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► **To cite this version:**

J. V. Niemi, H. Tervahattu, H. Vehkamäki, J. Martikainen, L. Laakso, et al.. Characterization of aerosol particle episodes in Finland caused by wildfires in Eastern Europe. *Atmospheric Chemistry and Physics Discussions*, 2005, 5 (2), pp.2469-2501. hal-00301234

HAL Id: hal-00301234

<https://hal.science/hal-00301234>

Submitted on 18 Jun 2008

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Characterization of aerosol particle episodes in Finland caused by wildfires in Eastern Europe

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Received: 31 January 2005 – Accepted: 4 April 2005 – Published: 22 April 2005

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Abstract

We studied the sources, compositions and size distributions of aerosol particles during long-range transport (LRT) PM_{2.5} episodes occurred on 12–15 August, 26–28 August and 5–6 September 2002 in Finland. Backward air mass trajectories, satellite detections of fire areas, and dispersion modelling results indicate that emissions from wildfires in Russia and other Eastern European countries arrived to Finland during the episodes. Individual particle analyses using scanning electron microscopy (SEM) coupled with energy dispersive X-ray analyses (EDX) showed that the proportion of S-rich particles increased during the episodes and they contained elevated fractions of K, which indicates emissions from biomass burning. These aerosols were mixed with S-rich emissions from fossil fuel burning during the transport, since air masses came through polluted areas of Europe. Minor amounts of coarse Ca-rich particles were also brought by LRT during the episodes, and they probably originated from wildfires and/or from Estonian and Russian oil-shale burning industrial areas. The ion chromatography analysis showed that concentrations of sulphate (SO₄²⁻), total nitrate (NO₃⁻+HNO₃(g)) and total ammonium (NH₄⁺+NH₃(g)) increased during the episodes, but the ratio of total amount of these ions to PM₁₀ concentration decreased indicating unusually high fractions of other chemical components. The particle number size distribution measurements with differential mobility particle sizer (DMPS) showed that the concentrations of 90–500 nm particles increased during the episodes, but the concentrations of particles smaller than 90 nm decreased. The reduction of the smallest particles was caused by suppressed new particle formation due to the vapour and molecular cluster uptake of LRT particles. Our results show that the emissions from wildfires in Russian and other Eastern European deteriorated air quality on very large areas, even at the distance of over 1000 km from the fire areas.

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1. Introduction

Large amounts of fine particles ($PM_{2.5}$) are emitted from various sources and transported long distances due to their slow removal from the atmosphere. Thus, in the areas with low local emissions, major fraction of the $PM_{2.5}$ mass is usually long-range transported. Fine particles are associated with adverse health effects (Dockery and Pope, 1994; Laden et al., 2000; Pope et al., 2002; WHO, 2002, 2003), and no threshold concentration has been found below which they have no effect on health (WHO, 2003).

The properties and sources of long-range transport (LRT) particles in Finland have been studied in association with several short-term campaigns and long-term monitoring measurements. The emissions and mass concentrations of $PM_{2.5}$ are generally quite low compared with more polluted regions of Europe (EMEP, 2001; Ruuskanen et al., 2001; EMEP, 2002). Even in the urban areas of Helsinki, 50–70% of $PM_{2.5}$ mass is caused by LRT (Vallius et al., 2003; Karppinen et al., 2004). In general, high concentrations of LRT $PM_{2.5}$ mass and typical chemical components of LRT particles, such as sulphate and nitrate, are observed in Finland when air masses arrive from Eastern and Central Europe (Pakkanen et al., 2001; Ruoho-Airola et al., 2004). In northern Finland, the emissions from Arctic Ocean and from Kola Peninsula industrial areas in Northern Russia also rise LRT particle mass concentrations (Virkkula et al., 1999; Yli-Tuomi et al., 2003). However, the sources and properties of aerosols during the strongest LRT periods are still poorly known in Finland, and only few LRT episodes have been studied in detail so far. The main sources of previously studied LRT episodes were marine aerosols from Atlantic Ocean (Tervahattu et al., 2002a, b), emissions from oil-shale burning industrial areas in Estonia and Russia (Tervahattu et al., 2004), and emissions from spring-time agricultural field-burning and fossil fuel burning in Russia and other Eastern European countries (Niemi et al., 2004).

In the present article we describe three LRT $PM_{2.5}$ episodes that occurred in Finland in August and September 2002. We studied mass and number concentrations of PM

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as well as its chemical composition using bulk and individual particle methods. Backward air mass trajectories, fire satellite observations, dispersion modelling results and emissions source data were also used to identify the origins and to characterize the dispersion of the LRT emissions. The main sources of the episodes were emissions from forest and peat fires mixed with emissions from fossil fuel burning. A comparison between an earlier LRT PM_{2.5} episode which occurred during spring-time agricultural field burning period (Niemi et al., 2004) is also carried out.

2. Material and methods

We used several measurement and sampling sites in this study and their locations are shown in Fig. 1. The summary of measurements and samplings performed in these sites are given in Table 1, and they are described in detail in following sections.

2.1. Meteorological and emission data

The meteorological conditions were observed using data collected by the Finnish Meteorological Institute at Kaisaniemi in Helsinki and by the Helsinki Metropolitan Area Council at Pasila in Helsinki. Backward air mass trajectories were produced using the vertical motion model in the HYbrid Single-Particle Lagrangian Integrated Trajectory (HYSPPLIT4) model (Draxler and Rolph, 2003) with the Final Analyses (FNL) meteorological database at NOAA Air Resources Laboratory's web server (Rolph, 2003). For detection of fire areas, we used Web Fire Mapper (<http://maps.geog.umd.edu>), which is part of the MODerate-resolution Imaging Spectroradiometer (MODIS) Rapid Response System (Justice et al., 2002). The Navy Aerosol Analysis and Prediction System (NAAPS) model results were used to determine the distribution of smoke, dust and sulphate aerosols. Details of the model and model results are available at the internet pages (<http://www.nrlmry.navy.mil/aerosol/>) of the Naval Research Laboratory, Monterey, California. Using data of the European Monitoring and Evaluation

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Programme (EMEP), we searched for information on trace gas and particle emissions of areas along the air mass routes (EMEP, 2001, 2002).

2.2. Particle mass and number size distributions

Particle mass and number size distributions are measured at a rural background station (SMEAR II) in Hyytiälä (Kulmala et al., 2001) by the University of Helsinki (Department of Physical Sciences). The particle mass fractions were measured with a Dekati PM₁₀ 3-stage impactor with a backup filter. The duration of sample collection was 2–3 days. The cut-off diameters (D_{50}) of the impactor stages were 10, 2.5 and 1 μm . The collection substrates on impaction stages were polycarbonate membranes (Nuclepore 800 203, diameter 25 mm) with no holes, and the backup filter material was made of teflon (Gelman Teflo R2P J047, diameter 47 mm) with 2 μm diameter pore size. The smearing of substrates to prevent particles from bouncing back from the substrates, and the gravimetric analysis are described in detail by Laakso et al. (2003). The PM₁₀ mass monitoring measurements performed in Virolahti, Helsinki (also PM_{2.5}) and Imatra are shortly described in Sects. 2.3 and 2.4.

Particle number size distributions were measured in the diameter between 3 and 500 nm using a differential mobility particle sizer (DMPS) (Aalto et al., 2001) in Hyytiälä. The time resolution was 10 min. Small particles with diameter less than about 10 nm were measured with a TSI3025 condensation particle counter (CPC) and Hauke-type differential mobility analyser (DMA) and whereas larger particles with a TSI3010 CPC and Hauke-type DMA.

2.3. Ion analyses

Rural background air quality is monitored at several measurement stations in Finland by the Finnish Meteorological Institute (FMI). We present ion measurement results from the EMEP stations in Ähtäri, Virolahti and Utö. Daily total suspended particle (TSP) samples were collected using open-faced 2-stage filter packs (NILU Products

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AS) containing cellulose filters (Whatman 40, diameter 47 mm). Sulphate (SO_4^{2-}), total nitrate ($\text{NO}_3^- + \text{HNO}_3(\text{g})$), and total ammonium ($\text{NH}_4^+ + \text{NH}_3(\text{g})$) were analysed with ion chromatography (IC, Dione DX-500 and Waters). The details of the sampling and IC methods are described in Paatero et al. (2001). We also compared the ion masses and PM_{10} masses measured in Virolahti. The PM_{10} mass was monitored with Eberline FH 62 I-R (Eberline Instruments), which is based on β -attenuation.

2.4. Individual particle analyses by SEM/EDX

Air quality is monitored at several measurement stations by the Helsinki Metropolitan Area Council in the Helsinki metropolitan area, and by the Environment Office of Imatra in the region of Imatra. We selected particle samples for individual particle analyses collected at Kallio and Luukki in the Helsinki area during all 3 episodes studied, and at Mansikkala in Imatra during the last episode. Particle samples were collected with Eberline FH 62 I-R samplers (Eberline Instruments) on fibreglass filter tape. In all, 17 particle samples (9 $\text{PM}_{2.5}$ and 8 PM_{10} samples), collected during the episode (13 August, 27 August, 28 August, 5 September and 6 September 2002) and reference (8 March, 10 March, 16 March, 18 August, 22 August, 17 September and 22 September 2002) days, were selected for the analyses.

The elemental compositions of individual particles and agglomerates were studied using scanning electron microscopy (SEM – ZEISS DSM 962) coupled with energy dispersive X-ray analyses (EDX – LINK ISIS with the ZAF-4 measurement program). The SEM/EDX samples were prepared by pressing a tape (Scotch Ruban Adhesive) attached to an aluminium plate onto the filter surface covered with particles. Then samples were coated with carbon (Agar SEM Carbon Coater) to make the sample surfaces conductive. The SEM/EDX was operated at 20 kV accelerating voltage, and counting time of the X-ray spectrum was 15 s. The elemental compositions of 100 randomly selected particles or agglomerates (minimum diameter $1 \mu\text{m}$) were analysed from each sample. Submicron-sized particles had formed abundant large agglom-

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erates during sampling and sample preparation for SEM/EDX analysis, and thus we could also obtain elemental information from submicron size-range by analysing these agglomerates. The relative elemental weight percentages (normalised to 100%) were calculated using ZAF-4 corrections for the following elements: Na, Mg, Al, Si, S, Cl, K, Ca, Ti and Fe. The ZAF-corrected elemental results are semiquantitative, but the accuracy is well sufficient to identify different particle types and to compare the differences in elemental ratios of the same particle type in different samples (Kupiainen et al., 2003; Niemi et al., 2004).

3. Results and discussion

3.1. General description of the LRT episodes

PM₁₀ and PM_{2.5} concentrations rose on extraordinarily high levels in Finland during three periods in August and September, 2002. We focus on the strongest periods of these episodes; the first episode (EPI-1) was on 12–15 August, the second (EPI-2) on 26–28 August and the third (EPI-3) on 5–6 September (Fig. 2). These episodes were also observed in Estonia and Sweden (Niemi et al., 2003; Räsänen et al., 2004). In Finland, the episodes were strongest in south, but they were observed weaker over a wide area, including as far north as Oulu, 600 km from southern coast of Finland. The maximum hourly PM₁₀ concentrations were 70–90 $\mu\text{g m}^{-3}$ in Southern Finland during the episodes, whereas the typical values in these areas are $\sim 15\text{--}20 \mu\text{g m}^{-3}$ (Anttila et al., 2003). The EPI-3 was especially strong in Southeastern Finland, where the maximum hourly PM₁₀ concentrations were 200–220 $\mu\text{g m}^{-3}$. Most of the particle mass was in the PM_{2.5} size range, and PM_{2.5} values observed during the episodes were multifold compared with long-term mean values. E.g. at Kallio in Helsinki, the maximum hourly PM_{2.5} values (maximum 24-h mean values in parenthesis) were 45 (33), 54 (42) and 81 (29) $\mu\text{g m}^{-3}$ during the EPI-1, EPI-2 and EPI-3, respectively, whereas the 3-year (1999–2001) summer-autumn mean was only 7.9 $\mu\text{g m}^{-3}$ (Laakso

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et al., 2003). The concentrations of coarse particles (low $PM_{2.5}$ - PM_{10} values in Fig. 2) increased also slightly during the episodes, especially during the EPI-1.

August and the beginning of September 2002 were exceptional warm and dry in Finland and in the neighbouring areas. For example, the monthly mean temperature at Kaisaniemi in Helsinki was 19.4°C in August, whereas the long term (1971–2002) mean value is 15.8°C . The total monthly precipitation values were 6 and 5 mm in August and September, while the long term (1971–2000) mean values are 78 and 66 mm, respectively. August 2002 was the warmest August and September 2002 the driest September of 100 year period. During the episodes at Pasila in Helsinki, the wind speed (hourly mean) was $2\text{--}6\text{ m s}^{-1}$, relative humidity 40–90% and temperature $15\text{--}28^{\circ}\text{C}$.

The backward trajectories showed that air masses arrived to Helsinki during the EPI-1 from Eastern Estonia, Russia and Ukraine (Fig. 3). During the EPI-2, trajectories curved along a more western route, and air masses arrived to Helsinki from Russia via the Baltic Sea and the Baltic countries (Estonia, Latvia and Lithuania). The EPI-3 was strongest in South-eastern Finland, and air masses arrived over Virolahti and Imatra from the direction of Russia and Eastern Estonia. The satellite observations of potential fire areas indicated that there were many wildfires in Russia, Ukraine and Belarus and in the Baltic countries during the episodes. The amount of fires near Finland was high already before and during the EPI-1 (Fig. 4). High emissions from large forest and peat fires led to accumulation of large smoke plumes over Eastern Europe (Goldammer, 2003). The NAAPS model results indicated that the strongest smoke plumes reached Finland during the EPI-2 and especially during EPI-3 (Fig. 5).

3.2. Particle mass and number size distributions

We compared the particle mass and number size fractions at Hyytiälä measurement station during the episode periods (12–15 August, 26–27 August and 6–8 September 2002) with reference periods surrounding the episodes (2 August–15 September 2002) (Table 2). The sample collection times for mass concentration measurements were 2–

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3 days, and therefore we had to select slightly different periods for EPI-2 and EPI-3 in this Sect. 3.2 compared with other sections of this paper. Most of the particle mass was in the submicrometre size range during all episodes as well as on the reference days. During the episodes the particle mass increased on average by a factor of 3 for the size classes PM_1 and $PM_{1-2.5}$ and by a factor of 2 for the size class $PM_{2.5-10}$, but above $10\ \mu m$ size class the concentration remained about at the reference level. During the three episodes, all these PM concentrations were close to those observed during the LRT episode in March 2002, which originated from agricultural field-burning emissions mixed with fossil fuel burning emissions (Niemi et al., 2004).

The particle number concentration increased on average by a factor of 1.8 in the accumulation mode and decreased (by a factor of 0.5) in the Aitken and nucleation modes (Table 2 and Fig. 6a). The decrease of the smallest particles was expected, since small molecular clusters and particles are scavenged efficiently by high amount of larger particles (see e.g. Mönkkönen et al., 2004). Abundant surface area of larger particles also consumes condensable vapours preventing particle formation and growth. The total concentration of particles with diameter less than 500 nm was fairly unaffected, since the decrease in nucleation and Aitken mode concentrations was balanced by the increase in accumulation mode concentrations.

Figure 6b shows how big a fraction of particles smaller than 500 nm in diameter belong to accumulation and nucleation modes. The ratio of accumulation mode concentration to Aitken mode concentration (Accum/Aitken) is also shown. The Aitken mode particles cannot be transported in the atmosphere from as far away as accumulation mode particles. The deposition efficiency of Aitken mode particles is high, they disappear by colliding and sticking to the accumulation mode particles, and they grow fast due to condensation of vapours. Furthermore, the sources of Aitken mode particles are scarce in the areas around Hyytiälä. Thus, Aitken mode concentrations in Hyytiälä normally exceed those of the accumulation mode, and Accum/Aitken ratio greater than one is typical for polluted air masses transported over long distances. The high values (>1) of Accum/aitken might thus be a good indicator LRT episodes in general. This

concept is also supported by the observations made during the LRT episode in March 2002.

3.3. Ion analyses

The 24-h mean TSP concentrations of sulphate, total nitrate, and total ammonium increased strongly at the rural EMEP station of Virolahti during the episodes (Fig. 7). The ion sum (total amount) of these chemical components was 8.2, 7.2 and 12.1 $\mu\text{g m}^{-3}$ during EPI-1, EPI-2 and EPI-3, respectively. The ion sum in Utö was about the same level as in Virolahti, whereas that in Ähtäri, more northern station (see Fig. 1), was clearly lower (factor of 2/3). The values during the episodes were 3–4 times higher than those usually observed at these three stations in this time of year (Leinonen, 2001).

We compared ion sum and PM_{10} concentration measured in Virolahti during the episodes and reference period (Fig. 8). The ion sum to PM_{10} ratios were lower than usually during the episodes; means were 0.28, 0.18, 0.17 and 0.33 during EPI-1, EPI-2, EPI-3 and reference period, respectively. This indicates that the particle matter contained unusually high fractions of other chemical components than sulphate, nitrate and ammonium during the episodes. The ion sum to PM_{10} ratio was especially low (0.14) during the peak stage of EPI-3 (6 September 2002), when strongest smoke plume from wildfires reached South-eastern Finland. Large emissions of organic and black carbon and other chemical components from wildfires was probably the main reason for the reduction of the relative fraction of ion sum during the episodes as will be discussed below.

3.4. Individual particle analysis with SEM/EDX

We analyzed the elemental composition of single particles and agglomerates with SEM/EDX from samples collected during the episodes and reference days. The particles were classified into 6 different groups based on the element that was most abun-

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dant in each particle: (1) S, (2) Si or Al, (3) Ca, (4) Fe, (5) Na or Cl, (6) low-Z. Particles in the low-Z class contained only elements with an atomic number lower than 11 (Na), such as C, N and O, which could not be analysed quantitatively with the SEM/EDX we used. We also classified particles with hierarchical cluster analysis (SPSS 10.0 statistical program) using Ward's method with squared Euclidean measures, which suits well to grouping of different particle types (Bernard and Van Grieken, 1992). However, the elemental composition of the main particle type (S-rich) during the episodes proved to be so homogenous that we show only some details of the clustering classification.

The amount of S-rich particles and agglomerates was extraordinarily high during the episodes (Fig. 9). The $PM_{2.5}$ samples of the urban background station at Kallio contained 65% and 71% S-rich particles during the peak day of EPI-1 and EPI-2, respectively, but only on average 10% (range 7–13%) during the reference days. The EPI-3 was clearly strongest in South-eastern Finland, but unfortunately no $PM_{2.5}$ samples were collected during that time there. Therefore we studied PM_{10} samples collected at urban station in Imatra. The fraction of S-rich particles was also very high (55%) during the peak day of EPI-3 in Imatra compared with the value (3%) of the reference day.

In addition to S, S-rich particles and agglomerates contained large amounts of C and O as well as some Na, Si and K (EDX spectra shown in Fig. 10). The presence of C was confirmed by analysing some S-rich particles and agglomerates directly on the gold coated surface of (C-free) fibreglass filters. The fractions of other elements were usually quite low, but some of the particles contained also elevated concentrations of Ca. The elemental ratios of S-rich particles in different samples were compared (Fig. 10). K was the only element the relative weight percentage of which had statistically significant ($p < 0.01$, Mann-Whitney U-test, SPSS 10.0 statistical program) differences between episode and reference samples. The relative weight percentages of K in S-rich particles were higher during all episodes (8.9% EPI-1, 8.9% EPI-2 and 8.2% EPI-3) compared with reference days (6.3%). This is a clear indication of emissions from biomass burning (Andreae, 1983; Andreae et al., 1998). S-rich particles also contained some Na and Si, which may have originated partly from biomass burn-

ing but also from many other sources, e.g. marine aerosols, fossil fuel burning and refuse incineration (Watson et al., 2001; Ooki et al., 2002).

The relative weight percentages of K in S-rich particles did not rise (range 8.2–8.9%) as high as during the LRT PM_{2.5} episode in March 2002 (range 11.4–19.3%), which originated from agricultural field burning emissions mixed with fossil fuel burning emissions (Niemi et al., 2004). The smouldering burning conditions are more dominating in boreal forest fires and peat fires compared with grass, shrub or cereal waste fires, which are mainly burned in flaming conditions (Echalar et al., 1995; Lavoue et al., 2000; Ortiz de Zárate et al., 2000; Soja et al., 2004). Thus it seems likely that aerosols from forest and peat fires were less enriched with K than aerosols from agricultural field burning since the fraction of K is lower in the emissions from smouldering fires compared with flaming fires (Echalar et al., 1995; Gaudichet et al., 1995; Robinson et al., 2004). Another reason to relatively low proportion of K was probably the mixing of emissions from fossil fuel burning since air masses arrived from the regions of Eastern Europe with high S emissions (EMEP, 2001). However, the fraction of fossil fuel burning emissions in LRT particle mass was probably smaller during the wildfire episodes compared with the March 2002 episode, because the relative weight ratio of S in S-rich particles did not rise as high as during the peak stage of the March episode (61%) (Niemi et al., 2004).

The PM measurements performed in Helsinki showed that mass concentrations of coarse (PM_{2.5–10}) particles also increased slightly during the episodes (see Fig. 2). Since Si-Al-rich particles are usually clearly the dominating group of PM₁₀ particles in Helsinki and other Finnish cities (Haapala, 1999; Räisänen et al., 2004), we compared the relative fractions of the Si-Al-rich group with the other groups. The ratio of Ca-rich to Si-Al-rich particle groups proved to be exceptional high during all the three episodes in Helsinki, and also in Imatra during EPI-3 (Table 3). The ratios were 0.24–0.53, which are 3–7 times higher than the values of the reference days and the long-term averages in Helsinki and Lappeenranta (Haapala, 1999; Räisänen et al., 2004).

The increased proportions of Ca-rich particles in PM₁₀ samples indicate that coarse

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Ca-rich particles were also brought by LRT during the episodes. Their mode and median sizes (geometric diameter) were 2 and 3 μm . The elemental combinations of Ca-rich particles were investigated with cluster analysis, and typical combinations were Ca, Ca-S-(Si-Na), Ca-(S-Si-Al) and Ca-Mg-(Si) (minor elements in parentheses). The elemental combinations suggest that major components of Ca-rich particles might have been CaCO_3 , CaSO_4 and $\text{CaMg}(\text{CO}_3)_2$.

Forest and peat fires were probably one of the major sources of Ca-rich particles during the episodes since Ca-rich particles are emitted from incomplete burning of biomass and most of their mass is in coarse ($D_p > 1 \mu\text{m}$) size fraction (Allen and Miguel, 1995; Osan et al., 2002; Li et al., 2003; Pagels et al., 2003). Intensive fires also mobilise particles from the surfaces of plants and ground (Allen and Miguel, 1995; Gaudichet et al., 1995; Pereira et al., 1996). The injection heights of particles from forest fires are in moderate surface fires usually ~ 2.3 km or less and in intensive crown fires ~ 5 – 12 km (Lavoue et al., 2000). Therefore the smallest particles of coarse size fraction may well be transported from the wildfire areas to Finland.

Ca-rich particles are also emitted from many other sources including fossil fuel burning and cement and metal industries (Hoornaert et al., 1996; Lee and Pacyna, 1999). The trajectories indicated that air masses arrived to Helsinki during EPI-1 and to Imatra during EPI-3 via Estonian and Russian oil-shale burning industrial areas located in Narva (see Figs. 1 and 3) and Slantsy (~ 25 km south from Narva). This area is one of the strongest particle sources of Northern Europe (EMEP, 2002), and the Ca content of emissions is high (Häsänen et al., 1997; Jalkanen et al., 2000; Tervahattu et al., 2004). These large emissions occasionally increase particle concentrations even in southern Finland under suitable meteorological conditions (Jalkanen et al., 2000; Tervahattu et al., 2004). Thus it is likely that Ca-rich particles may also have been transported from the regions of Narva and Slantsy to Helsinki during EPI-1 and to Imatra during EPI-3.

4. Summary and conclusions

Strong LRT PM_{2.5} episodes occurred on 12–15 August, 26–28 August and 5–6 September 2002 on the large areas of Finland. Backward air mass trajectories, satellite detections of fire areas, and dispersion modelling results indicate that emissions from wildfires in Russia and other Eastern European countries arrived to Finland during the episodes. The composition of individual particles and agglomerates was studied using scanning electron microscopy (SEM) coupled with energy dispersive X-ray analyses (EDX), and the bulk chemical concentrations of sulphate (SO₄²⁻), total nitrate (NO₃⁻+HNO₃(g)) and total ammonium (NH₄⁺+NH₃(g)) was studied using ion chromatography (IC). SEM/EDX analysis showed that the proportion of S-rich particles increased strongly during the episodes and that the relative weight percentage of K was unusually high in these particles. The high proportion of K indicates emissions from biomass burning. These aerosols were mixed with S-rich emissions from fossil fuel burning during the transport, since air masses came through polluted areas of Europe. Minor amounts of coarse Ca-rich particles were also long-range transported during the episodes, and they probably originated from wildfires and/or from the Estonian and Russian oil-shale burning industrial areas. The concentrations of sulphate, total nitrate, and total ammonium increased during the episode, but the ratio of total concentrations of these ions and PM₁₀ concentration decreased. This also confirms the presence of high amounts of particles originated from unusual (above mentioned) emissions sources. The composition of particles during the episodes were compared with earlier LRT PM_{2.5} episode originated from spring-time agricultural field burning, and the fraction of K was lower during the wildfire episodes, probably due to the dominance of smouldering burning conditions during boreal forest and peat fires.

The particle mass fractions were measured using a 3-stage impactor with a backup filter. Most of the LRT particle mass was in the submicrometre size fraction, and the mass of the submicrometre particles increased on an average by a factor of 3 during the episodes. The particle number size distributions were measured with differential

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mobility particle sizer (DMPS), and the number concentration of 90–500 nm particles increased by a factor of 1.8 during the episodes, but the concentrations of particles smaller than 90 nm were reduced. The reduction of the smallest particles was caused by suppressed new particle formation due to the vapour and molecular cluster uptake of LRT particles. These results show how strongly the LRT of particles affect not only the fine particle mass but also the number size distributions of submicron particles. The high values (>1) of accumulation mode to Aitken mode number concentration ratios seem to be a good indicator of LRT episodes, because Aitken mode particles cannot be transported in the atmosphere from as far away as accumulation mode particles.

Our results show that the emissions from wildfires in Russian and other Eastern European substantially affected PM_{2.5} concentrations on very large areas, even at distance over 1000 km from the fire areas. The wildfire season 2002 was exceptional severe in Eastern Europe. However, the measurements made by fire-monitoring satellites (<http://firemaps.geog.umd.edu>) and NAAPS model results (<http://www.nrlmry.navy.mil/aerosol/>) indicate that smoke from uncontrolled fires (and from agricultural field burning) is generally an important factor deteriorating air quality locally and even regionally in eastern and southern parts of Europe.

Acknowledgements. We acknowledge the Helsinki University Environmental Research Centre, the Finnish Cultural Foundation, the Helsinki Metropolitan Area Council and the Academy of Finland for funding this study. The Cooperative Institute for Research in Environmental Sciences, University of Colorado at Boulder, is gratefully acknowledged for a Visiting Fellowship (HT). We would like to thank our colleagues at the following institutes for their collaboration during the work: P. Aalto and V. Hiltunen (Division of Atmospheric Sciences, University of Helsinki), R. Hillamo, J. Kukkonen and T. Salmi (Finnish Meteorological Institute), M. Räisänen (The Geological Survey of Finland), K. Kupiainen (Nordic Envicon Ltd.), K. Lounatmaa (Helsinki University of Technology) and O. Hänninen (Department of Environmental Health, National Public Health Institute). The authors gratefully acknowledge the NOAA Air Resources Laboratory (ARL) for the provision of the HYSPLIT transport and dispersion model and READY website (<http://www.arl.noaa.gov/ready.html>) used in this publication. The Naval Research Laboratory is acknowledged for the NAAPS model results (website at <http://www.nrlmry.navy.mil/aerosol/>)

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Table 1. Description of measurement and sampling sites for different analysis.

Location	Environment	Particle mass fraction measurement	Particle number size distribution measurements	Samples for ion chromatography measurements	Samples for SEM/EDX measurements
Hyytiälä	Rural background	Size fractions ^a	x		
Virolahti	Rural background	PM ₁₀		x	
Utö	Rural background			x	
Ähtäri	Rural background			x	
Helsinki at Kallio	Urban background	PM ₁₀ , PM _{2.5}			x
Helsinki at Luukki	Rural background	PM ₁₀			x
Imatra at Mansikkala	Urban	PM ₁₀			x

^a $D_p < 1 \mu\text{m}$, $1\text{--}2.5 \mu\text{m}$, $2.5\text{--}10 \mu\text{m}$ and $>10 \mu\text{m}$

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Table 2. Mass and number size fractions in Hyytiälä during the episode (12–15 August, 26–27 August and 6–8 September 2002) and reference days (2 August–15 September 2002 excluding episode days). Sample change time was 5 a.m. UTC. We also show 3-year (1999–2001) mean summer-autumn values (Laakso et al., 2003), and values measured during March (17–19 March 2002) episode (Niemi et al., 2004).

	PM ₁	PM _{1–2.5}	PM _{2.5–10}	PM _{>10}	Nucl. (3–25 nm) # cm ⁻³	Aitken (25–90 nm) # cm ⁻³	Accum. (90–500 nm) # cm ⁻³	Total (3–500 nm) # cm ⁻³
	μg m ⁻³	μg m ⁻³	μg m ⁻³	μg m ⁻³				
Reference days	5.3	1.2	1.2	0.6	489 *343	1138	591	2219 *2072
Episode days	15.9	3.7	2.3	0.7	226 *174	588	1067	1880 *1828
Summer-autumn mean (Laakso et al., 2003)	4.6	1.2	1.3	–	*365	1065	710	*2130
March episode (Niemi et al., 2004)	15.4	6.9	2.3	0.3	273 *211	955	2703	3924 *3867

* Denotes the concentration calculated with nucleation mode limits 10–25 nm as in Laakso et al. (2003).

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Table 3. The number ratio of particles in Ca-rich particle group and Si-Al-rich particle group during the episodes (13 August EPI-1, 28 August EPI-2 and 5–6 September 2002 EPI-3) and reference days (Helsinki 22 August 2002 and Imatra 17 September 2002). We also show mean long-term values of the same ratio calculated from Haapala (1999) and from Räsänen et al. (2004).

Sampling place	EPI-1 Helsinki	EPI-2 Helsinki	EPI-3 Helsinki	EPI-3 Imatra	Reference Helsinki and Imatra	Haapala (1999) ^a Helsinki	Räsänen et al. (2004) ^b Lappeenranta
Number of analyzed Ca-rich and Si-Al-rich particles	77	81	91	35	107	700	1270
Ratio of Ca-rich and Si-Al-rich groups (SD)	0.24	0.53	0.45	0.49	0.08	0.07 (0.07)	0.11 (0.07)

^a 18 samples collected between March and April 1998

^b 13 samples collected between March and June 2003, Lappeenranta is a small town located ~35 km southwest from Imatra

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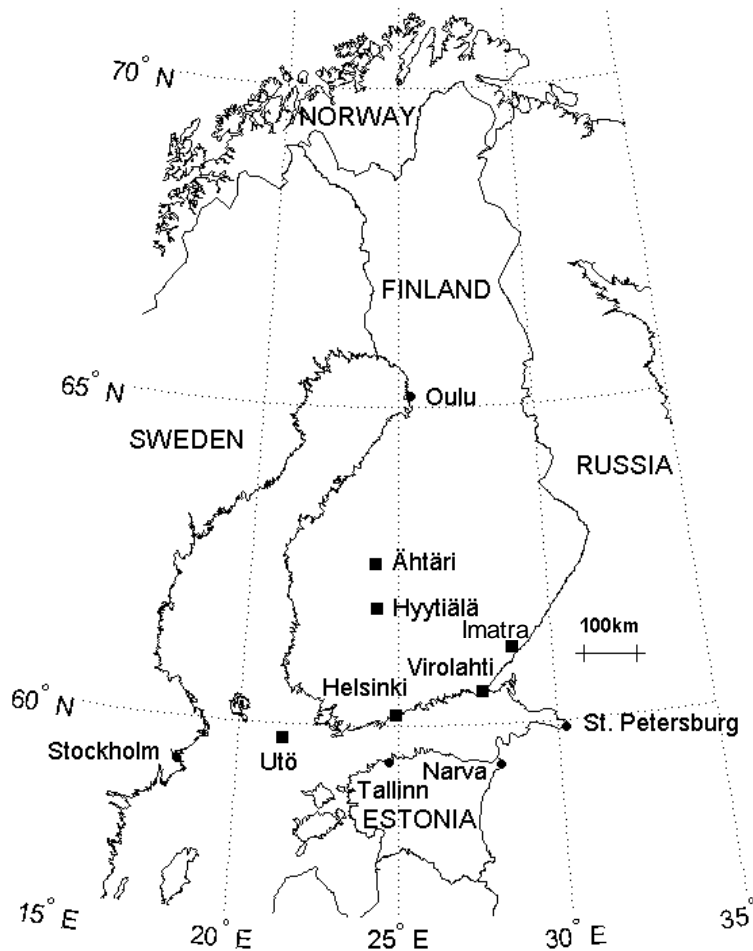


Fig. 1. Locations of the measurement and sampling sites (marked with squares).

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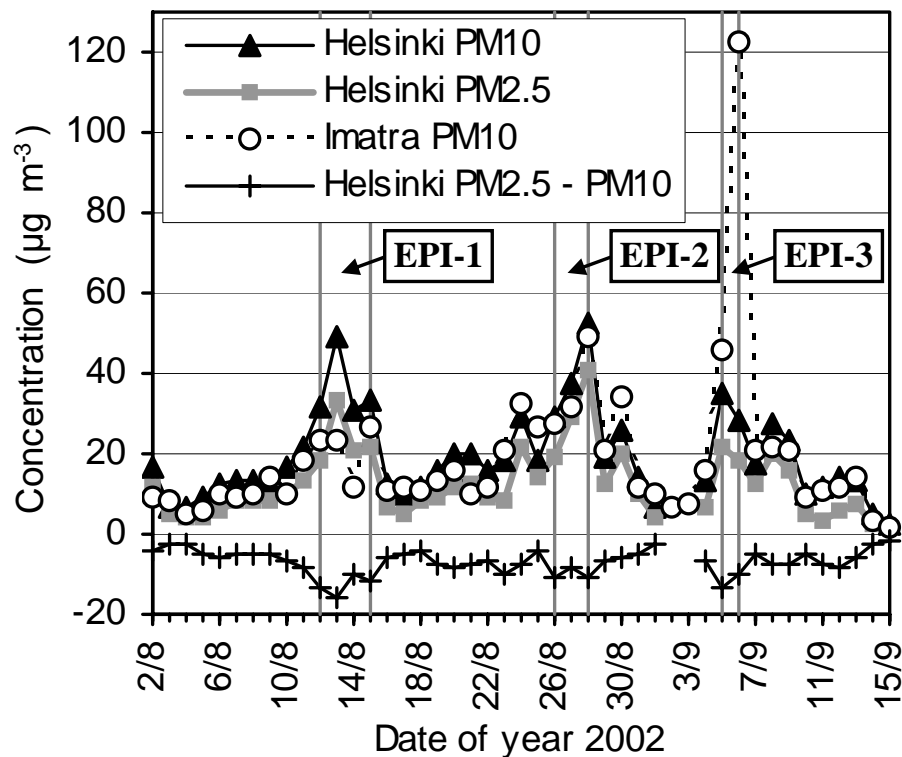


Fig. 2. Daily PM_{10} and $PM_{2.5}$ concentrations at Kallio in Helsinki and daily PM_{10} concentration at Mansikkala in Imatra on 2 August–15 September 2002. The three strong LRT episode periods (EPI-1, EPI-2 and EPI-3) are marked out with vertical lines. Note that the concentrations of coarse particles are depicted for figure clarity as a subtraction $PM_{2.5}-PM_{10}$ which gives negative values.

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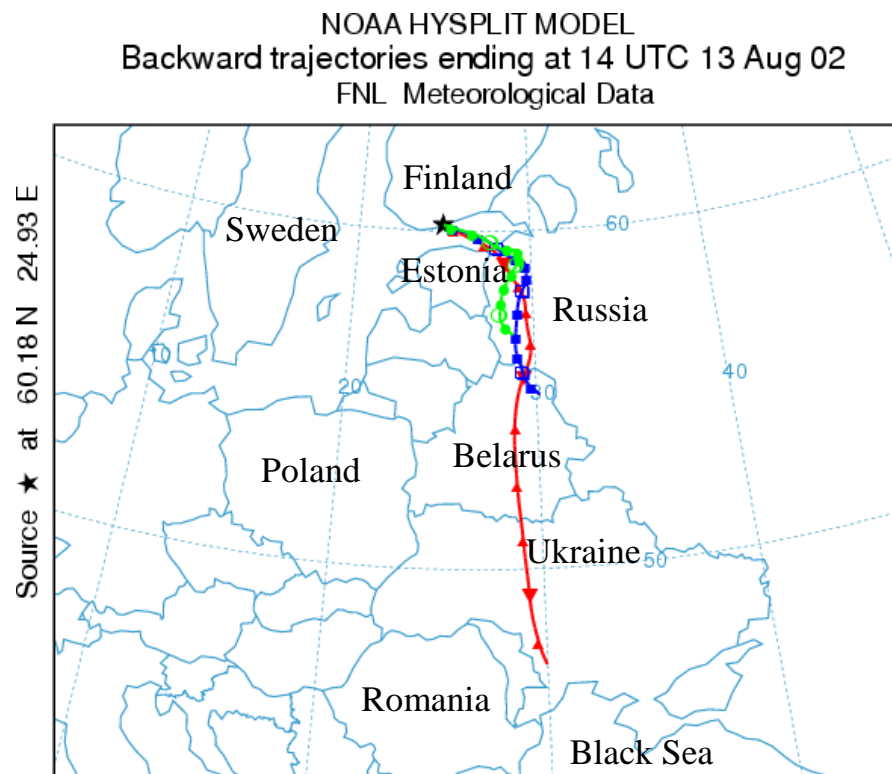


Fig. 3. Backward air mass trajectories (72 h) to Helsinki during the strongest stage of the EPI-1 (13 August 2002). Vertical heights at the start were 10 m (green circles), 100 m (blue squares) and 500 m (red triangles) above ground level.

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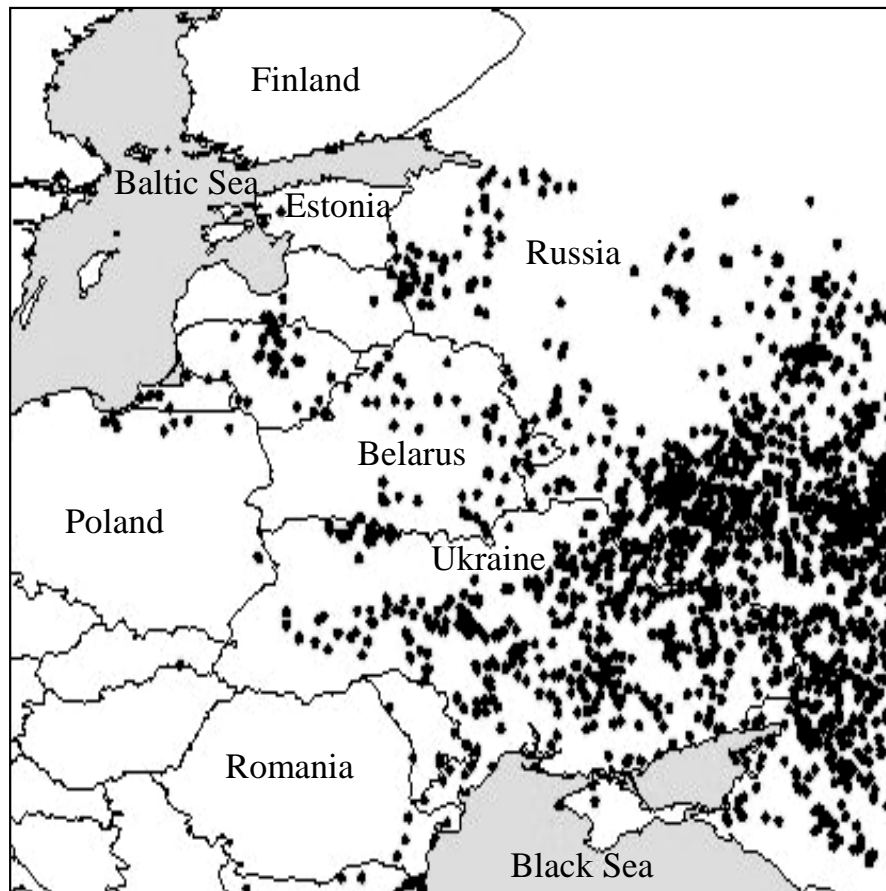


Fig. 4. MODIS fire detections in Eastern Europe during EPI-1 (10–15 August 2002).

NAAPS Surface Concentration ($\mu\text{g}\cdot\text{m}^{-3}$)
for 06:00Z 06 Sep 2002 Smoke

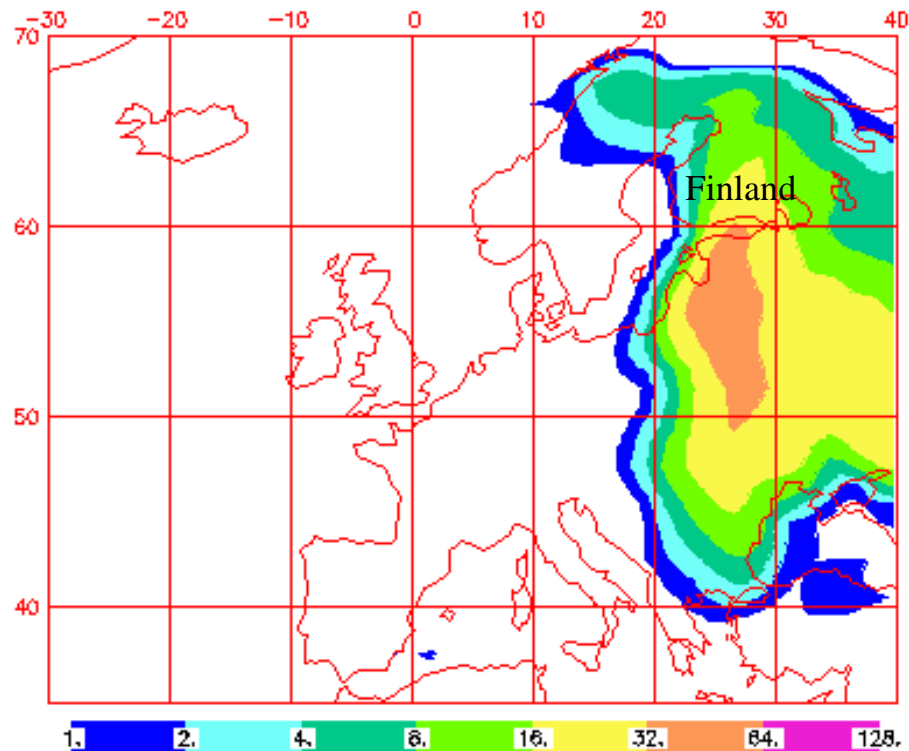


Fig. 5. NAAPS model results showing surface smoke concentrations for the strongest stage of EPI-3 (6 September 2002).

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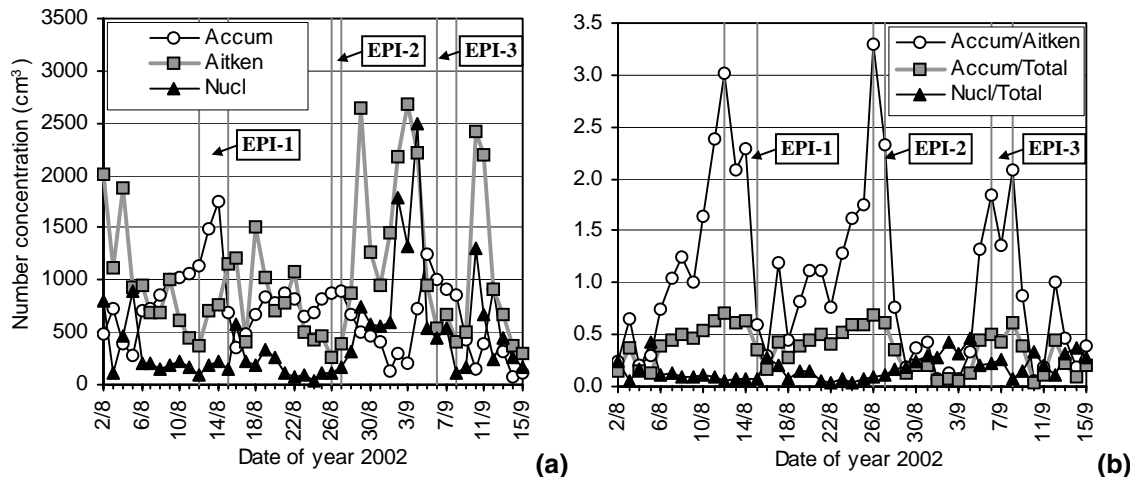


Fig. 6. (a) 24-h mean number concentrations of nucleation mode (Nucl, diameter 3–25 nm), Aitken mode (Aitken, 25–90 nm) and accumulation mode (Accum, 90–500 nm) particles, and (b) selected ratios of different modes and total amount (3–500 nm) of particles in Hyytiälä on 2 August–15 September 2002. Sample change time was at 05:00 (UTC).

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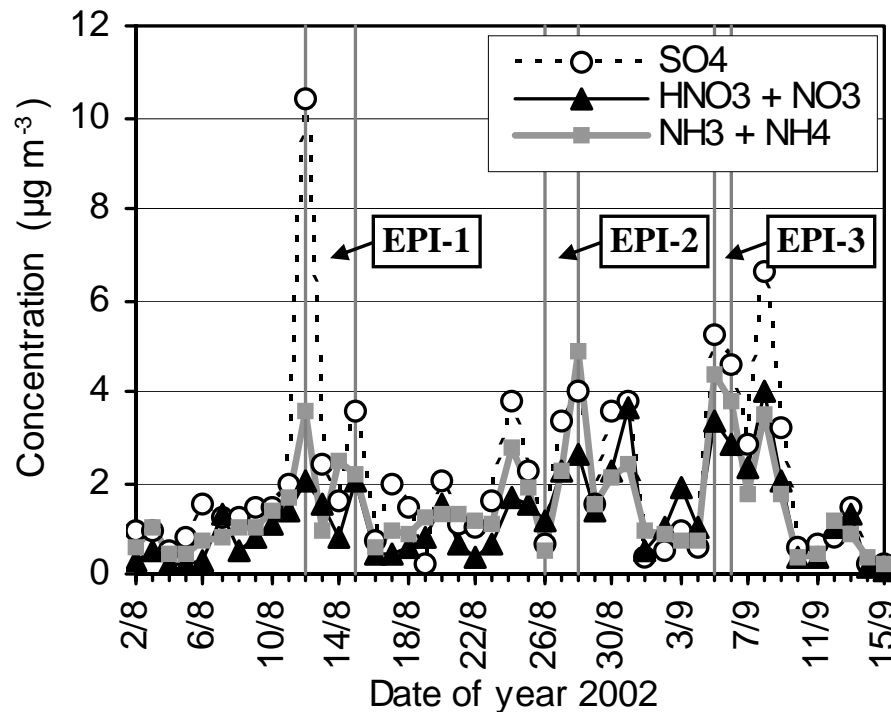


Fig. 7. 24-h mean total suspended particle (TSP) concentrations of SO_4^{2-} , total nitrate ($\text{NO}_3^- + \text{HNO}_3(\text{g})$), total ammonium ($\text{NH}_4^+ + \text{NH}_3(\text{g})$) in Virolahti on 2 August–15 September 2002. Sample change time was at 06:00 (UTC).

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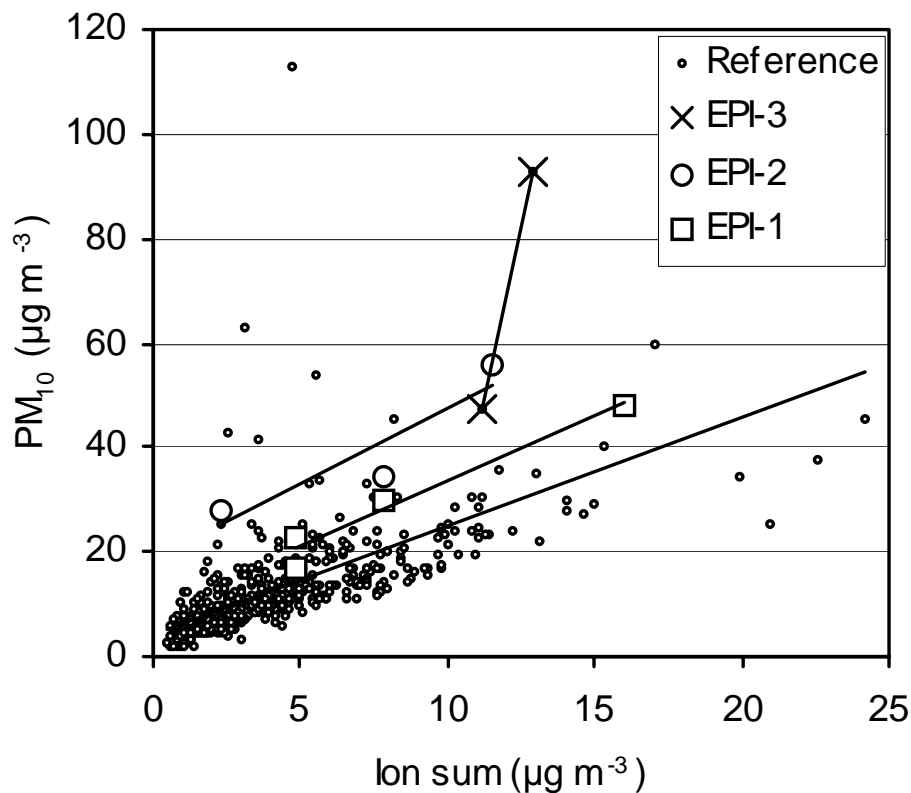


Fig. 8. Scatter plot and linear trend lines between 24-h mean ion sum and PM₁₀ concentrations in Virolahti during EPI-1 (12–15 August 2002), EPI-2 (26–28 August 2002), EPI-3 (5–6 September 2002) and reference days (3 July 2002–31 December 2003).

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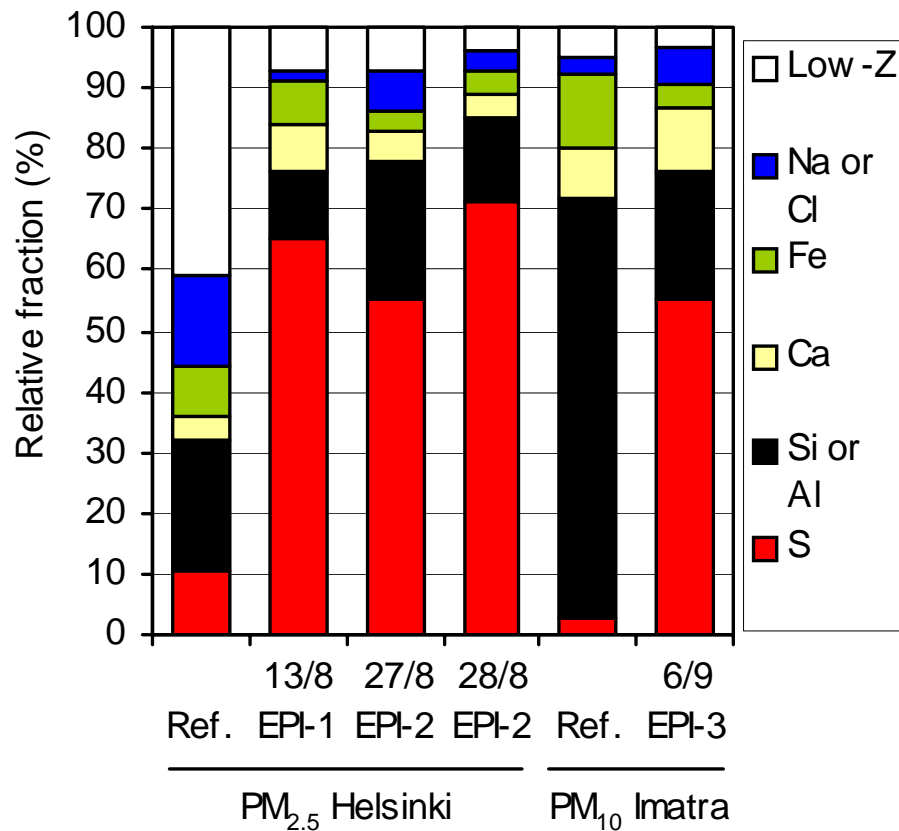


Fig. 9. Abundance of particle groups (%) in reference samples and episode samples collected at Kallio station in Helsinki during EPI-1 (13 August 2002) and EPI-2 (27 August and 28 August 2002) and at Mansikkala station in Imatra during EPI-3 (6 September 2002).

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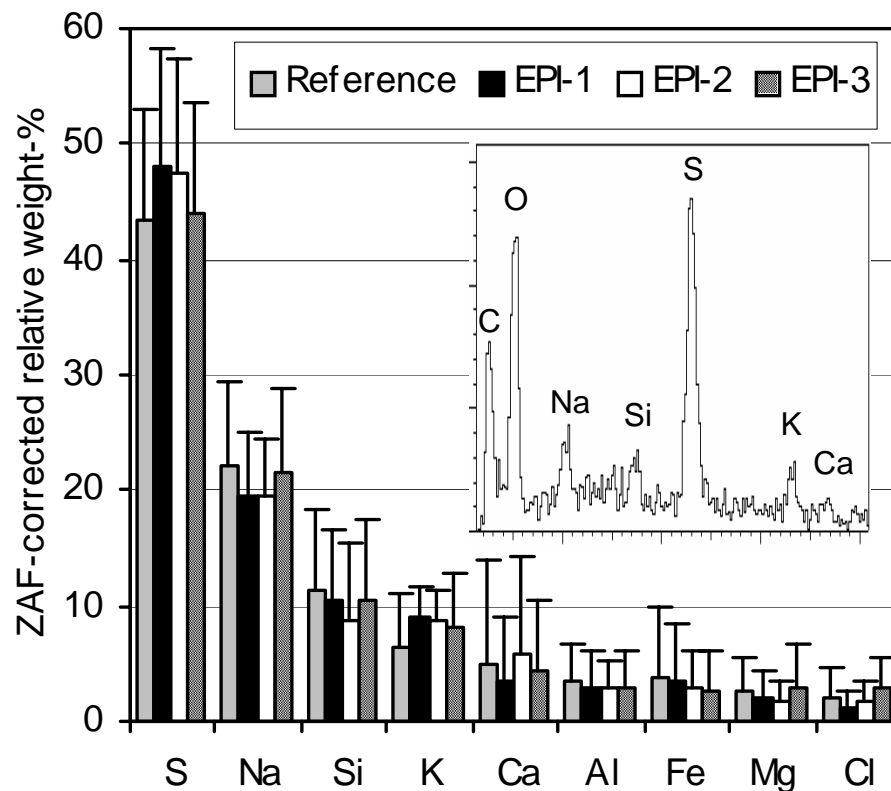


Fig. 10. Average weight percentages of selected elements in S-rich particles (typical SEM/EDX spectrum also shown) in $PM_{2.5}$ samples collected at Kallio in Helsinki during reference days, EPI-1 and EPI-2, and in PM_{10} samples collected in Helsinki and in Imatra during EPI-3. Error bars represent standard deviation (SD). Results are semiquantitative and weight percentages are normalized to 100%.

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