXPART simulation for the remainder of August 1998 suggests that during the second half of August the whole North Atlantic and European region gets polluted with CO from the Canadian forest fires. As an example we show the forest fire CO tracer mixing ratio in the lowest model layer for August 21 at 1200 UTC (Figure 4). Maximum CO tracer mixing ratios over the Atlantic Ocean are above 100 ppbv and values of the order of 30 ppbv are typical for large regions of Europe. Figure 5 presents timeseries of the average mixing ratios at 3000 m altitude within a box covering western and central Europe (10°W to 20°E, 40°N to 60°N) of the North American and European anthropogenic CO tracers, and the forest fire CO tracer, respectively. Because of model spin-up, mixing ratios of all tracers are low during the first days of August. However, it is clearly seen that during the second half of August CO tracer import from North America makes a larger contribution to the total CO tracer than the European emissions. Specifically, mixing ratios of the forest fire tracer are higher than those of both other tracers during most of the time. If real, such a strong signal should be detectable in CO measurement data from Europe. Mace Head is ideally located for this purpose because it normally receives low levels of pollution from the Atlantic, interrupted only by short episodes of transport from Europe [Ryall et al., 1998]. Furthermore, the FLEXPART results suggest that Mace Head is frequently located in the corridor of forest fire pollution on its way to continental Europe.

Figure 6 shows the CO mixing ratio (shaded curve) and the mixing ratio of the halocarbon CFC-11 (trichlorofluoromethane, solid curve) measured at Mace Head during August 1998. Carbon monoxide is a tracer for natural as well as for anthropogenic emissions, while halocarbons are produced solely by human activities [Lovelock, 1972], and increases in the concentrations of the halocarbons are an indicator for polluted air masses from anthropogenic sources [Ryall et al., 1998; Simmonds et al., 1996; Cunnold et al., 1986; Prinn et al., 1983]. Therefore the positive deviations of CFC-11 from its baseline value of ~261 pptv on August 9 and 14 and at the end of the month (August 29 to 31) can be taken as anthropogenic pollution events. Coincident peaks were also observed in other halocarbon data (not shown). From August 17 to 28 the CO mixing ratio shows enhanced values, while the time series of CFC-11 is at its baseline, indicating that the CO enhancement is not associated with anthropogenic pollution.
Consistent with the FLEXPART simulation (Figure 4), this suggests that the polluted air masses can be traced back to the forest fire emissions. The CO peak on August 11 and the first part of the CO peak on August 14 are also associated with baseline values in CFC-11 and therefore might be a result of the forest fires, too. The second half of the peak on August 14 is presumably linked to advection of an air mass from anthropogenic sources, which is indicated by the elevated CFC-11 values. The low values of CO on August 23 can be linked to a short episode of transport of tropical air, which is characterized by low CO.

The anthropogenic pollution events are well reproduced by the FLEXPART model. This is documented in Figure 7, which shows the modeled American (dashed curve) and American plus European (solid curve) anthropogenic CO tracers compared to the measured CFC-11 data; 97 ppbv were added to the modeled tracer mixing ratios in order to account for the CO background that is not simulated. This value was derived with a correlation analysis described below. It is somewhat higher than the typical background conditions during summertime in the Northern Hemisphere. A possible reason for this is discussed in section 4. Figure 7 documents nicely that the increase seen in the measurement data at the end of August is mainly a result of European emissions, while the smaller CFC-11 peak on August 14 was caused by transport from North America. During August 1998 the squared Pearson correlation coefficients of the European CO tracer, the North American CO tracer, and the sum of the two to the measured CFC-11 data were 0.61, 0.002, and 0.55, respectively. This means that more than half of the variance of the CFC-11 data can be explained by the simulation of the transport from Europe.

In order to find out the relative contribution of anthropogenic emissions and forest fires to the observed CO, we compared the measurements with the results of the FLEXPART model for the different tracers (Figure 8): the American anthropogenic CO tracer (dashed
curve in Figure 8a), the American plus European anthropogenic CO tracer (solid curve in Figure 8a), the forest fire CO tracer (solid curve in Figure 8b) and the sum of the anthropogenic and the forest fire tracers (solid curve in Figure 8c). Again a background of 97 ppbv was added to the modeled tracer mixing ratios. Figure 8c documents how well the model reproduces the observed CO concentrations. It is evident from Figure 8 that the observed peak on August 11, the first part of the observed peak on August 14 and the observed time series of CO during the second half of August cannot be explained without accounting for the forest fire CO. Between August 17 and 29, concentrations of the forest fire tracer exceed those of the anthropogenic tracers by more than a factor 3 and do not decrease before the end of the month. The strong pollution event on August 29-31, which is a result of European emissions for the most part (Figure 8a), is underestimated by the model (Figure 8c). Possible reasons for this are that the assumed European emissions were too low for this period, the simulated boundary layer heights were too high, or the simulated pathway of the polluted air mass missed a significant emission region that actually contributed to the measured CO.

To determine quantitatively the influence of the North American and European anthropogenic and the forest fire CO emissions on the CO mixing ratios at Mace Head during August 1998, we carried out a statistical analysis. For this we averaged all measurement data into 3-hour bins corresponding to the time resolution of the model output. This left 242 valid values. Then we correlated the modeled tracer mixing ratios with the measured CO concentrations, yielding a linear equation of the form $CO = CO_{bg} + S \times CO_{m}$, where $CO_{m}$ and $CO_{bg}$ are the measured and simulated CO mixing ratios, respectively, $CO_{bg}$ is the CO background that is not accounted for by the model, and the slope $S$ measures to what extent the model simulation overestimates ($S < 1$) or underestimates ($S > 1$) the contribution from a source. The variance of the measured CO explained by the fit is given by the squared Pearson correlation coefficient ($r^2$). We calculated several statistics: First, we correlated each CO tracer individually with the measurements; second, we correlated the sum of the anthropogenic tracers and the sum of all tracers with the measurements; and third, we carried out a multiple linear regression analysis using all three tracers as predictors, which allows to separate their contributions to the measured CO mixing ratios.

The results of the statistical analysis are reported in Table 2. It is evident that the forest fires alone account for almost half of the variance of the measurement data, whereas European and North American emissions alone explain much less (37% and 21%, respectively). The sum of all tracers together accounts for 72% of the variance. The mean simulated mixing ratio due to the forest fires (15.8 ppbv) is much higher than the means due to the anthropogenic emissions (3.6 and 1.6 ppbv). However, in order to estimate the true relative contributions we also have to consider the slopes of the fits, which indicate that the anthropogenic emissions have been underestimated. Since the slopes obtained from normal linear regression analysis are misleading if the tracers are correlated with each other, we use multiple linear regression to determine the contribution of each tracer to the measured CO. Multiple linear regression explains 82% of the variance of the measurement data. The slopes for the anthropogenic tracers are lower for the multiple linear regression fit than for the linear one, but they are still above unity, indicating that the model underestimates the anthropogenic contribution. On the other hand, the forest fire CO is slightly overestimated. Multiplying the mean modeled CO tracer mixing ratios with the corresponding slopes of the multiple linear regression fit, we can estimate the contribution of each source to the measured CO. The mean contributions to the total enhancement over the background level are 7.2, 2.2, and 13 ppbv, or 32%, 10%, and 58% for the European, North American, and forest fire tracer, respectively.
Similar to CO, black carbon is a tracer for natural as well as for anthropogenic emissions. However, as part of a multicomponent aerosol, black carbon can easily be washed out by rain. The solid curve in Figure 9a shows the black carbon data measured at Mace Head. The peaks on August 9 and at the end of the month are linked to increases in CFC-11 and can therefore be traced back to human activities (compare the solid curves in Figures 6 and 9a). In contrast to CO, black carbon does not show strongly enhanced values between August 17 and 28 (Figure 9a). Solely the elevated values on August 13 and around August 18 are likely due to the forest fire emissions, as they are not associated with anthropogenic pollution episodes. While much lower than during European pollution events, black carbon concentrations on these days are still above the normal background (5-40 ng m\(^{-3}\)) and exceed the maximum concentrations (75 ng m\(^{-3}\)) observed in marine air masses at Mace Head [Jennings et al., 1997]. The simulated forest fire aerosol tracer which was subject to washout qualita-
Table 2. Results of the Linear Regression and Multiple Linear Regression Analyses for the Different Tracers and the Measured CO Data.

<table>
<thead>
<tr>
<th>Tracer</th>
<th>$r^2$</th>
<th>CO$_{bg}$</th>
<th>S</th>
<th>Mean</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Linear Regression</strong></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Europe</td>
<td>0.37</td>
<td>111</td>
<td>2.1</td>
<td>3.6</td>
</tr>
<tr>
<td>North America</td>
<td>0.21</td>
<td>112</td>
<td>4.1</td>
<td>1.6</td>
</tr>
<tr>
<td>Fire</td>
<td>0.46</td>
<td>104</td>
<td>0.96</td>
<td>15.8</td>
</tr>
<tr>
<td>Europe + North America</td>
<td>0.55</td>
<td>107</td>
<td>2.3</td>
<td>5.1</td>
</tr>
<tr>
<td>All</td>
<td>0.72</td>
<td>97</td>
<td>1.0</td>
<td>21.0</td>
</tr>
<tr>
<td><strong>Multiple Linear Regression</strong></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Europe</td>
<td>0.82</td>
<td>96</td>
<td>2.0</td>
<td>7.2</td>
</tr>
<tr>
<td>North America</td>
<td>0.82</td>
<td>96</td>
<td>1.4</td>
<td>2.2</td>
</tr>
<tr>
<td>Fire</td>
<td>0.82</td>
<td>96</td>
<td>0.82</td>
<td>13.0</td>
</tr>
</tbody>
</table>

The squared Pearson correlation coefficient $r^2$, the derived background CO mixing ratio CO$_{bg}$, the slope S and the mean simulated mixing ratio. For the multiple linear regression, the mean simulated mixing ratio was multiplied with S in order to give the estimated contribution to the measured mixing ratio are reported.

The fact that enhanced black carbon occurred over periods when the wind direction was from the clean marine sector and the total aerosol number concentration was less than 500 cm$^{-3}$, which is indicative of unpolluted air masses. The covariance of CO and black carbon for August 13 (1000-1900 UTC) and August 18 (1900-2300 UTC) periods indicates a common source. Calculated black carbon to CO ratios for the two periods are of order 0.8 and 0.7 ng m$^{-3}$ ppbv$^{-1}$, respectively, which is intermediate between values measured for transatlantic marine air ($\sim$0.1 ng m$^{-3}$ ppbv$^{-1}$) and for polluted air ($\sim$4.0 ng m$^{-3}$ ppbv$^{-1}$) by Jennings et al. [1996].

The time series of ozone does not show strongly elevated values during the second half of August (Figure 9b), but the occasional positive correlation with CO indicates ozone production in the forest fire plume. However, as much of the transport occurred at low levels, this production may have been compensated by chemical destruction in the marine boundary layer.

4. Discussion

In our study we show that forest fire emissions from Canada caused pronounced haze layers and considerably enhanced CO concentrations over Europe during August 1998. The emissions were carried within the middle and lower troposphere over Greenland and the Atlantic in a mainly westerly flow and took $\sim$1 week

![Figure 9](image_url)

Figure 9. (a) Black carbon concentrations in ng/m$^3$ and (b) ozone mixing ratio in ppbv compared to carbon monoxide mixing ratios measured at Mace Head during August 1998.
to reach Europe. The fact that considerable amounts of forest fire aerosols and trace gases were detectable in the mid troposphere and the boundary layer even after a transport over such a long distance is related to the intense forest fire activity in Canada during 1998. It is reported that 10,838 fires burned in Canada in 1998, which corresponds to an area of \(4.7 \times 10^6\) ha of which \(>10^6\) ha were destroyed during the week from August 5-11, 1998, whereas the 10-year yearly average (1988–1997) amounts to 8937 fires and \(3 \times 10^6\) ha, respectively [Global Fire Monitoring Center (GMFC), http://www.uni-freiburg.de/fireglobe/ffn/country/ca/ca_6.htm, 1999]. These extreme conditions allowed us to unambiguously show that a range of trace substances emitted by the forest fires in Canada reached Europe. Although not shown in this study, it is highly probable that a less pronounced but still significant influence of the Canadian forest fires on trace substance budgets over Europe will be present during the summer months almost every year.

Furthermore, with the statistical analysis in section 3.2 we obtained a background CO concentration of 97 ppbv. This value is higher than one would normally expect during summertime in the Northern Hemisphere and implies that another unusual high CO source may not have been accounted for in the model simulations. Since Siberian forest fires can affect the total CO column over European parts of Russia [Yurganov et al., 1997], the elevated CO background documented in our study might be a result of intense forest fires in Russia. Approximately \(10 \times 10^6\) ha were destroyed by wildfires there [Kasischke et al., 1999]. This number is estimated from advanced very high resolution radiometer (AVHRR) satellite images, which show that 1998 was (like in Canada) an extremely severe fire year in Russia compared to other years in the 1990s. Boreal forest fires in Canada and Russia may therefore together strongly enhance the CO background of the Northern Hemisphere during extended periods of time and may exert also a nonnegligible influence on the radiative forcing of the Northern Hemisphere troposphere.

During the period studied, Canadian forest fire emissions had a much larger impact than North American anthropogenic emissions on the CO concentrations at Mace Head. An infrequent and relatively weak influence of North American anthropogenic emissions on measurement data at Mace Head was also reported by Ryall et al. [1998] for a longer period. In our study the relative contribution of North American anthropogenic emissions was found to be much greater at a level of 3000 m (see Figure 5), and it further increases with altitude (not shown). The reason for this is that anthropogenic emissions take place at lower latitudes than boreal forest fire emissions. Air masses from these lower latitudes regularly get entrained into warm conveyor belts which lift them to the upper troposphere [Stohl and Trickl, 1999], whereas air masses from higher latitudes, with lower potential temperature and a smaller chance of being carried over warm ocean waters, have a much lower probability of encountering warm conveyor belts and therefore stay longer in the lower troposphere [Stohl, 2001]. Deep convection, the other important process lifting surface emissions to the upper troposphere, is also much stronger at the lower latitudes. Therefore emissions from boreal forest fires in North America have a larger effect on the European lower troposphere than an equal amount of North American anthropogenic emissions. The period studied may be considered as typical in this respect.

The ozone sounding at Lindenberg (Figure 3) showed that ozone mixing ratios in the haze layer were \(20–25\) ppbv higher than in the adjacent layers. This ozone enhancement was likely caused by photochemical production from the forest fire emissions. If the FLEXPART result of \(60–80\) ppbv forest fire CO within this layer (see Plate 5a) is correct, this implies that up to 0.3 ozone molecules were produced per molecule CO emitted. This is significantly more than normally found in midlatitude forest fire plumes [Jacob et al., 1992; Mauzerall et al., 1996; Wotawa and Trainer, 2000] and is comparable to values typical for anthropogenic emission mixtures. This may be related to the more complete photochemical ozone production occurring over the very long distance this plume has traveled compared to the previous studies.

Our results show that if washout is ineffective, very pronounced haze layers in the free troposphere can be created by aerosols from forest fire emissions and can be transported over long distances. However, ineffective wet deposition is not the only requirement for a long-range transport of aerosols to occur. For this, the polluted air mass must be transported above the planetary boundary layer because otherwise deposition at the surface also removes the aerosols. As trajectory calculations (not shown) revealed, black carbon concentrations at the surface in Mace Head were moderately elevated only during those short periods of time when air masses traveled in the free troposphere and descended to the station only shortly prior to the arrival.

5. Conclusions

The conclusions of this study are as follows: Simulations with the particle dispersion model FLEXPART have shown that a haze lamina observed over Germany by four aerosol lidars on August 9 and 10, 1998 had its origin in large forest fires in Canada and traveled via Greenland to Europe. The haze lamina was so thick that meteorological observers reported cirrus cloud cover. Aerosols originating from boreal forest fires can thus have a strong impact on the radiative properties of the atmosphere on a hemispheric scale.

The FLEXPART simulation revealed that maps of the TOMS aerosol index, which are often used to trace the transport of forest fire plumes, strongly underestimate the true extension of the plumes. Elevated con-
centrations of nonsoluble trace gases (CO, O3, etc.) remain in the plumes long after aerosols have been removed by washout and thus the plumes have become invisible in the TOMS images. TOMS images can only prove the existence of plumes but not their nonexistence. Ozone concentrations within the haze layer were ~25 ppbv above the background, suggesting that substantial ozone production has occurred.

Three FLEXPART simulations of the transport of CO originating from forest fires and European and North American anthropogenic emissions were capable of reproducing the measured CO timeseries at Mace Head in August 1998 to a large extent, explaining 82% of the variance of the measurement data. At Mace Head, CO enhancements over the background during August 1998 were caused to 32% and 10% by European and North American anthropogenic CO emissions, respectively. At the same time 58% of the CO enhancement over the background can be traced back to the forest fire emissions from Canada. In addition, in summer 1998, even larger forest fires burned in Siberia. The CO budget of the Northern Hemisphere was thus at least as strongly affected by emissions from boreal forest fires as by anthropogenic sources.

The forest fire CO emission estimate of 4500 kg CO ha\(^{-1}\) of forest burned used in this and in previous studies was found to be compatible with the results of the FLEXPART simulation. From a comparison of the model simulation with measurement data at Mace Head, we derive a just slightly lower value of 3700 kg CO ha\(^{-1}\) burned. Given the range of uncertainty of forest fire CO emission estimates, which is between 3000 and 10,000 kg ha\(^{-1}\), this correspondence is remarkable. Our analysis thus supports an emission ratio close to 4000 kg CO ha\(^{-1}\) burned.

During most of the time black carbon concentrations measured at Mace Head in air masses influenced by the forest fires were close to their background due to washout, but two incidents of enhanced black carbon could be linked to the forest fires.

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