

**Biomass burning
smoke in Finland in
2006**

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Long-range transport of biomass burning smoke to Finland in 2006

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Abstract

Finland experienced extraordinary smoke episodes in 2006. The smoke was measured at the three SMEAR measurement network stations in Finland after it had been transported several hundreds of kilometers from burning areas in Eastern Europe. A trajectory method combining MODIS fire detections and HYSPLIT trajectories enabled us to separate the effect of biomass burning smoke from the measured concentrations and also study the changes in the smoke during its transport. The long-range transported smoke included at least NO_x , SO_2 , CO_2 , CO, black carbon and fine aerosol particles, peaking at 100 to 200 nm size. The most reliable smoke markers were CO and SO_2 , especially when combined with particle data, for which black carbon or the condensation sink are very effective parameters separating the smoke episodes from no-smoke episodes. Signs of fresh secondary particles was observed based on the particle number size distribution data. While transported from south to north, particles grew in size, even after transport of tens of hours and several hundreds of kilometres. No new aerosol particle formation events were observed at the measurement sites during the smoke periods.

1 Introduction

Biomass burning emissions have strong effect on air quality and climate being one of the key factors determining atmospheric composition of, e.g. greenhouse gases (CO_2 , methane), NO_x (precursor of ozone), CO and elemental and organic particulate matter (Langmann et al., 2009; Andreae and Merlet, 2001). Exposure to smoke is harmful for human health and can be dangerous for sensitive people like children and asthmatics (WHO, 2006).

Released greenhouse gases and soot particles (black carbon, BC) influence the radiative balance of the Earth due to absorption of radiation (positive radiative effect, Bond et al., 2013; Myhre, 2009). Aerosol particles exert an overall negative radiative

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effect (cooling) and change the properties and formation conditions of clouds (IPCC, 2007; Koch and Del Genio, 2010; Feingold et al., 2005). Soot deposition decreases ice and snow albedo (IPCC, 2007; Hansen and Nazarenko, 2004) and boreal forest fires increase the albedo of the burned area (Lyons et al., 2008). It is not clear whether boreal forest fires have an overall positive or negative influence on the radiative balance of the Earth (IPCC, 2007; Randerson et al., 2006).

Pollution due to biomass burning is not only a local problem. The smoke particles can travel even around the world (Damoah et al., 2004). Jaffe et al. (2004) demonstrated that long range transport of Siberian biomass burning emissions can impact ozone in North America, and Forster et al. (2001) describe the transport of emissions of Canadian fires to Europe. Interannual variability of CO at high northern latitudes has been observed to correlate with the area burned by forest fires in North America and Russia (Wotawa et al., 2001) and Simmonds et al. (2005) suggest that also the anomalies in tropospheric CO₂ concentration growth rates in 1996–2003 were related to large scale biomass burning events.

Compared to savanna and rain forest fire smoke plumes, emissions from boreal forest fires have been clearly less presented in the literature (Koppmann et al., 2005). As climate change is increasing the possibility of forest fires in boreal regions (Soja et al., 2007; Groisman et al., 2007), their importance as source of trace gases and particles in the atmosphere and long-range transported pollution episodes, should not be underrated. Especially smoke transported from boreal regions to the Arctic may have a substantial role in the warming of these regions (Law and Stohl, 2007; Liu et al., 2011). Black carbon contributes significantly to snow melt and may be a major factor in the near past Arctic ice retreat (Ramanathan and Carmichael, 2008) and has been estimated to be one of the main factors of Arctic climate warming (Shindell and Faluvegi, 2009).

Formation of secondary particles in fresh biomass burning smoke has recently been observed in a chamber (Hennigan et al., 2012), and majority of the biomass burning aerosol has been observed not to consist of black carbon (Kondo et al., 2011). Studies

of fine particle number size distributions gives us information of the origin of particulate mass. Understanding the properties of aged smoke plumes hundreds or thousands of kilometres from the fire areas helps to determine the climatic importance of biomass burning at the northern latitudes.

5 Exceptionally intensive wild fires occurred in large areas of Eastern Europe in spring and summer 2006. They were mainly caused by agricultural burnings, followed by an exceptionally warm and dry summer. Smoke was transported thousands of kilometres, and unusually high air pollution levels were measured even in Spitsbergen (Stohl et al., 2007; Treffeisen et al., 2007; Myhre et al., 2007) and the United Kingdom (Witham and
10 Manning, 2007). Modelled spatial distribution of the smoke plume can be found from the report of EMEP (2008).

In Finland the background values of the concentrations of fine particles and other anthropogenic pollution are low compared to e.g. central Europe (Asmi et al., 2011; Laakso et al., 2003). High concentrations are caused by long range transport of, in
15 particular, material emitted from wildfires or agricultural burnings in Eastern Europe (Niemi et al., 2009). From 25 April to 7 May and during August 2006, the meteorological situation was favourable for the transport of material from the large fire areas to Finland causing periods with exceptionally high concentrations of smoke. The smoke episodes in Helsinki have also been reported by Saarnio et al. (2010); Saarikoski et al.
20 (2007) and Bozier et al. (2007), and in Vironlahti (south-eastern Finland) by Anttila et al. (2008). Arola et al. (2007) studied the effect of the spring episode on aerosol optical properties over Finland and Estonia. Transported smoke included also radioactive material from the Chernobyl nuclear power plant accident fallout areas (Paatero et al., 2009), but their health effects were insignificant compared to the health effects
25 of the fine particles and their chemical components, that have been associated with increased mortality (Hänninen et al., 2008).

In this study, we used extensive data sets obtained from measurements at three field stations to characterize the properties of the air masses that had passed over extensive biomass burning areas, focusing on trace gas concentrations and aerosol size

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distributions. Continuous measurements together with satellite and air mass trajectory data, allowed us to observe changes in air mass properties as a function of time since it had passed over the burning area. This in turn allowed us to evaluate which processes affect the smoke properties during its transportation of several hundreds of kilometres from the source.

In this paper, Moderate Resolution Imaging Spectroradiometer (MODIS) thermal anomalies data was used together with HYSPLIT 4 trajectories (HYbrid Single-Particle Lagrangian Integrated Trajectory) to determine whether the air arriving at the measurement stations had passed over active fire areas. Trace gas concentrations and particle number size distributions in smoke plumes were studied, together with basic meteorological parameters. Results were compared to similar measurements during smoke-free reference periods.

2 Materials and methods

We combined data measured from three field stations with extensive atmospheric property measurements with calculated trajectories and satellite imaging to analyse two periods of biomass burning emissions transported over Finland. The smoke periods were compared to reference periods to estimate the relative impact of the smoke.

2.1 Observations at SMEAR-stations

The University of Helsinki has three atmospheric measurement stations (SMEAR, Station for Measuring Forest Ecosystem – Atmosphere Relations) in Finland: SMEAR I is a remote subarctic background station in Värriö, Lapland (Hari et al., 1994; Ruuskanen et al., 2003), SMEAR II is a remote background station in Hyytiälä, southern Finland (Kulmala et al., 2001b; Hari and Kulmala, 2005) and SMEAR III (operated together with the Finnish Meteorological Institute, FMI) is an urban background station in Helsinki (Järvi et al., 2009). The locations of the stations are presented in Fig. 1.

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Concentrations of several trace gases are measured continuously at all three stations. During the smoke episodes in 2006, O₃, NO and NO_x were measured at all three stations, SO₂ in Värriö and Hyytiälä, and CO and CO₂ in Hyytiälä. Particle number size distributions (from 3 to 1000 nm), as well as a suite of meteorological parameters, were measured at all three stations. Visibility was measured only in Hyytiälä.

2.1.1 SMEAR I – Värriö

The SMEAR I measurement station (69° 46' N, 29°35' E) is located in the Värriö strict nature reserve in eastern Lapland, less than 10 km from the border of Russia. There is practically no local sources of pollution. The distance from the nearest small road to the station is approximately 8 km and from the nearest major road about 100 km. There are no towns or industry nearby. The nearest major pollution sources are Montchegorsk located 150 km to the east and Nikel located 190 km to the north. The measurements in Värriö are made from different heights in a 15 m high tower located on top of a hill at 390 m a.s.l.. The measurements have been made continuously since 1992.

The gas concentrations used in this study were measured in the tower at a height of 9 m. The data are reported as hourly means. There was a measurement gap in NO and NO_x measurements from mid-May to mid-July in 2006. There was also a measurement gap in the middle of the spring smoke period: from 26 to 30 April. SO₂ measurements started on 21 April and there was a gap from mid-June to end of July.

2.1.2 SMEAR II – Hyytiälä

The SMEAR II station is located in a rather homogenous Scots pine (*Pinus sylvestris*) stand on a flat terrain at the Hyytiälä Forestry Field Station of the University of Helsinki (61° 51' N, 24°17' E, 181 m a.s.l.) 220 km north-west from Helsinki. The largest city near the SMEAR II station is Tampere, located about 60 km south-west from the measurement site and having about 200 000 inhabitants.

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Profiles of concentrations of several trace gases, temperature and wind speed, the properties of solar and thermal radiation of the stand, and the fluxes between the canopy and atmosphere are measured in a 73 m high mast. Measurements have been made continuously since 1996.

Data used in this study has been measured at a height of 67.2 m above the mast base. Precipitation was measured above the forest at 18 m height. The mast measurements are reported as half-hour means. Black carbon was measured by an aethalometer (Virkkula et al., 2011) and wavelength of 880 nm was used for this study.

2.1.3 SMEAR III – Helsinki

The SMEAR III measurement site is located in Kumpula, Helsinki (60° 10' N, 24° 57' E), 5 km north-east from downtown. The surrounding area of Kumpula is typical urban background. There is a major highway nearby (less than 200 m) south-east to the measurement site. There is a 31-m-high measurement tower on a hill, 26 m a.s.l.

The gas measurements are made at a height of 4 m above the mast base, and temperature, wind and radiation measurements at a height of 31 m. The data are reported as half-hour means. No measurements were available in the beginning of June 2006.

2.1.4 Data processing

The data were processed according to normal data processing routines performed at the stations, including calibrations and removal of erroneous results. For trace gas concentrations, we used half-hour average concentration values. The size distribution of aerosol particles is affected by several source and sink processes, with varying strengths of the respective sinks and sources. Therefore, it is useful to inspect several size ranges. Classically, atmospheric particle size distributions are divided in four size ranges: the nucleation mode, the Aitken mode, the accumulation mode and the coarse mode. Here we applied the size ranges used by Dal Maso et al. (2008): the nucleation mode with diameters from 3 to 25 nm, the Aitken mode from 25 to 90 nm and the

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accumulation mode from 90 nm to 1000 nm (the upper limit of DMPS measurements). Coarse mode ($> 1 \mu\text{m}$) particles were not measured for this study.

In addition to the number concentration, the atmospheric aerosol can be characterized by the average size of the particles and for example by its total surface area.

5 The surface area is of interest because it characterizes the interaction potential of the aerosol with condensing gas-phase species; in our study we therefore do not investigate the surface area but the “condensation sink” CS, which characterizes the maximum rate of condensation of surrounding vapour to particle surfaces (Kulmala et al., 2001a).

2.2 Air mass trajectories and fire locations

10 To determine whether the measurements had been made in an air mass that had passed over biomass burning areas, a combination of air mass trajectories and satellite data was used.

HYSPLIT 4 backward trajectories (Draxler and Hess, 1998) were calculated for an arrival height of 100 m, for hourly arrival times, at each of the three stations. Backward trajectories were determined with 1-h steps, until -96 h.

15 MODIS thermal anomalies data MOD14A1 collection 005 from Terra satellite were used to determine the location of fires (https://lpdaac.usgs.gov/products/modis_products_table/mod14a1). Spatial resolution of the daily data is $1 \text{ km} \times 1 \text{ km}$. Fire detection is based on temperature anomalies in cloud-free situations (Giglio, 2005).

20 An example of MODIS fire detections along an air mass trajectory, which arrived in Hyytiälä on 2 May 2006 at 18:00 LT, is presented in Fig. 1. There were numerous active fires along the four-day-back trajectory and signs of smoke in the measurements were to be expected.

25 To estimate how strongly the air mass had been affected by biomass burning emissions, we developed an indicator, called the “fire sum”. The fire sum (Eq. 1) describes the fire emissions by the number of active $1 \text{ km} \times 1 \text{ km}$ fire pixels along the four day back

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trajectory, with an exponential decay of fire products in time:

$$\text{firesum}(t) = \sum_{t=-96 \times 60}^0 \frac{N_f(t)}{A(t)} \times \exp\left(-\frac{t_0 - t}{\tau}\right), \quad (1)$$

where τ is a “life time” of the artificial fire products and t is back time along the trajectory. The distance between two hourly trajectory points were divided into 60 min. For every one-minute back trajectory point we defined the number of active MODIS fire detections below that air mass (N_f). The inaccuracy of the trajectories was estimated to be 10 % of the travelling distance (Draxler and Hess, 1998), and the number of fire pixels was normalized with that radius ($A = \pi r^2$). Trajectory height was not taken into account. Fires were expected to be intense enough to influence the whole vertical column of tropospheric air. Different formulas of the fire sum were tested, but Eq. (2) was selected, since it best expressed the features in the measured concentrations in our case.

By using back trajectories and fire maps we also estimated the time t that had passed since the air mass had last been over an active fire. By combining this information with measured trace gas and aerosol concentration, we could obtain a rough estimate of the timescale of the loss processes occurring during transport to the measurement site.

During transport, concentrations of emitted trace gases and aerosols will gradually decrease due to mixing with surrounding air masses and also several loss processes. For gases, these loss processes include deposition (both wet and dry) and chemical transformation. Chemical transformations for the gases considered in this study mainly include oxidation by hydroxyl radicals (CO and SO₂) and photolysis reactions.

For aerosols, in addition to deposition, losses are induced by coagulation (for aerosol number) and for specific size ranges also growth (by condensation) and shrinkage (by evaporation) can be considered as loss processes. Coagulation is generally a major loss process for aerosol particles in the smaller size ranges (nucleation and Aitken mode), while evaporation is not generally considered to be important under atmospheric conditions. Coagulation losses increase with increasing particle concentrations; in smoke plumes the atmospheric lifetime of ultrafine aerosol particles is of the

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order of minutes to hours. Condensation growth causes particle losses mainly in the smallest size ranges.

To estimate the atmospheric lifetime of fire-emitted trace gases and aerosols, we assumed that the added concentration of compound C decays by first-order processes, so that after time t its concentration will be

$$\Delta[C](t) = \Delta[C_0] \times \exp\left(-\frac{t}{\tau}\right), \quad (2)$$

where $\Delta[C] = [C] - [C_{\text{ref}}]$ is the concentration change measured at one of the stations. $\Delta[C_0] = [C_0] - [C_{\text{ref}}]$ and τ are parameters, corresponding to the concentration increase immediately after the air has left the fire area, and the atmospheric lifetime (e-folding time for the concentration increase). Using measured concentrations over the whole of the smoke episode, we applied a least-square fitting to the concentration increases as a function of time since the fire. This gave us an estimate of the average concentration increase at the fire site, as well as the lifetime of the added gases and aerosols.

One should note that this approach is very approximate, as the initial concentrations just after the air mass leaves the fire area are likely to vary with fire intensity and the type of burning biomass; also, the loss process rates are not likely to remain constant over the time of transport. Also, especially for aerosols, the two parameters τ and C_0 are not independent, as for example coagulation increases with higher C_0 , thus causing a lower τ . The values obtained from these fittings should therefore be considered only as a guide to the timescales of the loss of the fire signal in the trace gas and aerosol concentrations.

2.2.1 Definition of the smoke episodes and reference periods

As described above, the episodes were defined based on the backward trajectories. The spring episode lasted almost continuously from 25 April to 7 May. Smoke came from large areas in Eastern Europe and Russia, as shown in Fig. 1. A period of 12 days before and 12 days after the smoke period (13–24 April and 9–20 May) were

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selected as reference. During this reference period the air came from the sector south-west to north, over Scandinavia, where there were no fires detected by MODIS. Air was clean because of several weather front passages and rain.

During the spring smoke episode in Hyytiälä we noticed 20 exceptionally high particle concentration plumes, lasting from half an hour to some hours. Plume times were defined from the particle number size distribution data. During the plumes, the wind was always from the sector between 114–152 degrees, which means from the south-east. According to the MODIS fire detections and HYSPLIT trajectories, we conclude that these plumes came from some very extensive fires at the eastern shore of the Gulf of Finland, less than 300 km from Hyytiälä.

During August several shorter episodes occurred at the two southern stations. The smoke originated mainly from intense fires at the eastern shore of the Gulf of Finland and the eastern border of Finland. None of the smoke plumes were transported to the Värriö station. Reference values were calculated for the August months in each year of 1996–2008, except 2006. However, CO measurements started in Hyytiälä in 2002 and visibility and black carbon were measured only from 2005 onwards. For Helsinki the trace gas data were available only for Augusts 2007 and 2008. DMPS data are available for Hyytiälä from 1996 and for Helsinki from 1997, but only data for August 2005, 2007 and 2008 were included, because of the change in the number of measurement bins. The smoke and reference periods are presented in Table 1.

During the episodes, wind was mainly from the south-east. That is also the most common direction of pollutants arriving to Hyytiälä (Hulkkonen et al., 2012). During the spring reference period air came from the non-fire areas, mainly from the south-west (Atlantic ocean and the western Europe), from where there normally is not so much pollution transport. Therefore, it is important to have the fire parameters (fire sum) to estimate the impact of fires to the pollution concentration. For the August episodes the reference period is calculated from several years Augusts, and the wind directions vary more. It has not been ruled out that there might have been some fires, too. However, no such episodes have been reported before, except 12–15 and 26–28 August 2002

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(Niemi et al., 2005). Usually the smoke episodes are identified by chemical analysis of the particulate matter or when some special exceedings are noted from the data. The advantage of the trajectory and satellite based method of smoke identification is that it can be used also in the absence of the chemical analysers and it is not dependent on visual estimations.

3 Results and discussion

Two of the most obvious changes in an air mass passing over a smoking fire are reduced visibility and elevated carbon monoxide concentration (Fig. 2). Before and after the smoke episode shown in Fig. 2, the air was clean and visibility reduction was due to rain. During the smoke period there was no rain and RH was below 64 %, so visibility was reduced by smoke particles. Visibility in Hyytiälä was reduced to 16 km on minimum as compared to visibilities of over 50 km in non-rainy situations.

3.1 Concentrations of trace gases and aerosol particles

The difference in concentrations of atmospheric constituents between the smoke periods and the reference periods was studied by investigating the difference in the average concentrations of the respective periods (Table 2) and the frequency of occurrence of varying levels of trace gas concentrations (Fig. 3, for Hyytiälä). The extent of the measured values during a period was divided into 50 bins and the number of measurements in each bin was calculated; the frequency distribution was then normalized to one.

Trace gas concentrations in Hyytiälä are generally much lower in summer than in spring (Lyubovtseva et al., 2005). However, the difference between the smoke episodes and the reference period in spring is clear for CO, SO₂, NO_x and O₃ concentrations at all three stations (when measurements available). The summer smoke episode was the most intensive in Helsinki and not so visible in Hyytiälä. The smoke did not travel to Värriö in August 2006.

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In Helsinki NO and NO_x concentrations were elevated, especially in August. In Värriö NO_x and O₃ concentrations were elevated, but SO₂ not. Accumulation mode particle concentrations increased at all three stations, and the shape of the particle size distribution was similar.

3.1.1 Nitrogen oxides

Nitrogen oxides (mainly NO, NO₂) are expected to be emitted during biomass burning, especially if the fire burns with an open flame (Lapina et al., 2008). Agricultural areas in Eastern Europe are also expected to have large nitrogen loads from fertilization. NO_x concentrations were clearly elevated during both smoke episodes at all three stations. During the spring episode, NO_x levels were from 1.4 to 2.1 times higher at all the stations, up to 11 ppb at Hyytiälä. The NO_x increase was 7.4 ppb in Helsinki, 1.6 ppb in Hyytiälä 0.5 ppb in Värriö. The smoky air was travelling from south to north and NO_x was removed by photochemical reactions, deposition and dispersion of the smoke plume.

The NO concentration was elevated in Helsinki and Värriö, but not in Hyytiälä. Due to short atmospheric life time, NO can not be transported for days from the fire areas to the measurement stations in Finland. However, NO can be formed in the plume by photochemical reactions from the other constituents of the smoke. There are local NO sources in Helsinki, like traffic emissions. In Värriö there are no local sources of NO and there were no peaks in the data, but the general level was elevated by 0.02 ppb.

NO and NO_x measurements in Värriö had measurement gaps from 26 to 30 April and from 15 May onwards, so there is approximately 30 % less data during both the episode and the reference period.

During the summer episode, the increase in nitrogen oxides was most clearly visible in Helsinki.

The increase in NO_x was proportional to the elevation of CO, which clearly indicates that the increase in NO_x originates from the biomass combustion (see Fig. 4a for Hyytiälä). During the spring episodes, the correlation between the NO_x and CO

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increases was $R^2 = 0.66$ ($P < 0.05$, as for all the given correlations), and a linear fit for the dependency $\Delta(\Delta\text{NO}_x)/\Delta(\Delta\text{CO})$ gave a slope of 0.033. Although NO_x and CO_2 are both associated with flaming fires, in our case they correlated less ($R^2 = 0.36$) than NO_x and CO .

NO_x lifetimes within fire plumes have been estimated to be of the order of hours (< 7 h, Mebust et al., 2011 and references therein). In our case, the time since the air masses had been in contact with fire areas were several tens of hours. Our observation shows that the NO_x signal due to forest fires can survive for a period significantly longer than these estimates. The NO_x increase was inversely proportional to the time since the last fire; assuming first-order losses for NO_x during travel (Eq. 2), we arrived at a e-folding lifetime estimate of 63 h for the NO_x signal during travel to the site. The increase and correlation was not seen in NO concentrations. This is probably due to the oxidation of combustion-generated NO to NO_2 during the transport. In support of this assumption, we found a low and negative correlation between the increase in NO and the increase in ozone, the latter being one of the oxidants participating in the $\text{NO} \rightarrow \text{NO}_2$ conversion.

3.1.2 Carbon monoxide and carbon dioxide

The burning of organic matter leads to the emission of carbon oxides: carbon dioxide and carbon monoxide. CO emissions are associated with incomplete burning and smouldering fire (see e.g. Lapina et al., 2008). According to satellite studies, CO concentrations are known to correlate well with fire areas and aerosol optical depth (Edwards et al., 2004).

Carbon oxide measurements were available only for the Hyytiälä station and there CO seemed to be a good tracer of smoke periods (Fig. 2). The difference in CO concentration levels was clear during both spring and summer episodes (Fig. 3). During the spring episode in Hyytiälä CO levels were elevated by almost 30 %, while in summer the increase was slightly larger, approximately 40 % (Table 2). This is consistent

with a more smouldering fire during the summer, as the NO_x concentrations were lower during the same time. The carbon monoxide increase during smoke plumes (see e.g. Fig. 2) was quite typical for transported biomass burning: similar increases have reported by e.g. (Honrath et al., 2004).

In Hyytiälä the CO_2 concentration was elevated by 1 % (3.6 ppm) during the spring episode and no increase was visible during the summer episode on the average. During spring the distribution of CO_2 measurements is narrow and the difference clearly visible in Fig. 3. During summer the biological activity widens the distribution and the smoke effect is engulfed by biological uptake of CO_2 .

The combustion efficiency describes how much of the total carbon is burned to CO_2 , and it can be presented as modified combustion efficiency $\text{CO}_2/(\text{CO}_2 + \text{CO})$ (Koppmann et al., 2005, and references therein). In our spring smoke measurements in Hyytiälä, the modified combustion efficiency was very close to 1, so CO_2 excess dominated the emitted carbonaceous fire products. Also the relation of $\Delta\text{CO}/\Delta\text{CO}_2$ emissions was small, 0.01 on average, when it typically ranges from 2 to 20 % (Koppmann et al., 2005). One has to note that the travel distance in our study was longer than in other studies, and more CO had been oxidised to CO_2 . The CO_2 signal had also been modified by e.g. biological uptake during the transport. CO_2 correlation with other parameters is also weaker than correlations with CO ($R^2 = 0.24$).

3.1.3 Ozone

During the spring episode, ozone concentrations were consistently higher by ca 30 %. The elevated NO_x levels in the proximity of the fires lead to ozone formation, which can still be seen after several hours of smoke plume transport (Alvarado and Prinn, 2009). Significantly increased ozone levels have been observed even in 13–15 days old smoke plumes (Wotawa and Trainer, 2000; Honrath et al., 2004). During the spring episode, ozone concentrations increased by 10.8 ppb in Helsinki and 14.3 ppb in Hyytiälä. This is consistent with the situation of air travelling from south to north. After long distance from the sources, an increase of 13.7 ppb was measured in Värriö, slightly less than

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in Hyytiälä. However, even higher ΔO_3 values have been reported in the Arctic during the same pollution episode: ca 30 ppb in Spitsbergen and ca 40 ppb in Iceland (Stohl et al., 2007).

The elevation of the ozone levels was also correlated with the carbon monoxide elevation, as reported before e.g. by Honrath et al. (2004). In our cases the $\Delta O_3/\Delta CO$ slopes were 0.13 during the smoke episodes ($R^2 = 0.32$), and 0.11 during the reference period ($R^2 = 0.06$). These values are smaller than those reported by Honrath et al. (2004) (0.4 to 0.9, $R^2 = 0.4$ to 0.5), but in line with other results that have reported values below 0.2 (references therein). Stohl et al. (2007) observed very efficient ozone production of slopes of 0.34 to 0.53 in the same smoke plume further in the Arctic, resulting as record high ozone concentrations there.

Ozone and CO concentrations were quantities that quite clearly separated the smoke episodes from the background episodes (see Fig. 4c) and they were correlated with $R^2 = 0.35$. Figure 4c has similar feature as reported by Stohl et al. (2007) from Spitsbergen. Surprisingly, we also found that during the spring smoke episode, a very clear separation between the episode and non-episode observations could be found using CO₂ and ozone data: almost all smoke data points were in a region defined by $O_3 + 2.1 \cdot CO_2 > 850$, whereas in the reference period the data was in the region $O_3 + 2.1 \cdot CO_2 < 850$ (see Fig. 4e).

The ozone production has been noted to be highly dependent on the NO_x/CO ratio (McKeen et al., 2002), which here was 0.014 for the smoke period on average and 0.018 for the most extensive plumes in Hyytiälä, and resulted in ozone production of $\Delta O_3/\Delta CO = 0.29$, value typical to boreal and temperate regions (Jaffe and Wigder, 2012). The most extensive plumes had higher NO_x/CO ratios, i.e. more flaming combustion, but less ozone (see Fig. 4c), probably due to shorter travel distance. The ratio of O₃/CO stayed at 0.3 when comparing smoke episodes and reference period, but the ratio O₃/NO_x decreased from 42 during the reference period to 24 during the smoke episode on average and 18 during the most extensive plumes. In Helsinki the relation decreased from 4.0 during the reference period to 3.2 during the smoke episode.

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For the smoke periods, increasing CO₂ concentrations were accompanied by lower ozone concentrations; this is in contrast to the O₃–CO relation, which showed a positive correlation. We speculate that this could be caused by more efficient burning in the fires leading to lower ozone production, possibly via NO_x and hydrocarbon chemistry in the proximity of the fires. Due to the long transport time, details of this process are not observable in our data.

During the summer episode, ozone concentrations at the Hyytiälä station were still elevated by 6.6 ppb, but in Helsinki there was no change.

3.1.4 Sulphur dioxide

Soils and land biota are large reservoirs of sulphur (Seinfeld and Pandis, 1998) and during combustion of sulphur-containing matter, sulphur dioxide is emitted (see e.g. Lavoué and Stocks, 2011). For Hyytiälä, during both episodes, the sulphur dioxide was clearly the trace gas that exhibited the highest rise when compared to the reference period (Table 2). In spring, SO₂ was over four times higher than the reference average, while also in summer the rise was almost 60 %. However, at the more northern station in Värriö, no elevation was observed in the spring; this is most likely due to the short atmospheric lifetime of SO₂, which tends to get removed by oxidation (see also Sect. 3.2 for lifetime estimates). General levels of SO₂ at the remote Hyytiälä and Värriö stations are low and even during the high pollution episodes, average SO₂ concentration did not exceed 1 ppb (Table 2, Fig. 3), whereas the 99-percentile was 2.80 ppb.

3.1.5 Aerosol particle concentrations

Aerosol particles are a major pollutant emitted by biomass burning and also the main cause of many environmental effects such as visibility reduction and effects on human health. Mean particle concentrations are in general much higher in the urban Helsinki environment (on average 12 900 cm⁻³ during the spring reference period) compared to those at the more remote Hyytiälä or at the subarctic remote Värriö site (3390 and

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801 cm⁻³, respectively). The relative increase during the smoke episodes, however, is much larger at the more remote stations, where the total numbers increased by 40 % and 140 % on average for the Hyytiälä and Värriö stations, respectively. Particles were significantly larger at all of the three stations. In Helsinki the fine particles had a mean size of (61 ± 1) nm and in Hyytiälä and Värriö (111 ± 2) nm during both spring and summer episodes.

Accumulation mode dominated the particle size distributions, which is uncommon for the remote background stations, and associated with long range transport episodes. Only in Helsinki the number of the smallest particles in nucleation and Aitken mode was close to that of the accumulation mode. Particle number and size will be discussed more in detail in Sect. 3.3.

The increase in number and size transforms clearly into the condensation sink value calculated for the Hyytiälä station. It is higher by a factor of 2–4 for both episodes, making it an excellent marker for burning-emitted aerosol.

3.1.6 Black carbon

Black carbon measurements have been available in Hyytiälä since 2004 (Virkkula et al., 2007). Average concentration during the spring smoke episode was 1.01 µg m⁻³, reference values being four times smaller. Maximum concentrations were around 2.5 µg m⁻³. Concentration levels for the summer episodes were lower, 0.42 µg m⁻³ on average and around 1.5 µg m⁻³ on maximum. The measured values were extremely high compared to normal pollution levels in the remote environment of Hyytiälä (Hyvarinen et al., 2011).

3.1.7 Best indicators of fire

The differences between the smoke and the reference periods were analysed from scatter plots of the most important fire parameters versus each other (Fig. 4). Black carbon and carbon monoxide have been generally used as a marker of smoke periods, and

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they seem to correlate well in our study, $R^2 = 0.91$ during the smoke period (Fig. 4g, see also Fig. 2), and less during the reference period ($R^2 = 0.42$). Also condensation sink correlate better with CO during the episode ($R^2 = 0.78$) than during the reference period ($R^2 = 0.28$), but correlations with condensation sink and black carbon were good during both the episode ($R^2 = 0.78$) and reference period ($R^2 = 0.69$).

NO_x correlated well with all BC ($R^2 = 0.66$), CS ($R^2 = 0.68$) and CO ($R^2 = 0.66$). The correlation coefficient for NO_x and SO_2 was found to be $R^2 = 0.50$ and for CO and SO_2 $R^2 = 0.37$ for the whole spring period of study. O_3 and CO_2 correlated only slightly with the other studied parameters.

There is a clear separation between the concentrations of CS or BC during the reference period and the higher concentrations during smoke periods, whereas for most trace gases there is a less clear distinction. Also in CO concentrations, there are values overlapping with the smoke and reference periods, although most of the smoke values are high compared to the reference values.

We tried to find combinations of observations that would most clearly separate the smoke episodes from background, non-smoke observations. A markedly clear separation was found using CO_2 and O_3 for the variables (Fig. 4e). NO_x , SO_2 , CO and CS correlated much better during the episodes than during the reference period, but no such clear distinction was seen. The best correlation was found for the combination of $\text{NO}_x \times \text{CO}$ vs. CS ($R^2 = 0.73$), whereas the correlation during the reference period was $R^2 = 0.32$.

Suppression of nucleation mode particles and an increase in accumulation mode particle concentrations shows a bisected pattern in Fig. 4h, with high nucleation mode concentrations and low accumulation mode concentrations for clear sky conditions and vice versa for smoke episodes. When considering these parameters, the division between smoky and clean air is evident; the key for distinguishing between them is mainly in the accumulation mode particle concentration.

In the most extensive plumes in Hyytiälä, described in Sect. 2.2.1, all pollutant levels were increased even from the average episode values. The plumes had extremely high

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values of NO_x (4.34 ppb on average) and CO_2 (383.6 ppm on average). They also correlated well ($R^2 = 0.68$), better than during the episode on average ($R^2 = 0.30$). SO_2 concentrations were seven times higher than during the reference period (1.19 ppb on average) and they correlated the best with other fire parameters during the most intensive plumes of closer origin ($R^2 = 0.71$ with NO_x , $R^2 = 0.52$ with BC, $R^2 = 0.37$ with CO) than during the long range transport. This is reasonable, because the travelling time from the source areas is much shorter, some hours, compared to the smoke period in general (tens of hours).

To test the assumption that the elevated concentrations observed at the measurement stations were due to biomass burning, we investigated the relationship between the fire sum (Eq. 1) and the pollutants studied. Positive correlations were found between the fire sum and NO_x , SO_2 , CO_2 , CO and BC concentrations (Table 3). Consistent with this, there were negative correlations between the time when the air last was over an active fire and the same gases. The correlations between the pollutants and fire sum/time after fire indicate the fraction of measured concentration increases been caused by biomass burning emissions. In Eastern Europe there are several other sources of pollution, e.g. industry, but at least one fourth of variations in measured CO, CO_2 , NO_x and BC concentrations were caused by fire emissions ($R^2 = 0.27$, $R^2 = 0.27$ and $R^2 = 0.23$, respectively). Ozone concentrations did not directly correlate with the fire indicators. However, we have concluded that a large fraction of the increased concentrations of ozone was formed from the precursor gases emitted by the fires (like NO_x).

Particle number concentration correlated positively with the fire sum, especially for accumulation mode particles. Of particle parameters, condensation sink appeared to correlate best with the fire effects, with a correlation coefficient of 0.40. The latter indicates the removal of fire-induced aerosol particles. Our data shows a shift in the particle size distributions to larger sizes due to particle growth, which does not necessarily affect the total particle number. This will be further discussed in Sect. 3.3. Particle size did not reveal a correlation with the fire sum, but increased with increasing travelling

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time. The amount of precursors or condensing vapours did not seem to be the limiting factor of the particle size. Particle growth during the travel was also noted from the size distribution data (Sect. 3.3).

Fire sums were calculated for different characteristic life times of decay processes in Eq. (2). Life time of approximately 12 h revealed the best correlations with measured gas and particle concentrations, and is therefore presented in Table 3. Because the correlations are relatively good, we can conclude that the use of HYSPLIT 4 trajectories with MODIS fire detections works as an analysis tool to evaluate the effect of active fires on air mass properties down wind of the burning area. The fire sum appears to be a viable tool to describe the relative amount of “fire products” in the air.

3.2 Estimated emissions and life times

The relatively good correlations from backward trajectory analysis (Table 3) encouraged us to make an exponential fitting to the “back time” data, to get the first estimations of the emissions and atmospheric life times of the constituents under discussion (Eq. 2, Table 4). This method resulted in an atmospheric life time of 3.7 days for CO, around 2.6 days for NO_x and 33 h for SO₂. The estimated atmospheric life time of CO is much shorter than in the literature (30 to 90 days; Seinfeld and Pandis, 1998) but the estimates of NO_x and SO₂ compare favourably with values reported by Seinfeld and Pandis (1998): one day to a week for NO_x (depending on the NO/NO_x ratio) and 25 h for SO₂.

This method does not work for the behaviour of ozone because, as mentioned above, ozone is not directly correlated with other fire indicators. Its concentration is more likely to increase as a function of time (e.g. Alvarado and Prinn, 2009).

The atmospheric life time of particles in the accumulation mode size range was estimated with this method as around 2.5 days. In the literature somewhat longer life times are reported, i.e. around ten days for accumulation mode particles (Friedlander, 2000). Our method fails to estimate life times of nucleation mode particles because they are too short (from minutes to some hours, Friedlander, 2000) for them to travel

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from the expected sources (closest fires) to the measurement station. The exponential fit did neither succeed to estimate the life time of Aitken mode particles, although it is reported to be from several hours to a couple of days. The time air travelled from the nearest sources to our measurement stations was in the range of some tens of hours.

5 However, in the size distributions (Fig. 5) there were no clear Aitken modes visible in the smoke air either, except in the plumes from the nearest fires.

For the total particle concentration we got a life time estimate of more than four hours. Since almost two thirds of the total particle concentration consists of nucleation and Aitken mode particles, this supports our argumentation that the smallest particles
10 are very efficiently removed from the plume, probably by the larger particles.

Our study gives longer life time to black carbon than to any other constituent of study, 123 h. Atmospheric life time of BC is noted to be highly case sensitive and large variations in the models exist (Bond et al., 2013).

The average emissions were estimated by assuming an exponential decay since the
15 air left the fire area (Eq. 2). Estimated concentration increases immediately after the air has left the fire areas are presented in Table 4.

3.3 Particle number size distributions

The important advantage of the SMEAR station network is continuous particle size
20 distribution measurements at all three stations. We studied the difference between the size distributions during the clean and smoke periods by comparing the average number size distributions during the respective periods for these stations (Fig. 5a, c, d) as well as the volume size distributions at Hyytiälä (Fig. 5b). Differences between reference and smoke periods are clear, with higher particle concentrations and larger sizes during the smoke periods. The effect was smaller during the summer smoke episodes.

25 The shape of the aerosol number size distributions during smoke periods was similar at all three stations. The distribution peaked between 0.1 and 0.2 μm . The peak was the narrowest in Helsinki, broader in Hyytiälä and the broadest in Värriö. The relative amount of particles smaller than 50 nm was the largest in Helsinki, second largest in

Hyytiälä and the smallest in Värriö. The relative amount of particles smaller than 10 nm was the largest in Helsinki, smaller in Hyytiälä and minor in Värriö. Similar shapes of smoke aerosol number size distributions have been observed by Agus et al. (2008).

The particle volume distribution for Hyytiälä is shown to highlight, how the volume (mass) based measurements lose important information on particle size. It shows only the difference in particle total number, not the different size modes. More conclusions can be drawn from the particle origin based on the number size distribution data.

During the most intensive plumes (Sect. 2.2.1) the particle number size distribution revealed a very interesting pattern in Hyytiälä. There were two clear peaks: a nucleation mode (close to the Aitken mode size range) at around 20 nm with a concentration of 3500 particles cm⁻³ and an accumulation mode close to 200 nm with a concentration of 5700 particles cm⁻³. The peak in accumulation mode was similar to that observed during the smoke episode on average and the peak in nucleation mode was new. We concluded that the nucleation mode came from the fires only some hundreds of kilometres from the station and the larger mode consisted of aged fire particles, that had travelled many hundreds to thousands of kilometres to the station. This indicates secondary particle formation in a fresh biomass burning plume, and growth of the particles during the transport.

Our observations of the number size distribution data are in line with the assumption that we measured smoke originated from the same source at all three stations. They are also in line with the assumption that during their advection from the south northwards, smaller particles grew in size while their number was reduced by removal processes. The widening of the particle size distribution was caused by coagulation processes.

No new particle formation events were observed at the measurement sites during the smoke days. New particle formation has been observed to happen in a fresh smoke plume within the first hours after emission (Hennigan et al., 2012), but in the plumes older than this the number of grown particles most probably caused significant coagulation losses of the secondary particles. Other studies have shown that in polluted

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air masses, secondary aerosol formation and growth is possible only if the precursor vapour sources are large enough to overcome the high coagulation losses (Mönkkönen et al., 2005; Kulmala et al., 2005). This is for example the case in South Africa, where extensive new particle formation events are observed regularly even in the peak savanna fire period, most probably due to relatively high SO₂ background concentrations and clear sky conditions (Vakkari et al., 2011; V. Vakkari and L. Laakso, personal communication, 2012). In our measurements, only in Helsinki the number of particles smaller than 5 nm was large. This can be due to local emissions, but since the number of the smallest particles is remarkably higher during the smoke episodes than during the reference periods, it is also possible that there is secondary aerosol production in the smoke plume. Also the data from the most extensive plumes in Hyytiälä suggests new particle formation in a young smoke plume. However, DMPS are known for significant uncertainties at the smallest size classes (see Wiedensohler et al., 2012), so this may be a measurement artefact.

4 Conclusions

In this study MODIS fire detections were used together with HYSPLIT 4 backward trajectories to study episodes of biomass burning smoke over Finland. Combined with field measurements of several trace gases and aerosol particle number size distributions at three atmospheric measurement stations, we found the characteristic composition of the long-range transported smoke.

The smoke transported from Eastern Europe to Finland included at least NO_x, SO₂, CO₂, CO, black carbon and fine aerosol particles, peaking at 100 to 200 nm size. Differences between the smoke episodes and reference periods were clear. During the spring episode in Hyytiälä the BC and SO₂ levels were four times and NO_x levels two times as high as during the clear reference period. CO was increased by 26 % and number of fine particles by 41 %. Median particle size was two times higher as usual. Also ozone concentrations were increased during the smoke hours. However, ozone

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did not correlate with the developed fire parameters, most probably because of its nature as a secondary product.

We developed a satellite fire detection and trajectory based indicator, “fire sum” to estimate the effect of biomass burning emissions to measured pollution increases. The fire sum succeeded to detect fire periods and indicated that at least one fourth of the variations in measured NO_x, CO₂ and CO concentrations originated from fires detected by MODIS instrument onboard the Terra satellite. This is important, since there are several other sources along long range transport from fire areas in Eastern Europe to Finland. The method could be used for large data sets to determine the effect of biomass burning to air quality in Finland, or elsewhere.

The method was also successful in estimating the atmospheric life time for several compounds generated by the fires. Estimations of the characteristic life times of NO_x, SO₂, CO and accumulation and Aitken mode particles were approximately in line with values reported in the literature.

While transported from south to north, particles grew in size, even after transport of tens of hours and several hundreds of kilometres. The smallest particles were removed by larger particles. The emissions from the closest fires showed an interesting pattern in Hyytiälä, number size distribution having an additional peak in the 20 nm size. This indicates secondary particle formation in a fresh biomass burning plume, and particle growth during the transport. No new particle formation events were observed at the stations during the smoke episodes.

The total importance of biomass burning in the boreal regions has not been quantified so far. As large scale biomass burning is a common feature in the high northern latitudes and emitted particles and gases spread to large areas, their importance to global climate should not be underestimated. The Arctic is experiencing severe climate change within the next few centuries and its nature being especially sensitive to changes, understanding all factors affecting its climate should be of high priority. Our study indicates of high number of secondary particles formed in biomass burning

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plumes growing, spreading, travelling far away and having climatic importance as scattering sunlight and acting as cloud condensation nuclei.

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Table 1. Definition of the episodes and the reference periods.

spring episode	25.4.–7.5.2006, when fire sum > 0 at the station
spring reference	13.–24.4. and 9.–20.5.2006
summer episode	August 2006, when fire sum > 0 at the station
summer reference	August 1996–2005, 2007–2008, when data available

Table 2. Mean visibility, trace gas concentrations, particle concentrations and particle sizes at the three stations during the smoke episodes (epi) and reference periods (ref) in spring and summer 2006.

	Spring			Summer		
	epi	ref	ratio	epi	ref	ratio
Hyytiälä						
NO [ppb]	0.027	0.028	0.99	0.003	0.029	0.12
NO _x [ppb]	3.14	1.51	2.08	1.32	1.04	1.28
O ₃ [ppb]	60.6	46.3	1.31	37.8	31.2	1.21
SO ₂ [ppb]	0.86	0.21	4.15	0.26	0.17	1.55
H ₂ O [ppth]	4.52	5.22	0.87	10.80	11.95	0.90
CO ₂ [ppm]	381	377	1.01	367	367	1.00
CO [ppb]	223	177	1.26	164	115	1.43
BC [$\mu\text{g m}^{-3}$]	1.01	0.25	3.97	0.42	0.26	1.62
M_{tot} [cm^{-3}]	4790	3390	1.41	2150	2060	1.04
D_p [μm]	0.112	0.052	2.13	0.113	0.074	1.52
CS [s^{-1}]	0.0131	0.0041	3.17	0.0083	0.0035	2.38
Helsinki						
NO [ppb]	5.25	4.73	1.11	6.08	2.33	2.61
NO _x [ppb]	22.47	15.08	1.49	17.69	8.34	2.12
O ₃ [ppb]	38.7	27.9	1.39	23.96	24.13	0.99
M_{tot} [cm^{-3}]	16 500	12 900	1.28	9670	7210	1.34
D_p [μm]	0.060	0.033	1.81	0.061	0.044	1.41
CS [s^{-1}]	0.0271	0.0087	3.17	0.0164	0.0077	2.14
Värriö						
NO [ppb]	0.024	0.006	4.31	–	–	–
NO _x [ppb]	1.38	0.85	1.61	–	–	–
O ₃ [ppb]	53.4	39.7	1.34	–	–	–
SO ₂ [ppb]	0.42	0.48	0.89	–	–	–
M_{tot} [cm^{-3}]	1940	801	2.42	–	–	–
D_p [μm]	0.099	0.075	1.33	–	–	–
CS [s^{-1}]	0.0056	0.0015	3.64	–	–	–

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Table 3. Correlation coefficients for fire parameters and atmospheric composition in Hyytiälä in spring 2006. “Fire sum” is defined in Eq. (1) and t (latest fire) is the time as hours since the air was last over an active fire.

	fire sum	t (latest fire)
NO_x [ppb]	0.48	−0.30
SO_2 [ppb]	0.34	−0.48
CO_2 [ppm]	0.52	−0.40
CO [ppb]	0.52	−0.30
BC [$\mu\text{g m}^{-3}$]	0.48	−0.23
N_{tot} [cm^{-3}]	0.19	−0.30
N_{acc} [cm^{-3}]	0.30	−0.39
N_{Aitken} [cm^{-3}]	–	−0.21
N_{nucl} [cm^{-3}]	0.15	−0.20
D_p [m]	–	0.32
CS [s^{-1}]	0.40	−0.33

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Table 4. Estimations of emissions and atmospheric life times from Eq. (2) in Hyytiälä in spring 2006.

	$\Delta[C_0]$	τ [h]
NO _x	2 ppb	63
SO ₂	1 ppb	33
CO	66 ppb	88
BC	0.99 $\mu\text{g m}^{-3}$	123
N_{acc}	2872 cm^{-3}	59
N_{tot}	73 790 cm^{-3}	4

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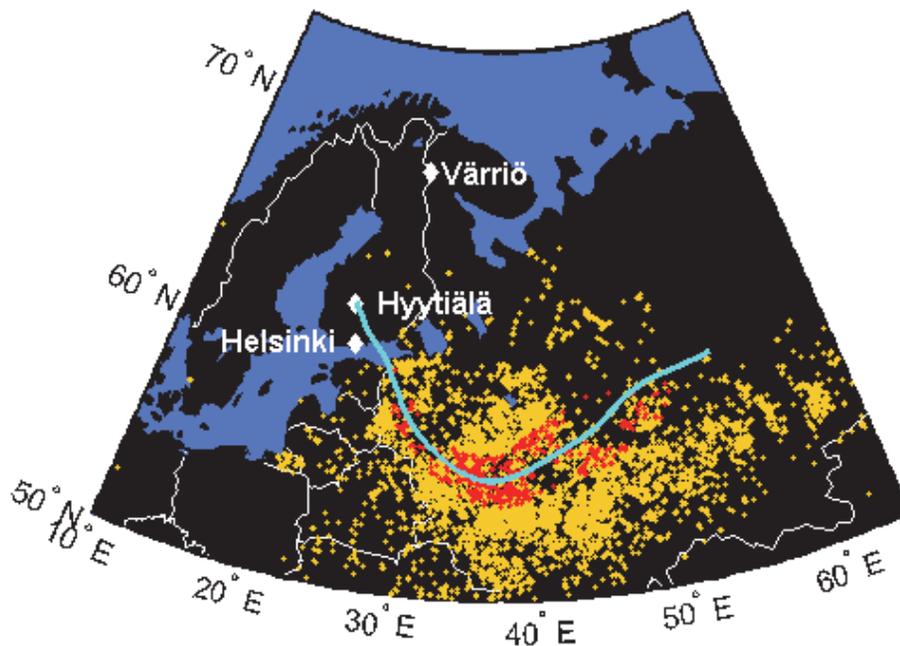


Fig. 1. Location of the measurement stations and a backward trajectory arriving in Hyytiälä on 2 May 2006 at 18:00 LT. MODIS fire detections during the four-day-back trajectory are indicated with the orange dots. Active fires along the trajectory are indicated in red.

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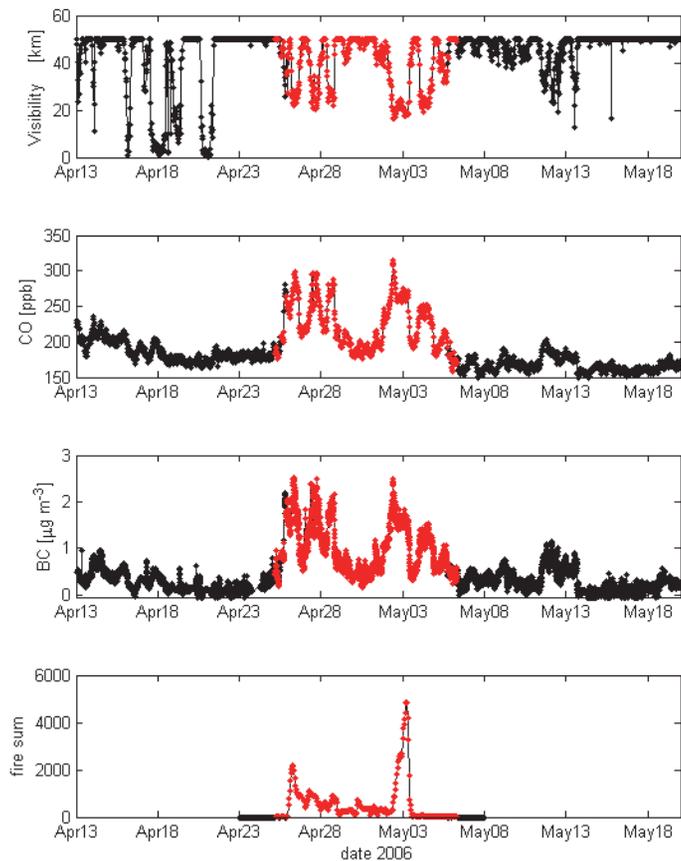


Fig. 2. Visibility, CO concentration, black carbon concentration and “fire sum” in Hyytiälä in spring 2006. Hours when the “fire sum” was positive are marked in red and indicate the spring smoke episode in Hyytiälä.

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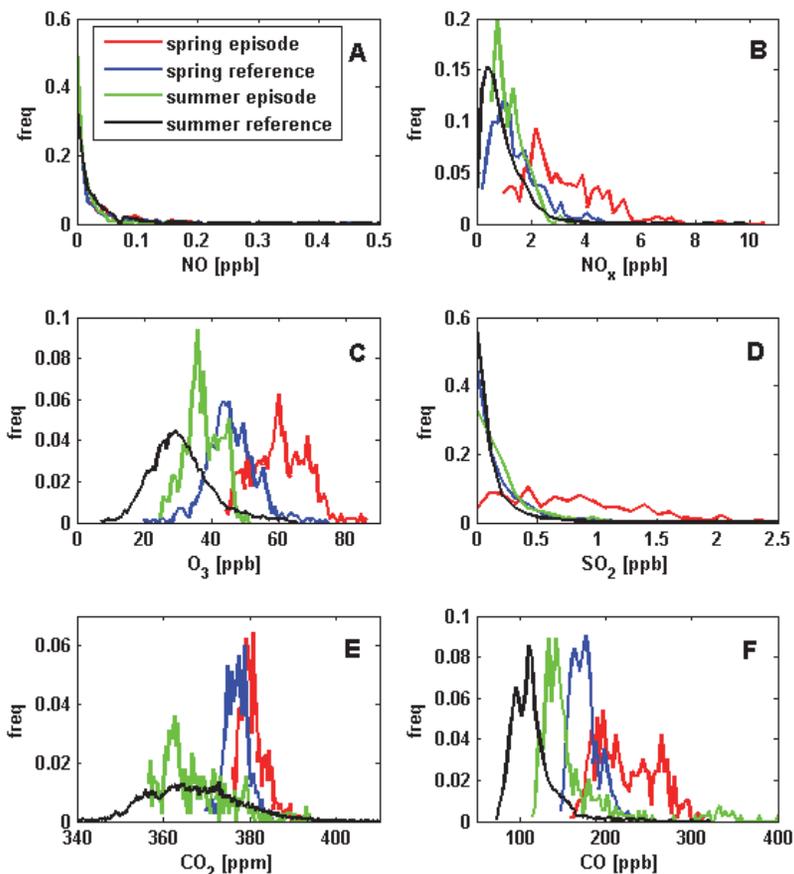


Fig. 3. Frequency distributions of trace gas concentrations in Hyytiälä for the spring and summer smoke episodes and reference periods. Frequency of occurrence of each of the concentrations is normalized to one.

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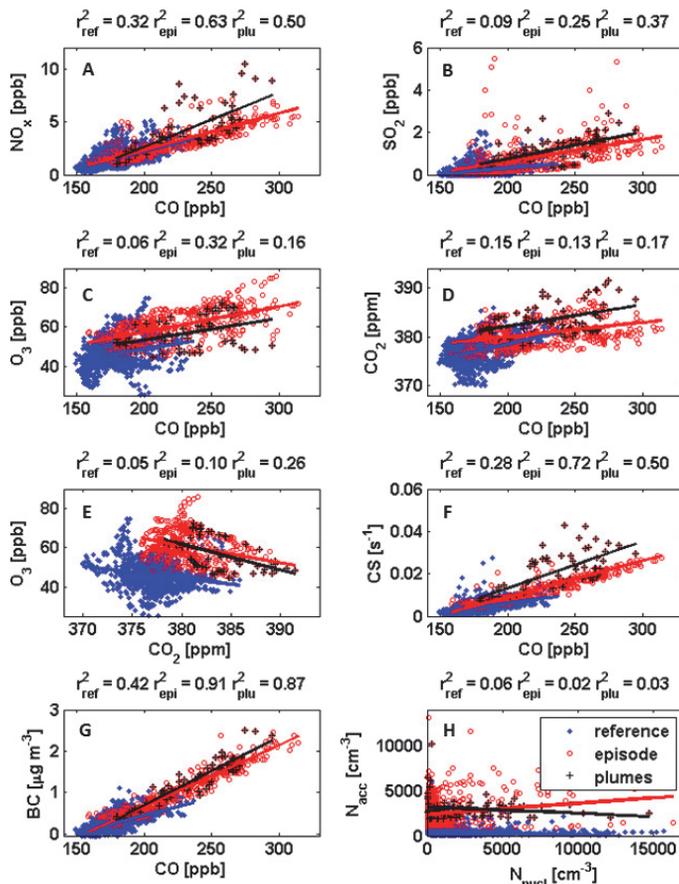


Fig. 4. Scatterplots of the most important fire parameters during the spring smoke episode (red) and the spring reference period (blue) in Hyytiälä. The most intensive plumes, as explained in the text, are marked with black crosses.

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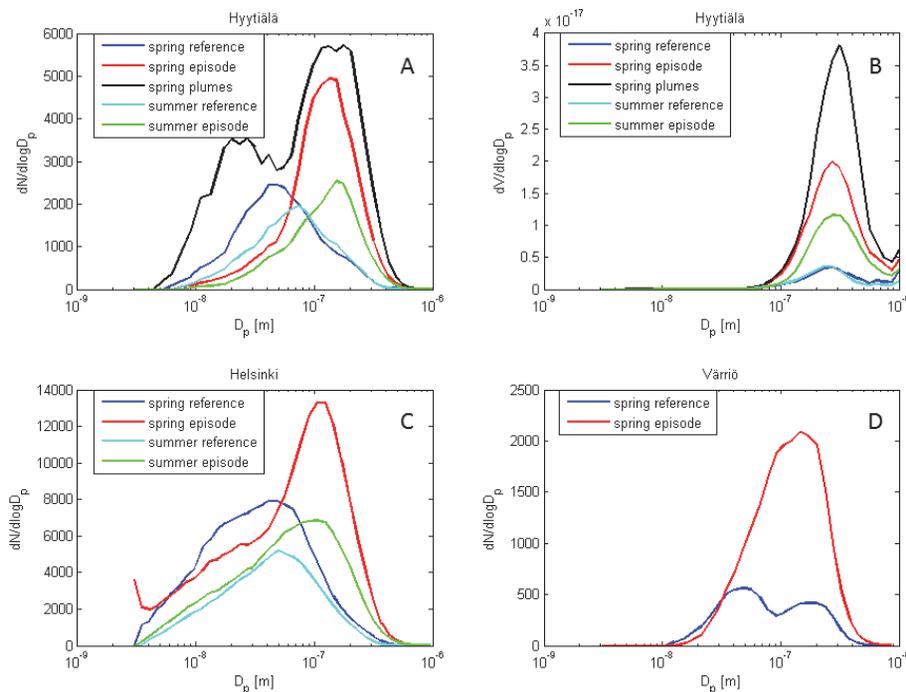


Fig. 5. Particle number size distributions at the three stations (**A**, **C**, **D**) and particle volume size distribution in Hyytiälä (**B**). Red represents spring smoke episodes, blue the reference period and green summer smoke episodes. The most intensive plumes observed during the spring smoke episode are shown in black and explained in more detail in the text.