

Biomass-burning smoke episodes in Finland from eastern European wildfires

Katri Leino^{1)*}, Laura Riuttanen^{1)*}, Tuomo Nieminen¹⁾²⁾, Miikka Dal Maso³⁾, Riikka Väänänen¹⁾, Toivo Pohja¹⁾, Petri Keronen¹⁾, Leena Järvi¹⁾, Pasi P. Aalto¹⁾, Aki Virkkula¹⁾⁴⁾, Veli-Matti Kerminen¹⁾, Tuukka Petäjä¹⁾ and Markku Kulmala¹⁾

¹⁾ Department of Physics, P.O. Box 48, FI-00014 University of Helsinki, Finland

²⁾ Helsinki Institute of Physics, P.O. Box 64, FI-00014 University of Helsinki, Finland

³⁾ Department of Physics, Tampere University of Technology, P.O. Box 692, FI-33101 Tampere, Finland

⁴⁾ Finnish Meteorological Institute, Research and Development, P.O. Box 503, FI-00101 Helsinki, Finland

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Biomass burning emissions from intensive wildfires in eastern Europe were observed in Finland in the spring of 2006 and in the late of summers 2006 and 2010. The smoke plumes were detected at three ground-measurement stations around Finland and in the lower troposphere after long-range transport from fire areas. The vertical extent of the smoke was estimated by flight measurements over southern Finland and the measurements were compared with CALIPSO satellite data from 29 July 2010. The history of the arriving air masses was analysed by using backward trajectories and MODIS fire detections. The smoke plumes had elevated concentrations of aerosol particle number, black carbon, CO, CO₂, SO₂, O₃ and NO_x, and the differences as compared with the background air were clear. The smoke was observed to be highly scattering, with a single-scattering albedo of 0.96 ± 0.01 . The median particle size was 60%–250% larger during the plume days than during July–August on average, and the growth of smoke particles was observed even after long-range transport of several hundreds of kilometres.

Introduction

Biomass burning is a major source of trace gases and aerosol particles into the global atmosphere (Andreae and Merlet 2001, Reid *et al.* 2005, van de Werf *et al.* 2010), affecting both air quality and climate (Randerson *et al.* 2006, Langmann *et al.* 2009, IPCC 2013, Mao *et al.* 2013, Reisen *et al.* 2013). Emissions from fire areas can be trans-

ported thousands of kilometres and are thereby not only a local problem (Damoah *et al.* 2004, Jaffe *et al.* 2004). Compared with clean or moderately-polluted air, air masses originating from biomass burning areas frequently have elevated concentrations of carbon monoxide (CO), nitrogen oxides (NO_x), volatile organic compounds (VOCs), ozone (O₃) and carbonaceous aerosol particles (e.g. Andreae and Merlet 2001).

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* These authors contributed equally

Numerous wildfires occur in Russian forests and peat bogs almost every summer due to high temperatures and drought (Konovalov *et al.* 2011). The mean annual area burned in this vast region increased during the past decades (Jaffe *et al.* 2004). Occasionally, air pollutants from the biomass burning taking place in Russia and elsewhere in eastern Europe reach Finland. Such smoke transport episodes took place, for example, during the spring and summer 2006 (Saarikoski *et al.* 2007, Anttila *et al.* 2008, Saarnio *et al.* 2010) and during the late summer 2010 (Portin *et al.* 2012, Mielonen *et al.* 2012, 2013). The 2006 smoke travelled far, as extraordinarily high pollutant concentrations were measured over the Arctic (Treffeisen *et al.* 2007, Stohl *et al.* 2007) and even in England (Witham and Manning 2007). The pollution from the summer 2010 fires was particularly severe in the Moscow region and its surroundings (e.g. Witte *et al.* 2011, Yurganov *et al.* 2011, Krol *et al.* 2013), causing remarkable local radiative cooling (Péré *et al.* 2014).

In this study, we investigated the character and both horizontal and vertical extent of long-range-transported smoke over Finland from the most severe large-scale biomass burning wild fires in eastern Europe during 2006 and 2010. In addition to comprehensive ground-level measurements at three continuously-operating stations, we made four airborne measurement flights over the eastern and southern Finland at the end of July 2010. These *in situ* measurements were complemented by air mass back trajectory calculations and satellite data. The scientific questions we aim to address here are the following: How did the most apparent smoke episodes differ from the background air in terms of pollutant concentrations and particle number size distributions? How far west and north could these episodes be detected in the surface air over Finland? Were the physical properties of the long-range transported smoke particles still changing when transported over Finland? How did the biomass burning plumes differ from each other? What is the benefit of combining data from very different types of measurements and data analysis approaches when investigating long-range-transported biomass burning smoke?

Methods and description of observations

In this study, we focused on the wildfire episodes observed in Finland during the spring of 2006 and the late summers of 2006 and 2010. The University of Helsinki has comprehensive long-term data sets of ground-level aerosol and ambient gas measurements at three field stations in Finland (Värriö SMEAR I, Hyytiälä SMEAR II and Helsinki SMEAR III). These long-term time series of pollutant concentrations from July–August period were used as a reference for the smoke episodes.

Several compounds were used in the analysis. Of trace gases, we concentrated on carbon dioxide (CO₂) and ozone (O₃), together with clearly observed and longer-lived combustion product carbon monoxide (CO) as well as nitrogen oxides (NO_x; NO + NO₂) and sulphur dioxide (SO₂). CO together with NO_x leads to photochemical formation of ozone close to and further away from the fire areas. The detected SO₂ emissions in Finland are usually from industry and from ship traffic, although the forest fires also generate SO₂ to the atmosphere. Concerning aerosol particles, we focused on the particle number concentration and size distribution, condensation sink (CS; *see* Kulmala *et al.* 2001a), and black carbon (BC) concentration. The mean values of the selected quantities on smoke days were compared with corresponding mean values during the July–August period averaged over several years.

The modified combustion efficiency (MCE) is defined as $MCE = \Delta CO_2 / (\Delta CO_2 + \Delta CO)$, where Δ was calculated as the difference between the measured concentration and corresponding monthly-minimum concentration. The MCE indicates the completeness of the burning process: as compared with flaming fires, smouldering fires produce more CO than CO₂, thus having higher MCE values (Ward and Radke 1993).

Measurements at research stations

The University of Helsinki has three SMEAR (Station for Measuring Forest Ecosystem–

Atmosphere Relations) stations for atmospheric measurements in Finland, equipped with versatile measurement capabilities (Table 1).

The SMEAR I (69°46'N, 29°35'E) research station is a remote subarctic background station in Värrö, Lapland (Hari *et al.* 1994, Ruuskanen *et*

Table 1. Instrumentation at the measurement stations. Differential Mobility Particle Sizer (DMPS) is presented more in detail in Aalto *et al.* 2001.

Station	Compound	Instrument	Model	Time
Värrö	SO ₂	UV-fluorescence analyser	Trace level model series 43, Thermo Fisher Scientific, USA	1992–Jan 2004
			API 100 AS, Teledyne Advanced Pollution Instrumentation, USA	Jan 2004–Mar 2007
	NO _x	Chemiluminescence gas analyser	Trace level model series 42, Thermo Fisher Scientific, USA, equipped with a molybdenum converter	Oct 1997–Jan 2004
			Trace level model series 42, Thermo Fisher Scientific, USA, equipped with a photolytic blue light converter by Droplet Measurement Technologies, USA	Apr 2007–
	O ₃	Ultraviolet light absorption analyser	API 200 AU Advanced Pollution Instrumentation, USA, equipped with a molybdenum converter	Jan 2004–Apr 2007
CO	Model series 49, Thermo Fisher Scientific, USA		1992–Jan 2004; Jan 2007–	
		API 400, Teledyne Advanced Pollution Instrumentation, USA	Jan 2004–Mar 2007	
		Horiba APMA 360, Horiba, Japan	April 2010–	
	Aerosol particle number size distributions	Infrared absorption analyser		1998–
Hyytiälä	SO ₂	UV-fluorescence analyser	Trace level model series 43, Thermo Fisher Scientific, USA	1997–
	NO, NO _x (NO + NO ₂)	Chemiluminescence gas analyser	Trace level model series 42, Thermo Fisher Scientific, USA, equipped with a molybdenum converter	1996–Feb 2007
			Trace level model series 42, Thermo Fisher Scientific, USA, equipped with a photolytic blue light converter by Droplet Measurement Technologies, USA	Mar 2007–
	O ₃	Ultraviolet light absorption analyser	Model series 49, Thermo Fisher Scientific, USA	1996–
	H ₂ O	Infrared light absorption analyser	URAS 4 H2O, Mannesmann Hartmann & Braun, Germany	1996–Sep 2011
	CO ₂		LI-840, LiCor Biosciences, USA	Sep 2011–
	CO	Infrared light absorption analyser	URAS 4 CO2, Mannesmann Hartmann & Braun, Germany	1995–Jun 2010
			LI-840, LiCor Biosciences, USA	Jun 2010–
			Horiba APMA 360, Horiba, Japan	2002–Mar 2010
		API 300EU, Teledyne Advanced Pollution Instrumentation, USA	Apr 2010–Jan 2011	
	Aerosol particle number size distributions	DMPS		1996–

continued

Table 1. Continued.

Station	Compound	Instrument	Model	Time
Helsinki	Black carbon (BC), aerosol absorption	aethalometer	Magee Scientific 7λ AE-31	2005–
	Aerosol scattering	nephelometer	TSI 3λ	2006–
	SO ₂	UV-fluorescence analyser	Trace level model series 43, Thermo Fisher Scientific, USA Horiba APSA 360, Horiba, Japan	Nov 2005–Sep 2006; Jun 2008– Sep 2006–Jun 2008
	NO _x	Chemiluminescence analyser	Trace level model series 42, Thermo Fisher Scientific, USA, equipped with a molybdenum converter	Nov 2005–Aug 2009
			API 200 AU Advanced Pollution Instrumentation, USA, equipped with a molybdenum converter	Aug 2009–
	O ₃	Ultraviolet light absorption analyser	Model series 49, Thermo Fisher Scientific, USA API 400, Teledyne Advanced Pollution Instrumentation, USA	Nov 2005–Dec 2009 Dec 2009–
	Aerosol particle number size distributions	DMPS		1998–

al. 2003). It is located in a nature park close to the Russian border. The local emissions of air pollutants in northern Fennoscandia (Lapland) are typically very small. The closest source of emissions is the mining industry in the Kola Peninsula. Trace gases (CO, SO₂, NO, NO_x, O₃), aerosol particles and meteorological quantities have been measured continuously at the station since 1992. A 15-m-high measurement tower is located on the top of a hill 390 m a.s.l. All data used in this study are from measurements at the 9-m level.

SMEAR II is a measurement station in the boreal forest site in Hyttiälä (Kulmala *et al.* 2001b, Hari and Kulmala 2005). The station is in southern Finland (61°51'N, 24°17'E) about 50 km to the northeast from the nearest city, Tampere, with about 200 000 inhabitants. The station is located 181 m a.s.l and the area has a rather homogenous Scots pine (*Pinus sylvestris*) base. The station is equipped with several aerosol and gas concentration measurement instruments and these have been operating continuously since 1996. The station has a 127-m-high mast equipped with instruments at different heights. In this study, we used trace gas concentrations of CO, NO, NO_x, H₂O, SO₂, O₃ and CO₂. We used gaseous data from the height of 16.8 m

above the mast base because it is the only level at which CO was measured during the summer 2010. We applied data as 30-minute averages. The black carbon concentration and aerosol optical properties, available only from this station, are measured with an aethalometer (measuring wavelengths of 370, 470, 520, 590, 660, 880 and 950 nm) and nephelometer (measuring wavelengths of 450, 550 and 700 nm, Virkkula *et al.* 2011). In reporting aerosol optical properties, the wavelength of 550 nm was used in this study.

The SMEAR III measurement site is located in Kumpula, Helsinki (60°10'N, 24°57'E), 4 km northeast from the center of Helsinki. The area is typical urban background and nearby, southeast from the station, is a busy highway. The station started operating in autumn 2004 and is run by both the Finnish Meteorological Institute and the University of Helsinki. The Kumpula site is located on a hill, 26 m a.s.l, and it has a 31-m-high measurement tower equipped with instrumentation at several heights (Järvi *et al.* 2009). Next to the tower there is an air-conditioned measurement container where the aerosol particle and trace gas measurement instrumentation is located. The concentrations of CO, SO₂, NO_x and O₃ were used in this study. Differential Mobility Particle Sizer measures the aerosol size range of

3–950 nm, while in two other stations it measures the size range of 3–1000 nm.

Airborne and satellite measurements

The measurement flights were made using a small aircraft, Cessna FR172F. The plane was modified from a four-seater to a two-seater, and the measurement rack with the instruments was located behind the seats of the pilot and the operator. The measurement flights were operated from and to Tampere-Pirkkala airport located in southern Finland. The flights were flown with the airspeed of around 130 km h⁻¹.

The airborne instrumentation in summer 2010 included an Ultrafine Condensation Particle Counter (UCPC, model TSI-3776) tuned and calibrated for 3 nm cut-off size, CO₂/H₂O gas analyser (Li-COR LI-840), pressure sensor, temperature/relative humidity sensor (Rotronic HygroClip-S) and GPS receiver (Garmin). The time resolution of the instruments was 1 Hz.

The UCPC, CO₂/H₂O gas analyser and GPS were located inside the cabin and set into the instrumentation rack. The measurement data from the instruments was collected with a computer located in the rack. This computer was linked to another laptop kept by the operator during the flights so that the operator would have a real-time opportunity to view the measurements. The sample-air inlet and the relative humidity/temperature probe were located ahead of the plane's right wing. The sample-air inlet was a modified and down-scaled model designed originally for measurement flights of a DC-8 aircraft (McNaughton *et al.* 2007). The inward-expanding form of the inlet and the flow-rate of 50 lpm make the flow laminar and isokinetic. The sample air is piped to the instruments via a stainless-steel pipe with inner diameter of 22 mm and a length of 4 m. The outflow exited via a venturi located in the main gear of the plane. The flow rate of this main flow was controlled manually with a flow meter (TSI 4000 series) and a control valve mounted in the operator's foot space. The electricity required by the measuring instruments was produced by rechargeable 12-volts (DC) batteries. For previous airborne measurements performed with the same aircraft

with partly similar installation *see* Schobesberger *et al.* (2013).

The total particle number and carbon dioxide concentrations from the flight measurements were studied. The CO₂ profile was corrected by the H₂O profile. The concentrations were normalized to a standard temperature of 288.15 K and a pressure of 1013.25 mbar; due to low altitudes these corrections were minor. Altitudes are given in meters above mean sea level, and times are UTC + 2 hours.

In order to study the vertical extent of the smoke plume, the airborne measurements were compared with vertical profiles from a polarization-sensitive, two-wavelength lidar (CALIOP; the Cloud-Aerosol Lidar with Orthogonal Polarization) onboard the CALIPSO satellite (the Cloud-Aerosol Lidar and Infrared Pathfinder Satellite Observation; Winker *et al.* 2009). It observes a narrow 70-m swath width, with 30-m vertical and 333-m horizontal resolution (below 8.5 km).

The MODIS (Moderate Resolution Imaging Spectroradiometer; Remer *et al.* 2005) Aqua aerosol optical depth data were used to study the horizontal extent of the smoke plumes. The Level 2 "Aerosol Optical Depth Land and Ocean" data Collection 051 with 10 km resolution was used in the study. The Aqua satellite passes over northeastern Europe after 10:00 local solar time.

Air mass trajectories and fire location analysis

We complemented the ground-based and flight measurements with backward air mass trajectory calculations and satellite-based active fire detections. These gave us information about the air mass transport routes and whether the measured air had passed over active fire areas. Air mass trajectories were calculated by HYSPLIT 4 (Hybrid Single Particle Lagrangian Integrated Trajectory Model; Draxler 1999) by using the GDAS (Global Data Assimilation System, <https://ready.arl.noaa.gov/gdas1.php>) data as input. Backward trajectories were calculated hourly for 100 m, 500 m and 3000 m arrival heights. Active fire detections and fire radiative

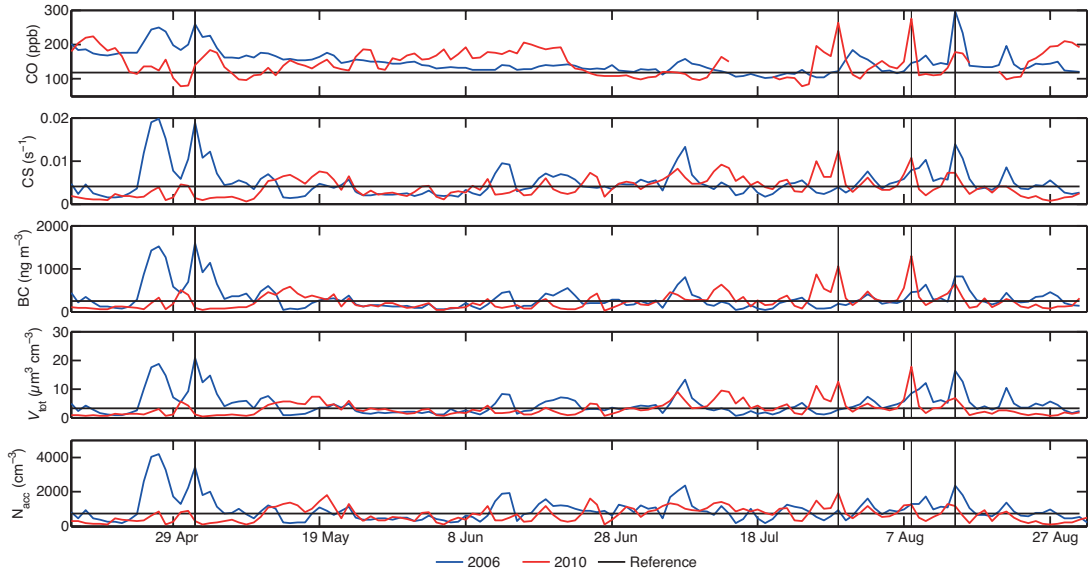


Fig. 1. Time series of daily mean values of carbon monoxide (CO), condensation sink (CS), black carbon (BC), total particle volume concentration (V_{tot}) and particle number concentration in accumulation size range (90–1000 nm, N_{acc}) at SMEAR II in Hyytiälä, Finland, in April–August 2006 (blue lines) and 2010 (red lines). Reference values are averages over July–August of several years (Table 2). Four vertical lines are drawn for the plume days 2 May 2006, 29 July 2010, 8 August 2010 and 14 August 2006.

power were obtained from the MODIS Aqua and Terra thermal anomalies data (MYD14A1 and MOD14A1), and they were compared visually with the air-mass trajectories. The relative source contribution fields were calculated based on the *in situ* measurements in Hyytiälä and HYSPLIT backward trajectories with a 100-m arrival height for 24 July–8 October 2010 (see Riuttanen *et al.* 2013).

Results and discussion

The most clearly-observed characteristics of the biomass burning smoke episodes at the ground level at SMEAR II station in Hyytiälä were elevated carbon monoxide (CO) and black carbon (BC) concentrations, as well as condensation sink (CS), total particle volume concentration and number concentration of particles in the accumulation mode size range (Fig. 1). Based on the concentration time series measured at the SMEAR II station in Hyytiälä and SMEAR III station in Helsinki (Tables 2 and 3), we selected four days for a more detailed investigation: 2 May 2006, 14 August 2006, 29 July 2010 and 8

August 2010. The air mass backward trajectory analysis confirms that during those days the air came from active fire areas (Fig. 2). In case of the SMEAR I station in Värriö, 3 May 2006 and 27 July 2010 were selected as the study days based on back trajectory analysis and increased SO_2 , NO_x and aerosol particle number concentrations and size distributions (Table 4).

The Hyytiälä and Helsinki stations had, unfortunately, data gaps on 8 August 2010: the particle number size distribution data from Hyytiälä were available only after the mid-day (67% of that day's data missing), and the particle number size distribution, CO and SO_2 data from Helsinki were available only between 7:00 and 14:00 and after 18:00 (58% of data missing).

General characteristics of the biomass burning episodes

On 2 May 2006, the observed smoke came from large and varying fire areas five hundred to several thousands of kilometres away. High BC and total particle number concentrations were observed in Hyytiälä, being 6.8 and 2.6 times

higher than those during the reference period, respectively (Table 2). The maximum particle number concentration was $22\,900\text{ cm}^{-3}$. A strong correlation between BC and CO concentrations was found (linear regression analysis: $r^2 = 0.88$). The SO_2 , NO_x and O_3 concentrations were 9.7, 3.2 and 2.1 times the corresponding concentrations during the reference period. The correlations between the CO_2 and CO concentrations ($r^2 = 0.37$) as well as between the O_3 and CO concentrations ($r^2 = 0.22$) were weak.

On 14 August 2006, the observed smoke came from smouldering fires on the eastern coast of the Gulf of Finland about 300 km from Hyytiälä, with a high CO concentration and the highest correlation between BC and CO concentrations ($r^2 = 0.92$). The smoke had less CO_2 than the other episodes and the correlation between CO_2 and CO was the lowest of the studied episodes ($r^2 = 0.11$). The O_3 concentration was 10% higher than that during the reference period and

the correlation between the O_3 and CO concentrations was clear ($r^2 = 0.50$). The SO_2 and NO_x concentrations were 62% and 92% higher than the corresponding concentrations during the reference period.

On 29 July 2010, the observed smoke came from intensive fire areas more than 800 km to the southeast from Hyytiälä. The MODIS Terra observed a radiatively-weak fire point on the eastern shore of the Gulf of Finland about 300 km from Hyytiälä. The correlation between the BC and CO concentrations was the weakest of the studied smoke days ($r^2 = 0.46$) and also the correlations between the CO_2 and CO concentrations ($r^2 = 0.18$) and between the O_3 and CO concentrations ($r^2 = 0.24$) were weak. The SO_2 concentration was the highest of the studied days, on average 930% higher than during the reference period. The O_3 concentration increased by 81%, while NO_x decreased by 42%, as compared with those of the reference period.

Table 2. Mean trace gas and total particle concentrations (N_{tot}), median particle sizes (D_p) and mean condensation sinks (CS) at the SMEAR II station in Hyytiälä during two smoke days in summer 2010 (27 July 2010 and 8 August 2010) and 2006 (2 May 2006 and 14 August 2006) and several-year July–August mean values as reference. Years included into the reference period are stated in the last column.

Hyytiälä	2 May 2006	14 Aug. 2006	29 June 2010	8 Aug. 2010	Ref	Ref period
CO (ppb)	259	298	262	276	118	2002–2010
CO_2 (ppm)	381	371	396	405	372	1996–2010
SO_2 (ppb)	1.26	0.21	1.34	0.35	0.13	1997–2010
O_3 (ppb)	65.3	34.0	55.7	42.0	30.8	1996–2010
NO (ppb)	0.01	0.01	0.02	0.02	0.02	1996–2010
NO_x (ppb)	3.05	1.82	0.41	0.55	0.95	1996–2010
H_2O (ppth)	4.7	13.3	16.5	18.8	12.5	1996–2010
BC (ng m^{-3})	1626	839	1054	1270	240	2005–2010
D_p (nm)	129	146	117	152	73	1996–2010
N_{tot} (cm^{-3})	5558	2983	3496	1806	2139	1996–2010
CS (s^{-1})	0.019	0.014	0.012	0.011	0.004	1996–2010

Table 3. The same as in Table 2, but for the SMEAR III station in Helsinki.

Helsinki	2 May 2006	14 Aug. 2006	29 June 2010	8 Aug. 2010	Ref	Ref period
CO (ppb)	–	–	251	433	170	2007–2010
SO_2 (ppb)	–	–	1.03	0.86	0.62	2007–2010
O_3 (ppb)	44.2	26.6	45.9	41.3	26.1	2006–2010
NO (ppb)	4.44	2.56	1.00	0.52	2.44	2006–2010
NO_x (ppb)	22.62	8.66	9.84	7.87	9.37	2006–2010
D_p (nm)	74	68	67	70	21	1998–2010
N_{tot} (cm^{-3})	18079	8990	6999	6041	9627	1998–2010
CS (s^{-1})	0.039	0.018	0.014	0.018	0.007	2006–2010

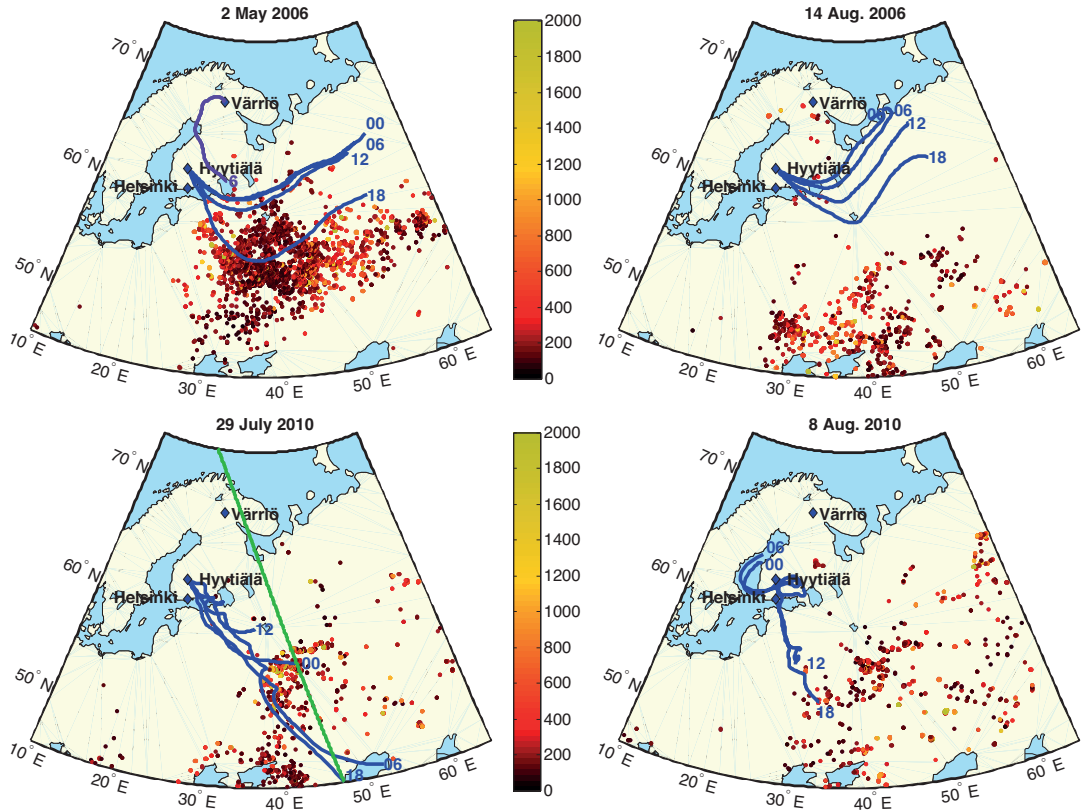


Fig. 2. Four-day-back trajectories arriving at Hyytiälä on 2 May 2006, 14 August 2006, 29 July 2010 and 8 August 2010 at 00:00, 06:00, 12:00 and 18:00 local time (EET = UTC +2). Backward trajectory arriving at Värriö on 5 May 2006 at 06:00 is marked on the first map. MODIS fire detections during the four days are marked on the maps, the colour indicating fire radiative power (W m^{-2}). CALIPSO flight on 29 July 2010 is also marked in the figure (green line).

On 8 August 2010, the afternoon smoke was transported to Finland from fire areas more than 800 km away from Hyytiälä, with fewer but larger particles (median diameter 152 nm), compared with the reference period (73 nm). Before the noon, high measured CO_2 concentrations

were indicative of flaming fires, probably those with high radiative signal on the eastern coast of the Gulf of Finland. The correlation between the BC and CO concentrations was clear ($r^2 = 0.63$), between the CO_2 and CO concentrations even clearer ($r^2 = 0.75$), whereas the correlation

Table 4. The same as in Table 2 but for the SMEAR I station in Värriö during two smoke days (3 May 2006 and 27 July 2010) and several-year July–August mean values as a reference.

Värriö	3 May 2006	27 July 2010	Ref	Ref period
NO (ppb)	0.00	0.03	0.01	1997–2010
NO _x (ppb)	2.06	0.49	0.18	1997–2010
SO ₂ (ppb)	0.68	–	0.35	1991–2010
O ₃ (ppb)	65.7	20.1	25.8	1991–2010
N _{tot} (cm ⁻³)	3004	1174	1232	1998–2010
D _p (nm)	161	134	51	1998–2010
CS (s ⁻¹)	0.0136	0.0043	0.0020	1998–2010

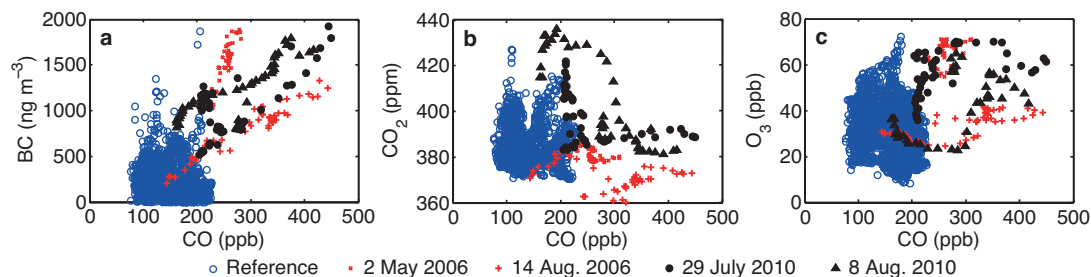


Fig. 3. Relations between black carbon (BC), carbon dioxide (CO_2), ozone (O_3) and carbon monoxide (CO) measured in Hyytiälä in spring and summer 2006 and summer 2010. The results are presented as 30-min averages. The coefficients of determination (r^2) are: (a) ref: no correlation, 2 May 2006: 0.88, 14 August 2006: 0.92, 29 July 2010: 0.46 and 8 August 2010: 0.63; (b) ref: no correlation, 2 May 2006: 0.37, 14 August 2006: 0.11, 29 July 2010: 0.18 and 8 August 2010: 0.75; (c) ref: no correlation, 2 May 2006: 0.22, 14 August 2006: 0.50, 29 July 2010: 0.24 and 8 August 2010: 0.31. The number of the data points for the reference period ranged from 1545 to 2708 and for the smoke days from 43 to 48, $p < 0.01$ for existing correlations.

between the O_3 and CO concentrations was moderate ($r^2 = 0.31$).

During the reference period (including time series 1 June 2010–15 July 2010 and 15 August 2010–21 August 2010), no correlations between the BC, CO_2 , O_3 and CO concentrations was found in Hyytiälä, in contrast to the smoke days when significant correlations of variable strength were found (Fig. 3). BC is a well-known biomass burning tracer, and its positive correlations with CO, CO_2 and O_3 indicate clearly that the elevated pollutant concentrations were caused dominantly by biomass burning. However, as there were large differences in the correlations, we conclude that the composition of the smoke varied between and within the plume days, indicating differences in the sources, burning conditions and chemical transformations taking place during the atmospheric transportation of the plumes. Differences between the 2006 episodes have also been reported by Saarikoski *et al.* (2007) and Saarnio *et al.* (2010), who studied the chemical composition of the smoke aerosols. They concluded, based on potassium and elementary carbon (EC) contents, that the spring 2006 episode originated mainly from flaming fires, whereas the August 2006 episode originated from smouldering fires. This is consistent with our results, as the modified combustion efficiency (MCE) was high on 2 May 2006 (0.993) and lower on 14 August 2006 (0.986). During the 2010 episodes, the MCE was 0.991 on 29 July 2010 and 0.992 on 8 August 2010. These

values are high, which may indicate that other sources of CO_2 during the transport distract the applicability of the MCE in long-range transported smoke plumes.

It should be noted that the applicability of many of the trace gases produced by wildfires to investigating the long-range transport of fire smoke is limited by the atmospheric lifetime of these trace gases and the potential presence of other important sources for them. For example, the atmospheric lifetimes of both NO_x and SO_2 are short, typically less than two days (Seinfeld and Pandis 1998), in addition to which they are emitted in large quantities by traffic and several types of industrial activities. The lifetime of O_3 varies from a few days in summer to several weeks in winter. However, being produced continually during the atmospheric transportation, O_3 cannot be considered an ideal tracer for fire pollution. The atmospheric lifetime of CO is of the order of 30 to 90 days, making it very suitable for investigating the long-range transport from wild fires.

Horizontal extent of biomass burning smoke in Finland based on ground-based observations

The SMEAR stations are located in different parts of Finland, several hundreds of kilometres apart from each other. As compared with those in Hyytiälä (SMEAR II), the average concentra-

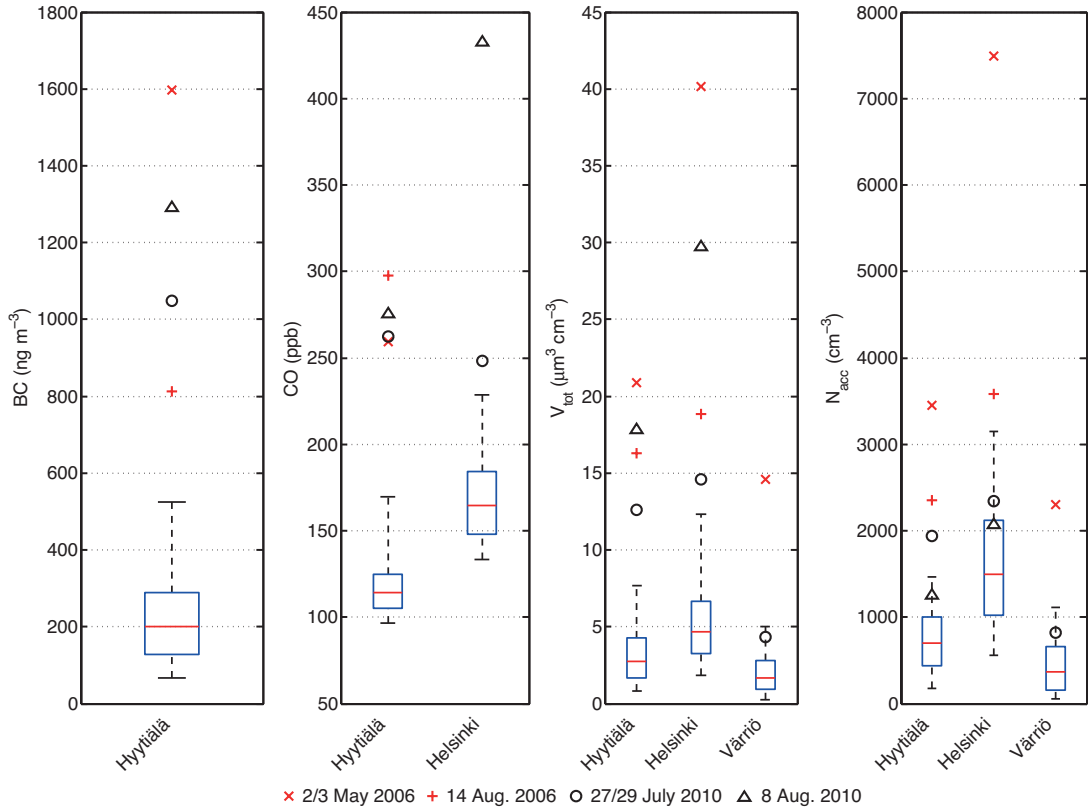


Fig. 4. Boxplots of black carbon (BC), carbon monoxide (CO), total particle volume concentration (V_{tot}) and particle number concentration in accumulation mode size range (N_{acc} , 90–1000 nm) from the measurements at three SMEAR stations during July–August over several years, when data were available. In each box, the red line is the median; the top and bottom of the box are the 25th and 75th percentiles, respectively; the whiskers are the 5th and 95th percentiles; and blue and red circles and crosses are the daily mean values for the plume days.

tions of air pollutants are higher in the urban background air in Helsinki (SMEAR III) and much smaller at the northern remote Värriö (SMEAR I) station.

Two of the four smoke episodes in 2006 and 2010 were observed at all three stations but at slightly different times. At all the sites, high CS values were measured during the episodes (Tables 2–4). The median particle sizes were 67–74 nm in Helsinki, 117–152 in Hyttiälä and 134–161 nm in Värriö, and these values are 60%–250% greater than those during the reference period. The CO and SO₂ concentrations were increased as compared with mean levels in Helsinki, with much variability in NO, NO_x and O₃ concentrations. The concentrations of NO_x, O₃ and SO₂ were exceptionally high in Värriö on 3 May 2006.

For the four smoke days, the daily-mean concentrations of BC, CO and total particle volume in Hyttiälä and Helsinki were clearly over the 95th percentile calculated for the reference period of July–August for several years (Fig. 4). Regarding particle number in the accumulation mode size range, only the 2006 episodes were over the 95th percentile. In Värriö, the daily mean concentrations on 3 May 2006 were clearly above the 95th percentile, whereas the episode on 27 July 2010 did not stand out from the reference data that clearly, yet the daily-mean values were above the 75th percentile. We conclude that the long-range transported smoke episodes were among the highest air pollution cases in the history of summer measurements at these three stations.

The average particle number size distributions (Fig. 5) showed that most particles are typically

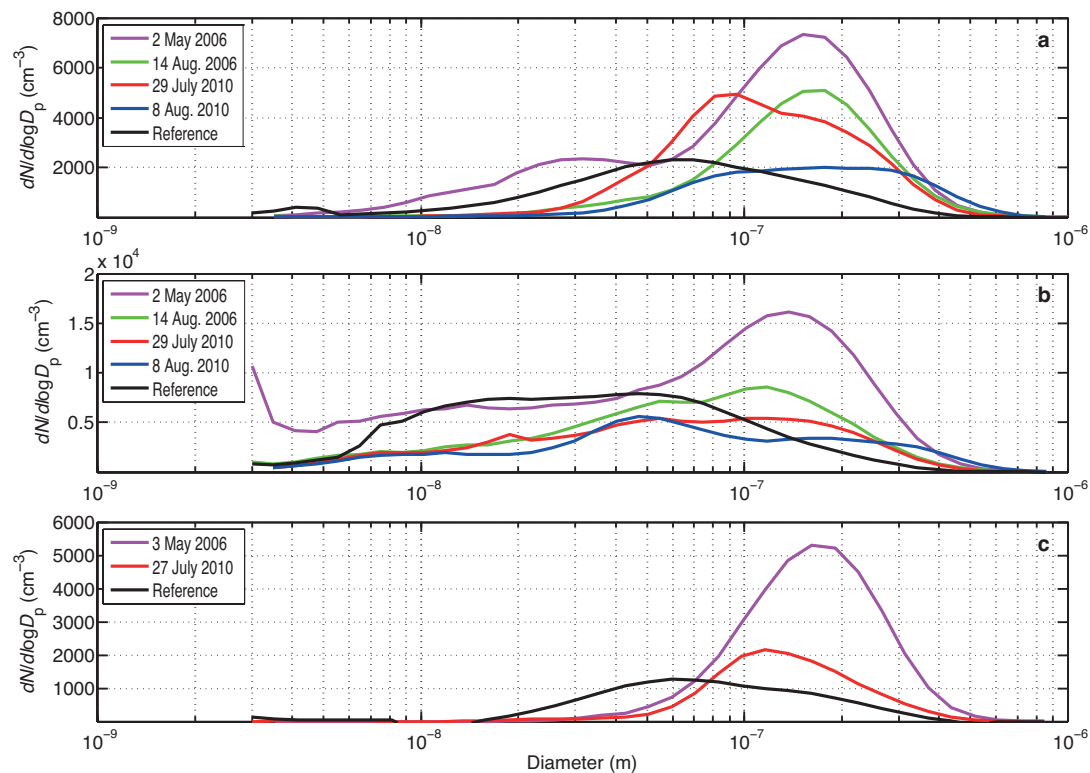


Fig. 5. Particle median size distributions measured at three SMEAR stations in Finland: (a) Hyytiälä, (b) Helsinki and (c) Väriö. Blue and red lines represent the plume days and the black line size distributions during July–August of several years.

in the Aitken mode size range (25–90 nm) at all three stations. During the smoke episodes, the particle number size distribution peaked in the accumulation mode size range (90–1000 nm), and the numbers of nucleation (< 25 nm) and Aitken mode particles were clearly lower than average.

Aerosol optical properties measured at Hyytiälä revealed that the smoke scattered strongly solar radiation. The scattering coefficients were 116 ± 31 (mean \pm SD) on 29 July 2010 and 206 ± 12 on 8 August 2010, and these values are high as compared with the long-term summertime values of 17 ± 13 (table 2 in Virkkula *et al.* 2011). The absorption coefficients were also high (5.2 ± 1.2 on 29 July 2010 and 7.2 ± 0.8 on 8 August 2010) when compared with the long-term summertime values (1.4 ± 1.2 , Virkkula *et al.* 2011). During both days, the smoke had high single-scattering albedos (0.96 ± 0.002 on 29 July 2010 and 0.97 ± 0.005 on 8 August 2010) compared with long-term summertime values (0.91 ± 0.05 , Virkkula

et al. 2011). The high single scattering albedos on both days, combined with clearly higher-than-average BC concentrations, is indicative of strong secondary aerosol formation associated with long-range transport in these plumes. Also the Ångström exponent of scattering was high, 1.73–2.04, indicating the dominance of small aerosol particles. The maximum value (2.05) of this exponent was obtained on 29 July 2010, the day with the smallest median size of aerosol particles. The Ångström exponent of absorption was also high, 1.3–1.9, indicating a high fraction of organic compounds in aerosol particles. The 2010 smoke plumes were observed to be highly scattering also around the Moscow region, close to their actual sources, with single scattering albedos of 0.95–0.96 (Péré *et al.* 2014).

The horizontal extent of plumes can be studied by using satellite-based aerosol optical depth measurements (Fig. 6). The MODIS instrument detected increased aerosol loadings at 550 nm

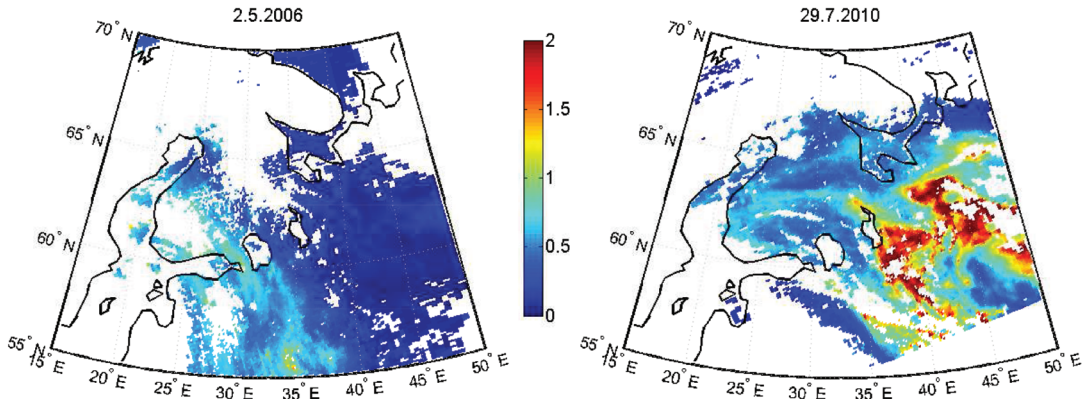


Fig. 6. MODIS Aqua aerosol optical depth for the northeastern Europe on 2 May 2006 and 29 July 2010.

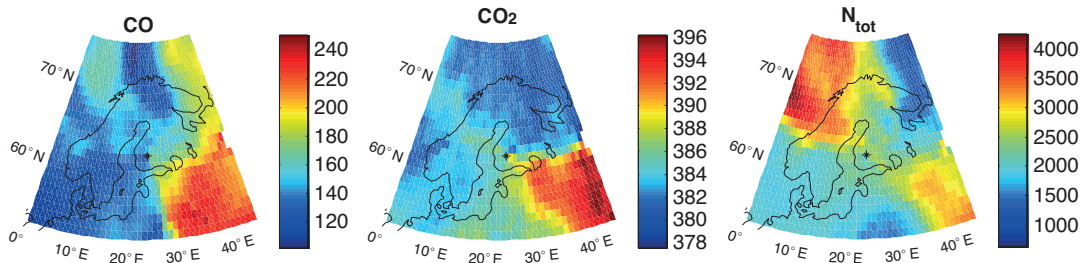


Fig. 7. Relative source contribution fields of CO, CO₂ and total fine particle number. Colours represent measured concentration in Hyytiälä according to HYSPLIT backward air mass trajectories that arrived at the measurement station during the measurement hour on 1 June–31 August 2010.

wavelength over a long band extending from SE to NW on 2 May 2006. Optically thick aerosol layers were observed on 29 July 2010 SE from Finland, with only minor parts of them reaching Finland. Although the smoke covered large areas, it was horizontally very inhomogeneous.

The relative source contribution fields based on air mass back trajectories calculated for the summer 2010 showed clear differences between the smoky air and surrounding air masses (Fig. 7). The CO and CO₂ concentrations were clearly higher in air masses coming from SE as compared with those in air masses coming from other transport directions. The same applied to SO₂, O₃, and NO_x concentrations (not shown). The relative source contribution fields of fine particles in the accumulation mode size range showed very similar patterns to the corresponding fields of CO, CO₂ and SO₂, confirming that these pollutants came from the same source areas. Particles smaller than 90 nm (nucleation and Aitken mode size range) came from

the opposite direction, namely they were clean Arctic air masses, a feature typical for Hyytiälä (Sogacheva *et al.* 2005). Therefore, two separate source fields for total particle concentration can be seen in Fig. 7. For NO_x the relative source contribution field was not as uniform, indicating that local emissions and chemistry played a significant role in addition to long-range transport. Also satellite detections see NO_x production over the fire areas, together with CO and aerosol production, but only CO and aerosols are long-range transported (*see* Mielonen *et al.* 2012).

The synoptic analysis (Fig. 8) revealed that on 2 May 2006 there was a blocking high over the northeastern Europe, and this situation lasted for two weeks. Smoke from the biomass burning area was directly transported all the way to southern and northern Finland, and even to Spitsbergen in the Arctic (Stohl *et al.* 2006, Treffeisen *et al.* 2007). On 29 July 2010 and 8 August 2010, the smoke came to Finland in a warm sector of a low-pressure system (*see* also

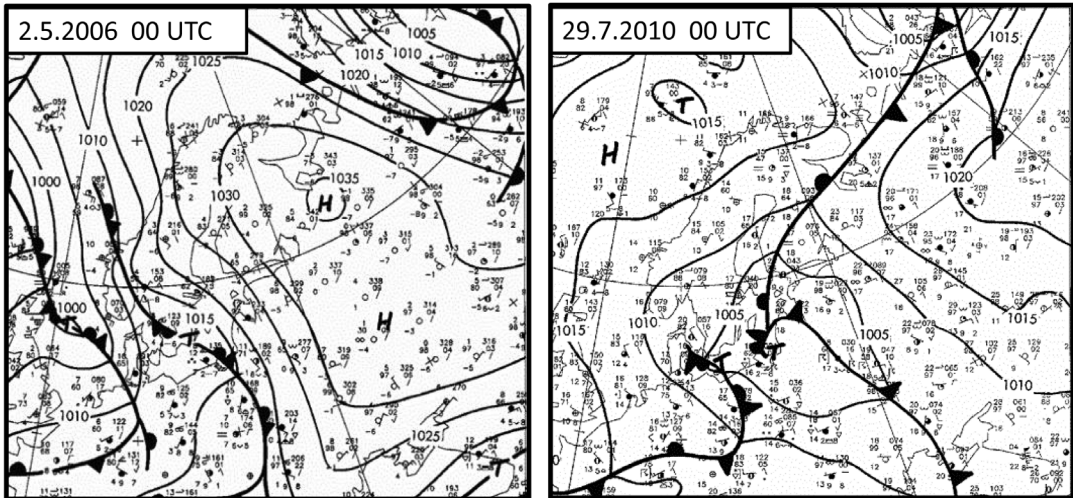


Fig. 8. Synoptic analysis for the northeastern Europe on 2 May 2006 and 29 July 2010 at 00 UTC. Figure adapted from Deutsche Wetterdienst (reproduced with permission from the copyright owner).

Witte *et al.* 2011). Smoke episodes lasted for less than one day and smoke transport was dependent on the dynamics of the low-pressure system.

The spring 2006 episode provided us an exceptional case to investigate aerosol dynamics in an already aged fire plume. The smoke was detected first at the southern measurement stations in Helsinki and Hyytiälä, and then a couple of days later at the northern Värriö station (Fig. 2, *see* also Fig. 8). The growth of particles during their transport over Finland was apparent (Fig. 9). The median particle diameter was 67 nm on 30 April in Helsinki, reached 93 nm on 1 May in Hyytiälä, and 117 nm on 5 May in Värriö, whereas the corresponding median diameters were 21, 73 and 51 nm during the reference period. The peak diameter of the size distribution increased from about 100 nm to 200 nm during the air mass transport over Finland.

Vertical extent of biomass burning smoke in southern Finland

At the end of July 2010, we operated four flights to eastern and central Finland to measure total particle concentration and carbon dioxide in the lower atmosphere (Fig. 10). The aim of these measurement flights was to investigate the vertical and horizontal extent of biomass burning

smoke in Finland from the Russian wildfires at that time. The measurement flights were performed at altitudes between the ground and about 3.8 km. One measurement profile is considered to include a climb from the ground level up to 3.8 km or a descent from 3.8 km down to the ground level. Altogether the measurements included 16 profiles during two days.

The first flight on 29 July 2010 at 7:45–10:30 was carried out from Pirkkala to Savonlinna. The second measurement flight of that day was performed at 12:30–14:30 from Savonlinna back toward Pirkkala, but now via Joensuu and Jämsä. During each flight, a total of four profiles were measured (*see* Fig. 10).

The first measurement day was very hot and hazy in Finland with poor visibility during both flights. The maximum temperature at the Hyytiälä measurement station was 32.7 °C, the minimum visibility and relative humidity were 9.7 km and 32.2%, respectively. Clouds had accumulated throughout the day up to the 3.8 km altitude. Especially around the Jämsä area, one could clearly smell the smoke. The maximum total particle number concentration reached about 6000 cm⁻³ over the Jämsä area between the altitudes of 2.5 and 3.8 km (Fig. 11g and h). During the morning flight we measured higher particle number concentrations in the boundary layer (500–700 m) as compared with those in the

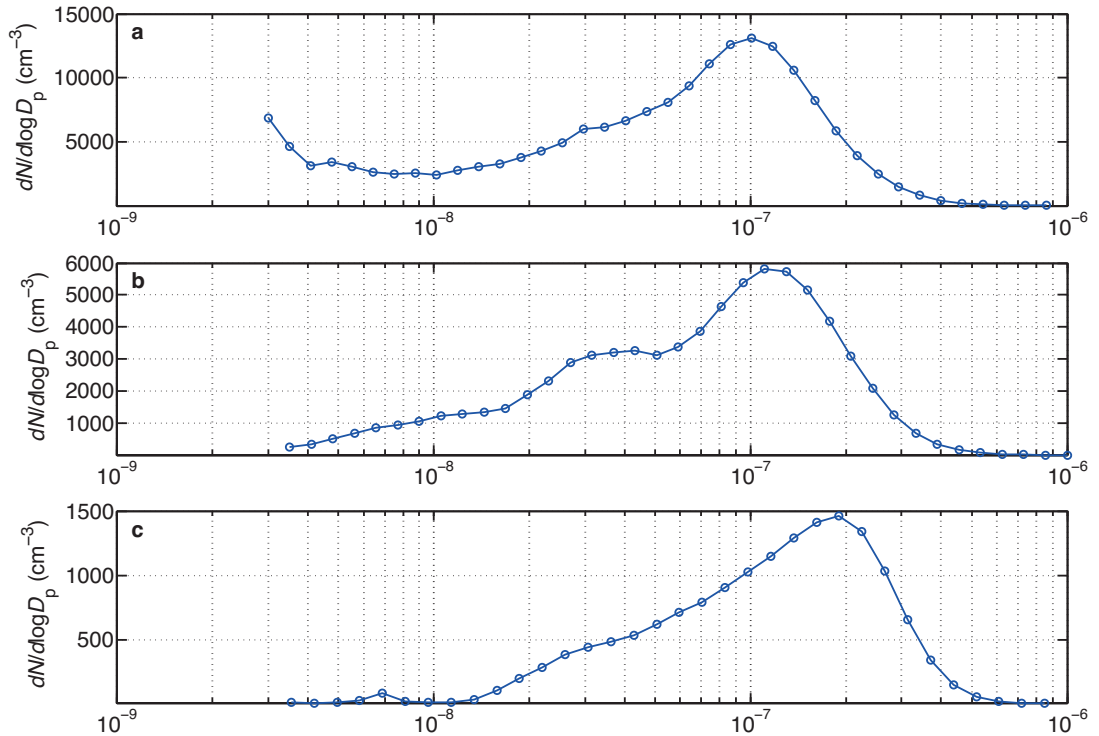


Fig. 9. The growth of the mean size of particle population during few days measured with DMPS instruments at three stations in Finland during Russian wildfires in summer 2006. Particle size distribution measured in (a) Helsinki on 30 April, (b) Hyytialä on 1 May, and (c) Värriö on 5 May 2006.

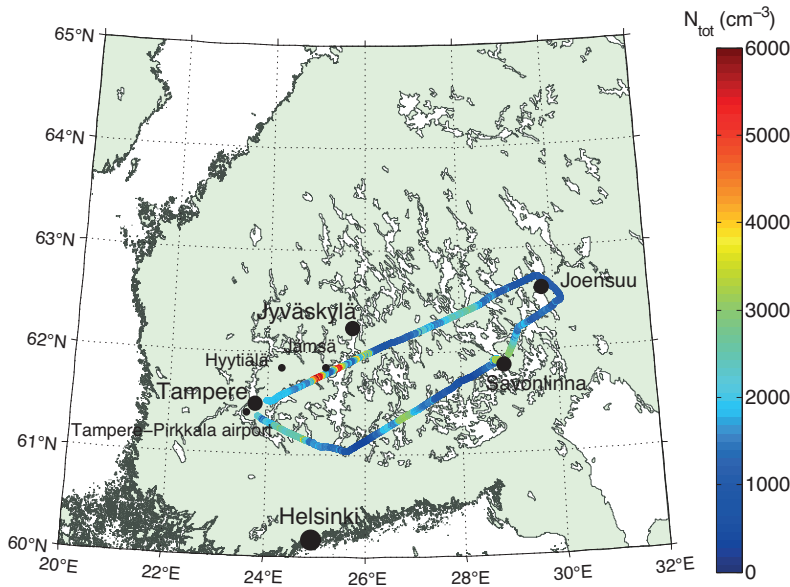


Fig. 10. The route of the flight on 29 July 2010. The flights were operated from and to Tampere-Pirkkala airport. The colours of the line indicate the total particle concentration (cf. the scale).

free troposphere; the concentrations were mainly in the range of 2000–5000 cm^{-3} in the boundary layer and $< 2000 \text{ cm}^{-3}$ above it. The measured

concentrations on 29 July 2010 afternoon were higher than typically in the free troposphere above the southern Finland (Schobesberger *et*

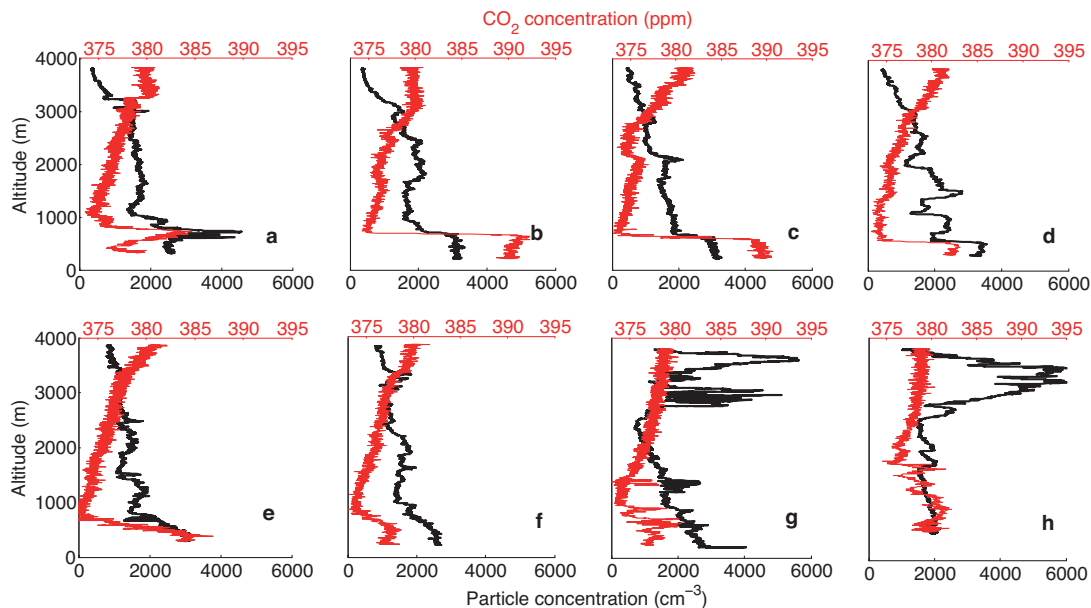


Fig. 11. Total particle concentrations and carbon dioxide during 8 measurement profiles: (a) first climb, (b) first descent, (c) second climb, and (d) second descent in the morning of 29 July 2010; and (e) first climb, (f) first descent, (g) second climb, and (h) second descent during the afternoon flight on the same day.

al. 2013). On 29 July 2010 at the SMEAR II station in Hyytiälä, the total particle number concentration varied between about 2000 and 5000 cm⁻³, the mean \pm standard deviation being 2139 ± 1536 cm⁻³ during July–August. The smoke plumes were thus observed more clearly in the Jämsä area in the free troposphere than at the ground level in Hyytiälä just 50 km away. The MODIS AOD detections (Fig. 6) confirm that there was smoke around that area on the same day, and that the smoke was not uniformly distributed over southern Finland.

The vertical distribution of the CO₂ concentration followed roughly that of the particle number concentration inside the boundary layer. In the most intensive plume observed in the free troposphere around the Jämsä area, there was no apparent increase in the CO₂ concentration. Taken together, the smoke from wildfires seemed to arrive to Finland on 29 July 2010 in several layers. During this measurement day, the air mass back trajectories at the 100, 500 and 3000 m levels all came to Finland from east or southeast. Moreover, during the transition the air masses were lifted from 1500–2000 meters altitude to 3000 meters.

The wind direction changed during the night of 29/30 July 2010, so that on the next day the air masses came to southern Finland from the south and southwest and therefore not from the fire areas. No clearly-elevated particle or carbon dioxide concentrations were measured during the flights on 30 July 2010.

The vertical extent of the long-range-transported smoke plume can also be seen from the CALIOP satellite based lidar images (Fig. 12, CALIPSO flying route in Fig. 2). Over the fire areas (ca. 50–55 °N), the smoke was detected up to 5 km above the ground level. Northward from the fire areas about 700 km east towards Hyytiälä (62 °N), the main plume was getting lower and high backscattering was detected in several layers between 0 and 5 km. A layered structure of the smoke was also reported on 29 July 2010 and 8 August 2010 in Kuopio (Mielonen *et al.* 2012) and on 30 July 2010 in Pallas and Sodankylä, northern Finland (Mielonen *et al.* 2013). Figure 12 can also be compared with Fig. 6, as in the morning of 29 July 2010 Aqua and CALIPSO satellite overpasses over north-eastern Europe differed by less than 10 minutes. We conclude that the CALIOP instrument

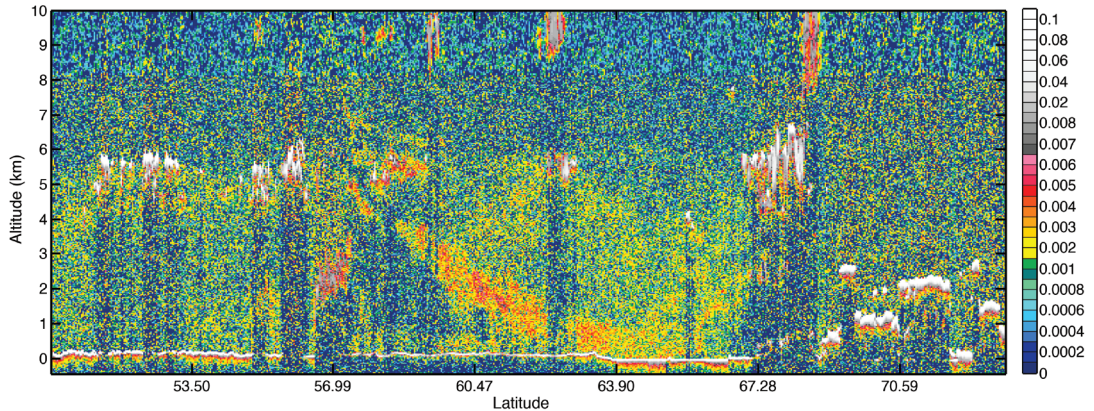


Fig. 12. Total attenuated backscatter ($\text{km}^{-1} \text{sr}^{-1}$) at 532 nm wavelength measured by CALIOP lidar onboard CALIPSO satellite on 29 July 2010 around 10 AM UTC. The flight route is marked in Fig. 2.

catches one of the main, long-range-transported plumes, and that although in Finland the aerosol particle concentrations were high, we probably did not measure the densest parts of the smoke in our measurements.

Summary and conclusions

Biomass-burning smoke from eastern European wildfires was observed in large parts of Finland in 2006 and 2010. Observations at the three SMEAR stations in different parts of Finland, complemented by flight measurements over southern Finland, enabled us to comprehensively study the horizontal and vertical extent of long-range transported smoke. In support of the observations, we analysed the movements of the arrived air masses with HYSPLIT 4 backward trajectory model, together with satellite-based MODIS fire detections and CALIOP vertical aerosol profiles. Several studies have already been published on the chemical and optical properties of these smoke plumes in different parts of Finland. In our study, we were able to combine this information with large-scale transport processes due to concurrent measurements at the three stations, as well as with the vertical structure of the smoke.

The difference between the four most intense smoke episodes and background air was clear. The most reliable smoke markers in our observations were carbon monoxide (CO) with daily

concentration increases of 48%–155% as compared with the reference period, black carbon (BC) with increases of 249%–578%, and the number concentration of aerosol particles in the accumulation mode size range reflected by the increase in the condensation sink by 100%–580%. Some of the episodes showed elevated concentrations of CO_2 , SO_2 , O_3 and NO_x as well. The smoke was observed to be highly scattering, with the single-scattering albedo of 0.96 ± 0.01 . The median particle size was 60%–250% larger during the smoke episodes as compared with that in the background air, indicating substantial growth of sub-100 nm particles into the accumulation mode during the long-range transport of fire pollution. During one of the episodes, such growth was observed to continue in the already-aged plume over Finland.

While the wildfire plumes originating from eastern Europe could be detected over the whole Finland, the horizontal and vertical extent of the plumes varied considerably. The most widely-spread and persistent of the smoke episodes was caused by a blocking high-pressure system located over the northeastern Europe, whereas in three shorter episodes the smoke came in a warm sector of a low-pressure system. At least one of the smoke plumes had a layered structure, and in southern Finland the highest particle number concentrations were measured above the boundary layer.

Our analysis demonstrates the importance of using multiple methods in analysing the charac-

ter of long-range transported pollution, including that originating from remote wild fires. The remote sensing methods used here confirmed the source areas of the observed pollutants, whereas the back trajectory analysis revealed us the differences between the air masses. Synoptic analysis — in addition to satellite images, back trajectories and *in situ* measurements — enabled us to determine the horizontal extent and inhomogeneity of the smoke plume.

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