Aerosol measurements and modeling
Some recent advances in Finland

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CONTENTS

The spatial and temporal variation of measured urban PM mass in the Helsinki Metropolitan Area in 1997-99

[Pohjola, M A, Kousa, A, Kukkonen, J, Härkönen, J, Karppinen, A, Aarnio, P, Koskentalo, T, 2002. The Spatial and Temporal Variation of Measured Urban PM10 and PM2.5 concentrations in the Helsinki Metropolitan Area. International journal on Water, Air and Soil Pollution: Focus 2 (5-6), pp. 189-201.]

Measurements

Figure 1 shows the location of the air quality monitoring stations in the Helsinki metropolitan area and the pollutants monitored in 1999. The network contains six permanent multicomponent stations; these are located in Helsinki city districts (Töölö, Vallila and Kallio 2), in suburban in the cities of Espoo and Vantaa centres (Leppävaara and Tikkurila), and in a rural area in Espoo (Luukki). The stations used in this study represent urban (Töölö and Vallila) and suburban traffic environments (Leppävaara), together with the urban background (Kallio 2). Regional background concentrations were monitored in a rural environment in Luukki, approximately 20 km to the north-west of downtown Helsinki. Two urban monitoring stations, Töölö and Vallila, are located in the Helsinki downtown area.

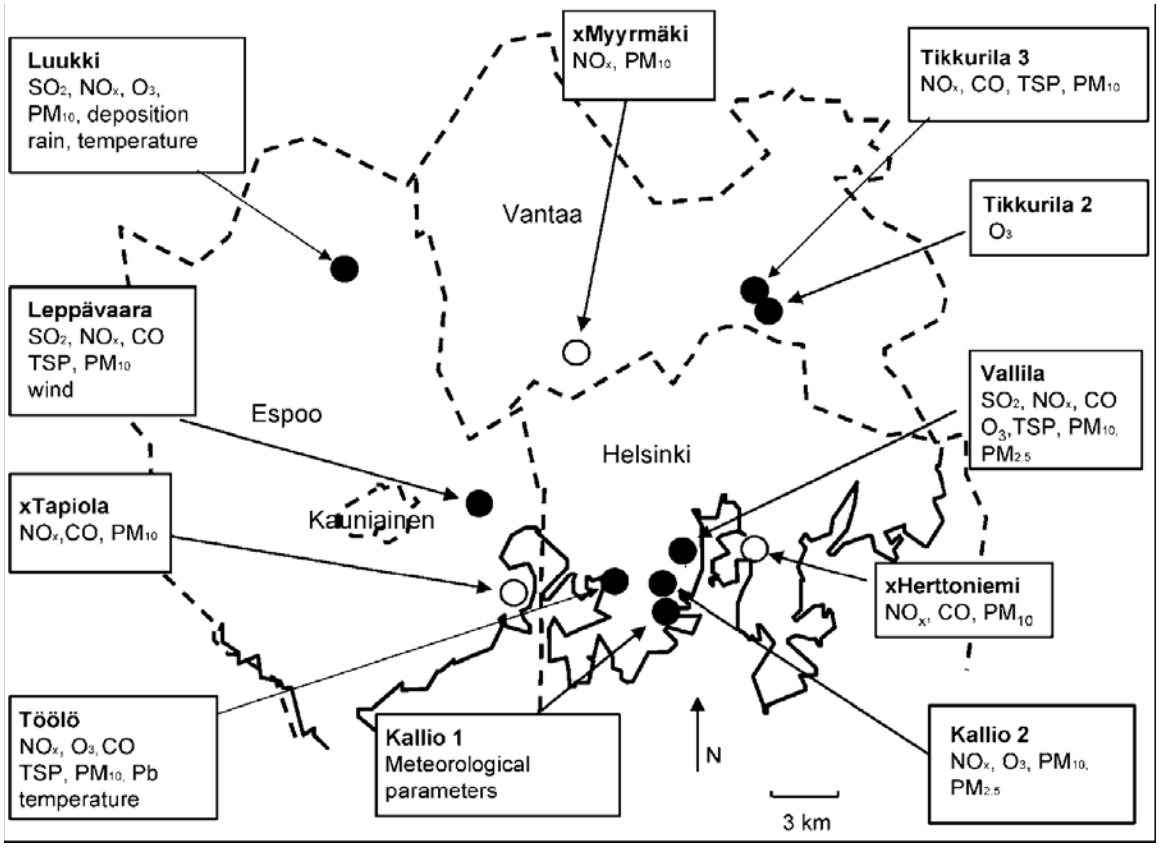

Figure 1. Urban air quality monitoring network in the Helsinki Metropolitan Area in 1999. The legends show the name of the station and the pollutants measured. X indicates a mobile station.

Diurnal variation of PM concentrations

Data has been compiled on the diurnal variation of the PM10 and PM2.5 concentrations at the various monitoring stations during 1997–1999. As an example, we have presented the diurnal variation of PM10 concentrations at the various stations in Figures 2a–c, averaged over each of the years 1997–1999, and PM2.5 concentrations at Vallila averaged over the same years and additionally at Kallio 2 in 1999 in Figures 3a–b. Clearly, the diurnal variation of traffic-originated pollutant concentrations depends on the day of the week; for these figures we have selected data from working days (Monday–Friday) only. The concentrations of PM10 show a clear diurnal variation. The concentrations increase continuously during the morning rush hours, from approximately 6 to 8 a.m.; as expected, this increase took place irrespective of the year (during the period 1997–1999) and the season of the year. Subsequently the concentrations decrease slowly during the rest of the daytime hours, also showing in some cases peak values during the afternoon rush hours, from approximately 3 to 6 p.m. The more moderate diurnal variation of the PM10 concentrations, compared with the traffic flows, could be caused by resuspension of PM from street surfaces. Resuspended PM increases with increasing traffic flow; however, it can reach a saturated state, in which a further increase of traffic cannot cause any more resuspension.

During working days, there is a very clear diurnal variation of local vehicular traffic. Despite this, the PM2.5 concentrations are temporally fairly uniform during working days, except for a moderate increase during the morning rush hours. The diurnal variation of local vehicular traffic flows seems to have no substantial correlation with the PM2.5 concentrations. In 1999, the temporal variation of PM2.5 concentrations at both monitoring stations was also very similar. This indicates that the PM resuspended from street surfaces and other sources has only a minor effect on the PM2.5 concentrations, and that a large fraction of the PM2.5 concentrations most likely originates from regional or long-range scale sources.

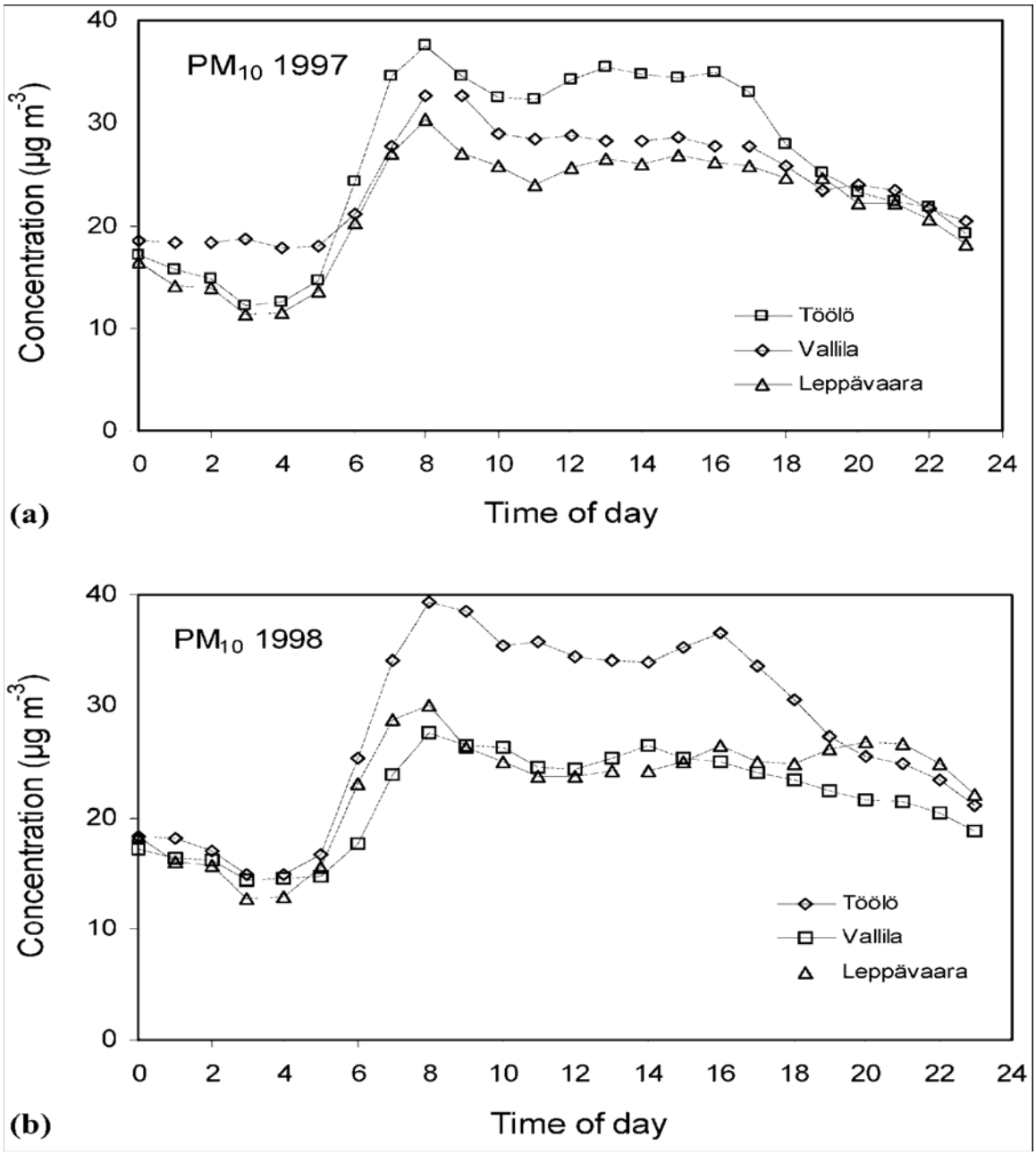

Figure 2. The diurnal variation of PM10 concentrations at the stations of Töölö, Vallila and Leppävaara during 1997–1999, and at the station of Luukki, during 1999. Figures include data from working days only.

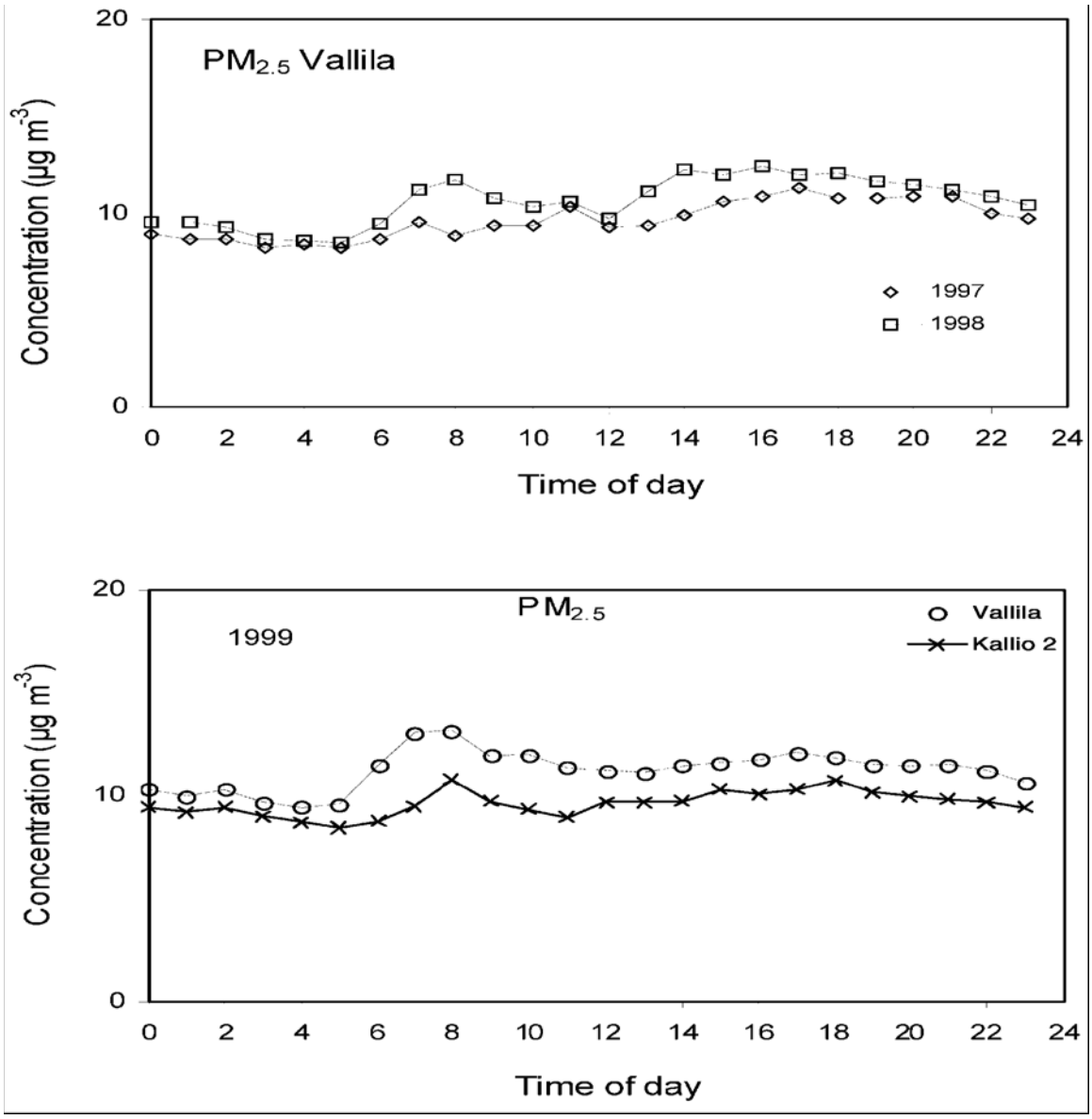

Figure 3. The diurnal variation of PM2.5 concentrations at the station of Vallila during 1997–1998, and at the stations of Vallila and Kallio 2 during 1999. Figures include data from working days only.

Conclusions

The diurnal variation of the PM10 concentrations was clear, irrespective of the year and the season of the year. This variation partly follows the corresponding variation of local vehicular traffic flows. On the other hand, both the spatial and temporal variation of the fine particle (PM2.5) concentrations was moderate. The results provide indirect evidence indicating that the PM10 concentrations are originated mainly from local vehicular traffic (direct emissions and resuspension), while the PM2.5 concentrations are mostly of regionally and long-range transported origin. This result is qualitatively in agreement with source apportionment studies in the same area

Analysis and evaluation of selected PM10 pollution episodes in the Helsinki Metropolitan Area in 2002,
[Päivi Aarnio, Jyrki Martikainen, Tareq Hussein, Ilkka Valkama, Hanna Vehkamäki, Larisa Sogacheva, Jari Härkönen, Ari Karppinen, Tarja Koskentalo, Jaakko Kukkonen, Markku Kulmala, 2007. Atm. Env., doi:10.1016/j.atmosenv.2007.02.008]

We developed methods to distinguish the long-range transport (LRT) episodes from local pollution (LP) episodes. The first method is based on particle number concentrations ratio between accumulation mode (diameter >90 nm) and Aitken mode (diameter 25–90 nm). The second method is based on a proxy variable (interpolated ion sum) for long-range transported PM2.5. The ion-sum is available from the measurements of sulphate, nitrate and ammonium at the nearest EMEP stations. We also utilised synoptic meteorological weather charts, locally measured meteorological data, and air mass back-trajectories to support the evaluation of these methods.

We selected nine time periods (i.e. episodes) with daily average PM10 > 50 μgm^{-3} in the Helsinki Metropolitan Area during year 2002. We characterized the episodes in terms of PM10 and PM2.5 concentrations and the fraction of fine particles in PM10 at an urban traffic and regional background air quality monitoring sites. Three of these episodes were clearly of local origin. They were characterized by a low average fraction of PM2.5 (<0.2) in PM10 at the urban traffic monitoring site, low ratio between PM10 concentrations at the regional background site and at the urban traffic site (<0.2), low average ion sums (1.5–2.5 μgm^{-3}) and low accumulation to Aitken mode ratios (0.13–0.26). Four of the episodes had distinct LRT characteristics: a high fraction of fine particles in PM10 (0.5–0.6) at the urban traffic site, a high ratio between PM10 concentrations at the regional background site and at the urban traffic site (0.7–0.8), high interpolated values for the ion sum (6.6–11.9 μgm^{-3}), and high accumulation to Aitken mode ratios (0.75–0.85). During the remaining two episodes there was significant contribution from both local sources and LRT. A detailed analysis of meteorological variables and air mass back-trajectories gave support to these findings.

PM and NO concentrations at the air quality monitoring sites in the Helsinki Metropolitan Area in 2002

Station	Station classification	PM_{10} median ($\mu g\,m^{-3}$)	PM_{10} 24 h max ($\mu g\,m^{-3}$)	$PM_{10} > 50$ ($\mu g\,m^{-3}$) no of days	$PM_{2.5}$ median ($\mu g\,m^{-3}$)	$PM_{2.5}$ 24 h max ($\mu g\,m^{-3}$)	$PM_{2.5}/PM_{10}$ median ($\mu g\,m^{-3}$)	NO median ($\mu g\,m^{-3}$)
Töölö	Urban traffic	19	97	32				35
Vallila	Urban traffic	18	137	19	8	52	0.48	11
Kallio	Urban background	14	85	10	7	43	0.57	4
Leppävaara	Suburban traffic	17	144	27				10
Tikkurila	Suburban traffic	17	118	22				23
Luukki	Regional background	10	58	2				1

These characteristics can be utilised in a simple procedure to distinguish between LRT and LP episodes. Further quantitative investigations to these characteristics provide an indication to the episode strength. There was a good correlation between the 24-h

averages of PM2.5/PM10 at the urban traffic site (Vallila) and the accumulation to Aitken mode ratio at the urban background site (Kumpula). There was a moderate to good correlation also between PM2.5/PM10 ratio and the interpolated ion sum, and between the ion sum and the accumulation to Aitken mode ratio. The scatter plots for these correlations are shown in Figure 1, which also show the grouping of the different types of episodes. The quantitative results presented in the current study are applicable to the Helsinki Metropolitan Area and similar cities. Nevertheless, developing these methods for other cities require analyses of the meteorological conditions, behavior of the PM concentrations, and air-mass back trajectories for that specific city.

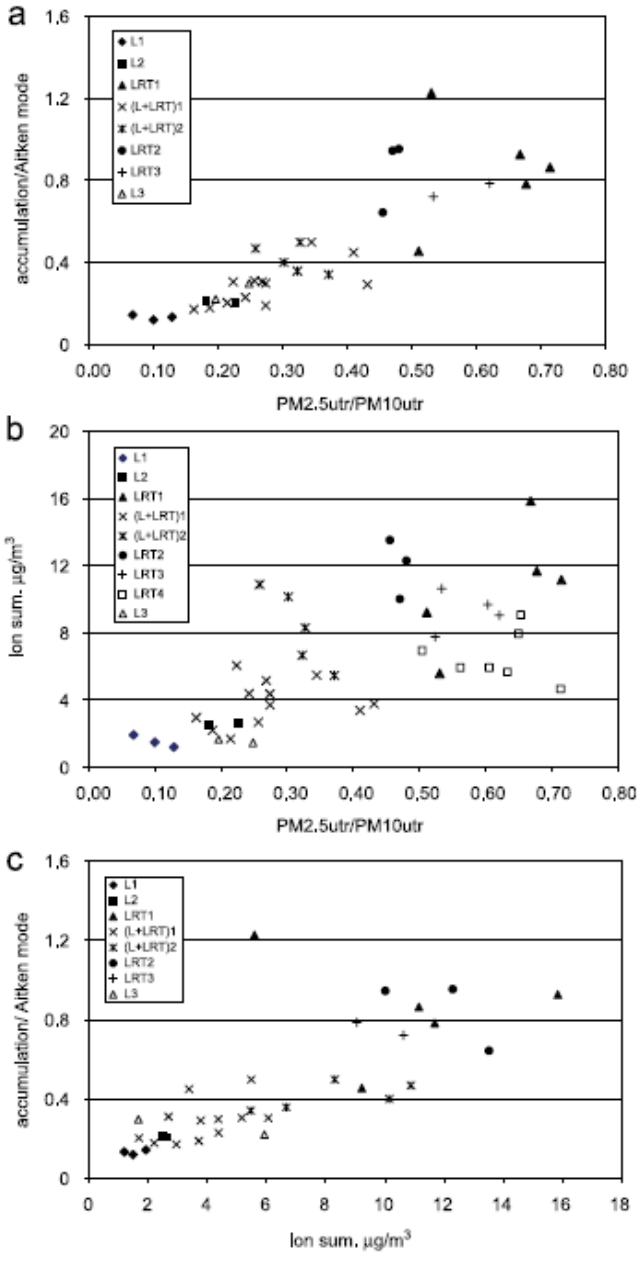

Figure 1. a) The PM2.5/PM10 ratios at the urban traffic monitoring site vs. accumulation to Aitken mode ratios measured at the urban background site.

(b) PM2.5/PM10 ratios at the urban traffic monitoring site vs. the ion sums.

(c) ion sums vs. the accumulation to Aitken mode ratio

Temporal and spatial patterns of PM mass in Finland

[Pia Anttila and Timo Salmi, 2006. Characterizing temporal and spatial patterns of urban PM_{10} using six years of Finnish monitoring data. *Boreal Env. Res.* 11: 463–479]

Six years (1998-2003) of continuous PM_{10} mass concentration measurements from 25 stations in 20 cities in different parts of Finland were used to examine the temporal and spatial patterns of urban PM_{10} in Finland (Anttila and Salmi 2006). PM_{10} data was extracted from the Finnish Meteorological Institute's Air Quality Monitoring Data Management System (ILSE). PM_{10} was measured with automatic analyzers based either on the tapered element oscillating microbalance or the beta-attenuation method.

Long term means of PM_{10} at twenty five Finnish urban stations vary between 11-24 μg m^{-3}, the highly trafficked urban centers tending to have higher concentrations than the suburban stations or small towns (Fig. 1). Year to year variation at each station is very low, typically only 2-4 μg m^{-3}. The national background concentration was estimated to be about 5 μg m^{-3}, and there is some indication that when traffic influence is eliminated a decreasing trend of PM_{10} from south/southeast to north emerges.

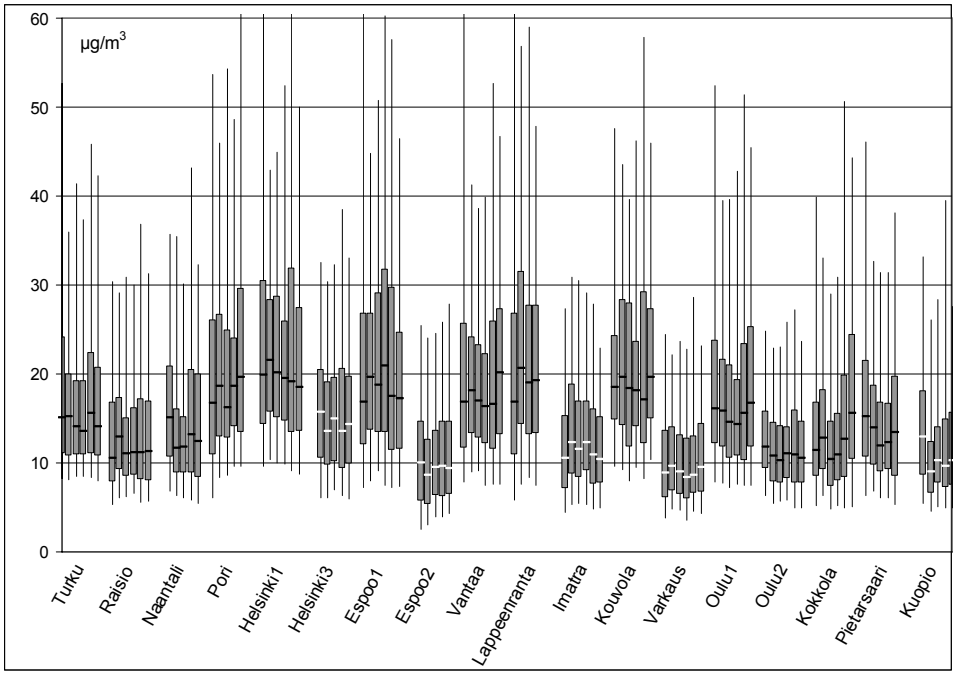

Figure 1 Annual distribution of 24-hr average PM_{10} concentrations from 1998 to 2003 at 18 stations. Values for the 5^{th}, 25^{th}, 50^{th}, 75^{th} and 95^{th} percentiles are shown. At the non-traffic stations the medians are denoted with white horizontal lines.

The seasonal variation of PM_{10} at all stations is dominated by the maximum during spring; in March-April the PM_{10} concentrations are about twice as high as during the rest of the year. This spring peak of PM_{10} covers practically the whole snowmelt period, which also is the driest period of the year.

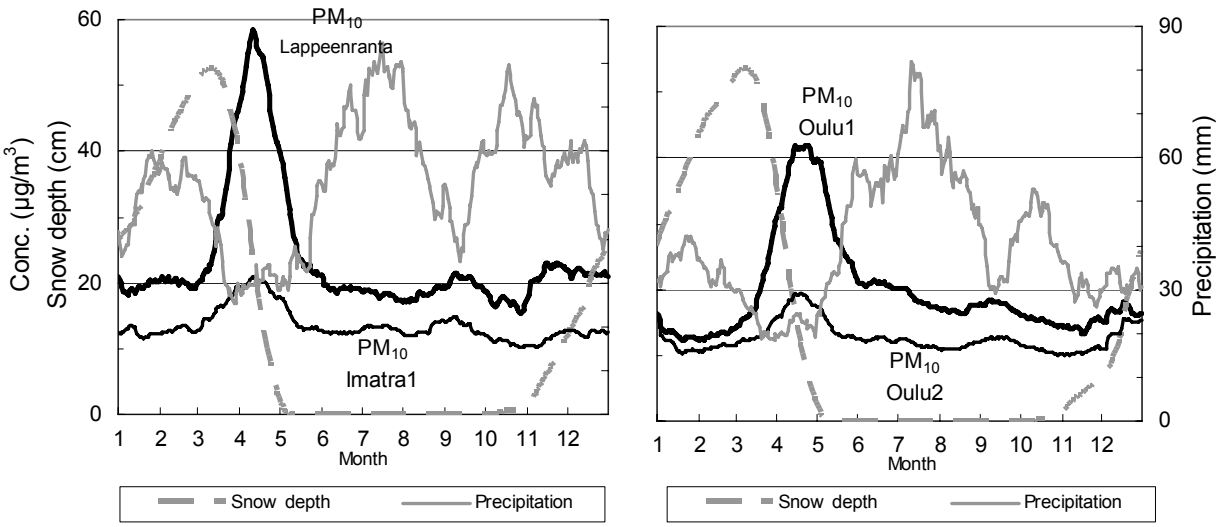

Figure 2. Annual variation of the PM_{10} concentration (black lines, left axis) and snow depth (grey dashed line, left axis) and precipitation (grey line, right axis) averaged over years 1998-2003 in southwestern Finland (Lappeenranta and Imatra1) and norhern Finland (Oulu1 and Oulu2). Meteorological data are from Lappeenranta and Oulu airport synoptic stations.

In spring also the Sunday concentrations at all urban stations are significantly lower (from 13% to 40%) than the weekday concentrations which implies a strong traffic influence on the PM_{10} concentrations. The spring time dry period with increasing temperatures, radiation and evaporation enables the effective suspension of the dust accumulated from multiple sources to road surfaces and shoulders and initiates the elevation of PM_{10} concentrations. However the highly synchronized day to day variation at a variety of sites across the country highlights the role of large scale weather patterns also in the formation of spring episodes. In March and April the exceedances of 50 $\mu g/m^3$ as a daily mean are frequent at almost all urban stations, however the EU limit value (35 exceedances) is exceeded only at the most trafficked street canyons in Helsinki.

Every year, most often in August, September and October, there are also 1-5 irregular regional PM_{10} episodes, lasting from one day to six days and being most likely originated from long range transported particles. During these regional events the PM_{10} concentrations may well reach the typical spring peak concentration levels. Similar regional LTR-events are probable also in spring but they get masked behind the overwhelming road influence.

Long term fine particle ($PM_{2.5}$) mass measurements are still scarce in Finland. Longest time series are from Helsinki metropolitan area and Oulu (since 2002) and from the FMI background station Virolahti (since 2004). Figure 3 shows the monthly means of $PM_{2.5}$ from these stations during 2002-2005 together with the co-located PM_{10} measurements. At the Virolahti rural background station the distinctive spring maximum of the PM_{10} is missing and the ratio $PM_{2.5}$ to PM_{10} typically varies between 0.6 to 0.8 being highest in early spring February and March. At the urban stations the ratio of $PM_{2.5}$ to PM_{10} drops

to 0.2 in spring being in the midwinter at its highest 0.7. From site to site the fine particle concentrations vary much lesser than the thoracic particles. The study period (Fig.3) mean of the fine particles in Virolahti is 7.3 µg/m^3 and in Oulu 8.5 µg/m^3.

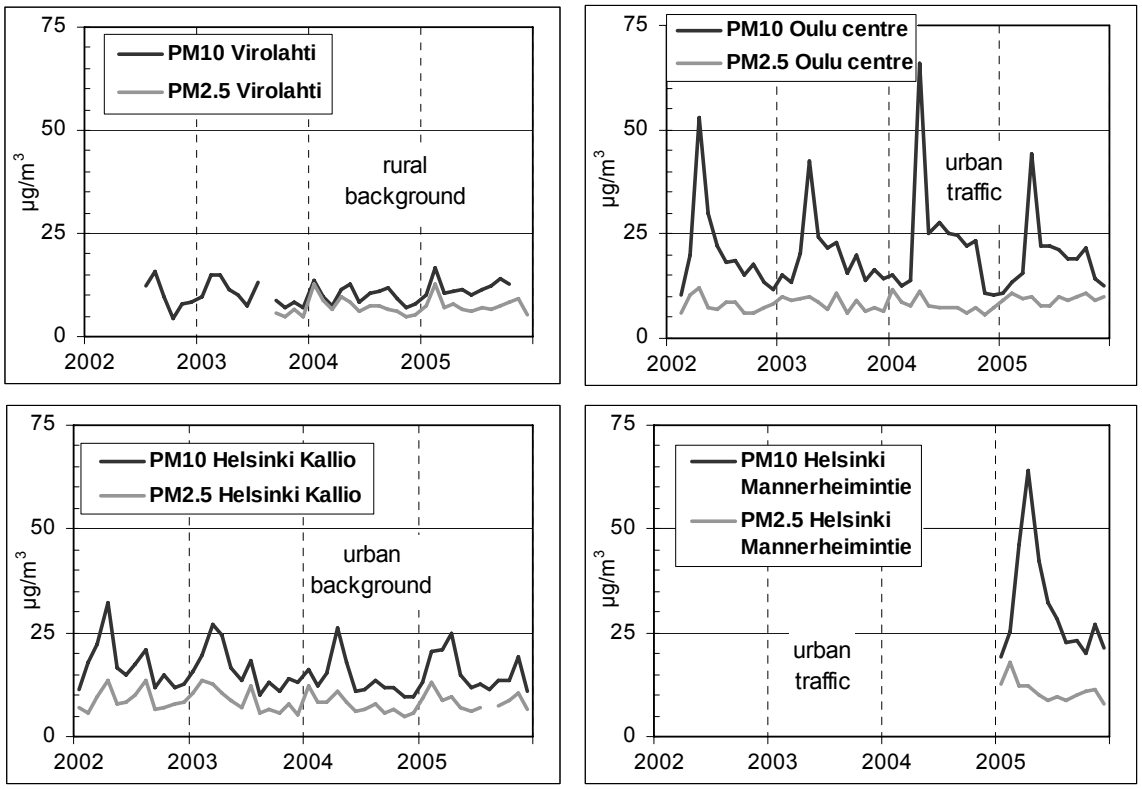

Figure 3. Monthly means of PM_{10} and $PM_{2.5}$ masses at different types of monitoring stations in 2002-2005.

MODELING FINE PARTICLE CONCENTRATIONS IN HELSINKI METROPOLITAN AREA

[Kauhaniemi, M., Karppinen, A., Härkönen, J., Kousa, A ., Koskentalo, T., Aarnio, P. and Kukkonen, J., 2007. Refinement And Statistical Evaluation Of A Modelling System For Predicting Fine Particle Concentrations In Urban Areas. In: Ranjeet S. Sokhi and Marina Neophytou (eds): Proceedings of the 6th International Conference on Urban Air Quality, Limassol, Cyprus, 27-29 March 2007, CD-disk: ISBN 978-1-905313-46-4, University of Hertfordshire and University of Cyprus (pp. 68-71)]

We present a combined modelling system that addresses particulate matter on an urban scale and long-range transported aerosols (LRT). We have evaluated the advantages and limitations of the statistical model (Karppinen et al., 2004) for computing the LRT'ed contribution to $PM_{2.5}$ in U.K. and in Finland (Kukkonen et al., 2007). We also aim to evaluate the performance of the combined modelling system against measured $PM_{2.5}$ data in Helsinki.

Meteorological and Air Quality Measurements

The relevant meteorological parameters for the models are evaluated using data produced by a meteorological pre-processing model (Karppinen et al., 1997, 1998). The location of the study area, the meteorological stations and the background air quality measurement stations are presented in Figures 1a and b. We used a combination of the data from the stations at Helsinki-Vantaa airport and Helsinki-Isosaari. The mixing height of the atmospheric boundary layer was evaluated using the meteorological pre-processor, based on the sounding observations at Jokioinen (90 km northwest) and the routine meteorological observations.

The EMEP stations ('Co-operative programme for monitoring and evaluating of the long-range transmission of air pollutants in Europe') that are located nearest to Helsinki are shown in Figure 1a; these are Utö, Ähtäri and Virolahti. The following concentrations are measured daily at the EMEP stations: (i) SO_4^{2-} (sulphate), (ii) the sum of NO_3^- (nitrate) and HNO_3 (nitrogen acid), and (iii) the sum of NH_4^+ (ammonium) and NH_3 (ammonia). The fine particle ($PM_{2.5}$) measurements of the Helsinki Metropolitan Area Council (YTV) monitoring stations at Vallila and Kallio were used. The location of the YTV monitoring stations in Helsinki metropolitan area is shown in Figure 1b. Monitoring station of Vallila represents urban roadside conditions. The Kallio station is an urban background monitoring station.

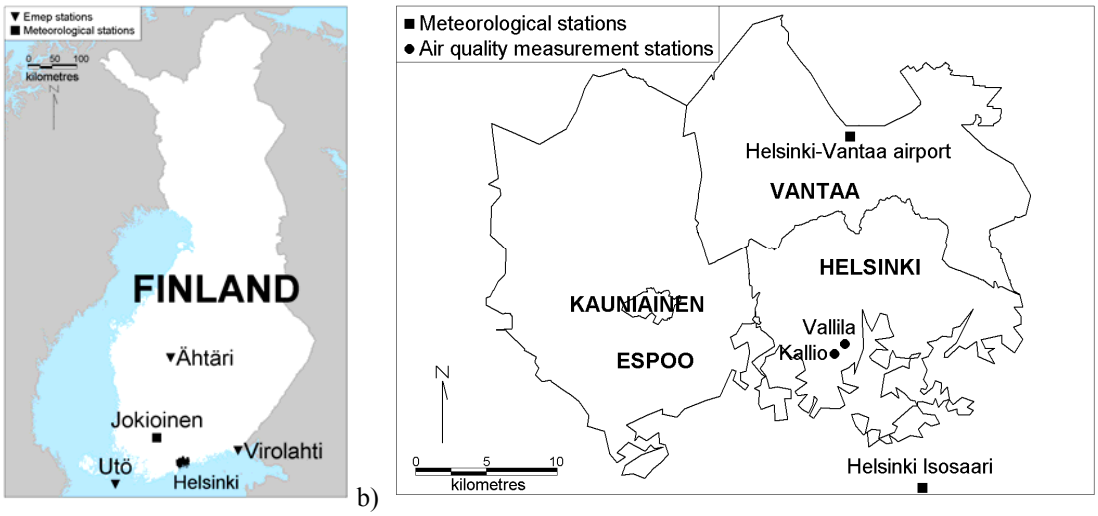

Figure 1. The meteorological and air quality monitoring stations in the Helsinki metropolitan area in 2002 (edited YTV, 2003).

Modelling System

The atmospheric dispersion of vehicular emissions is evaluated using a roadside dispersion model, CAR-FMI (Härkönen, 2002). The dispersion equation is based on an analytic solution of the Gaussian dilution equation for a finite line source. The dispersion parameters are modelled as function of the Monin-Obukhov length, the friction velocity and the mixing height. Traffic-originated turbulence is modelled with a semi-empirical treatment. The model includes the basic reactions of nitrogen oxides, oxygen and ozone, and the dry deposition of the fine particles. The model also takes into account the effect of the non-exhaust vehicular emissions and particulate matter suspended from the street surfaces, using empirical correlations.

The long-range transported contribution to urban particulate matter was evaluated with a statistical model (Kukkonen et al., 2007) that utilises, as input values, the daily sulphate, nitrate and ammonium ion concentrations measured at the EMEP stations. Currently, there is also an option to use representative regional background $PM_{2.5}$ concentration measurements in the Helsinki Metropolitan area. The regional background $PM_{2.5}$ concentrations can also be determined using the European-scale computations using the regional and continental-scale dispersion model SILAM (Sofiev et al, 2006).

Results

The mean, the maximum and the standard deviation, together with the statistical parameters for the predicted and observed daily average time series of $PM_{2.5}$ concentrations in 2002, for Vallila and Kallio monitoring stations are presented in Table 2.

Table 2. The statistical analysis of the predicted and observed daily average time series of $PM_{2.5}$ concentrations at the Vallila and Kallio monitoring stations for 2002 (the days 01.01.02 and 31.12.02 are omitted).

Statistical Parameter	Vallila		Kallio	
	Predicted	Observed	Predicted	Observed
Mean ($\mu g/m^3$)	11.2	9.90	7.92	8.64
Maximum ($\mu g/m^3$)	39.0	52.0	35.2	42.5
Standard deviation ($\mu g/m^3$)	5.31	6.75	4.80	5.89
Index of agreement (IA)	0.83		0.86	
Pearson's correlation coefficient (COR)	0.74		0.77	
Normalised mean square error (NMSE)	0.16		0.16	
Fractional bias (FB)	0.13		-0.09	
Number of data	360	360	358	358

At both stations the predicted $PM_{2.5}$ concentrations agree fairly well with the measured data. The model has slight over prediction for the station at Vallila and minor under prediction for the station at Kallio. The scatter plots of the predictions and observations at Vallila and Kallio in 2002 is presented in Figures 2a and b.

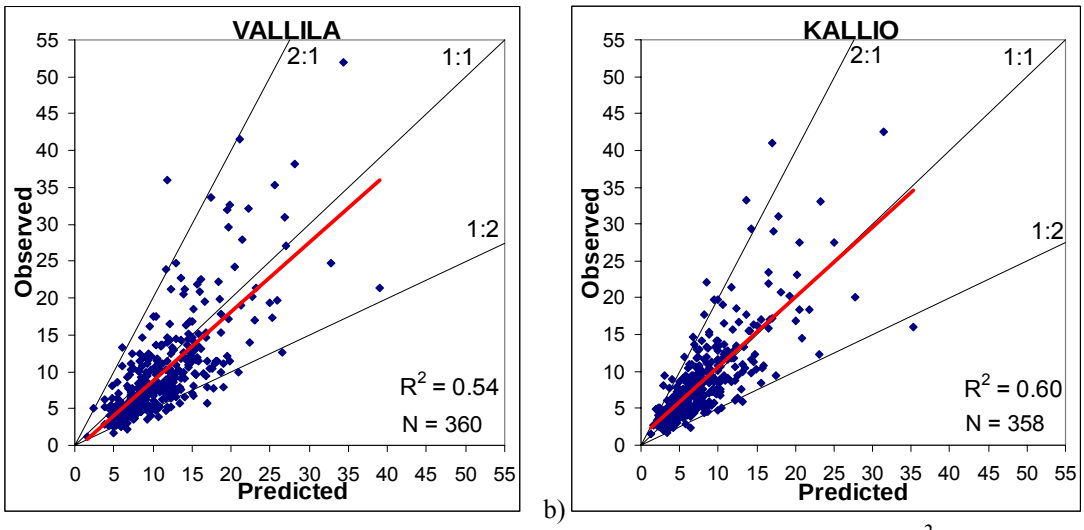

Figure 2. The scatter plots and the correlation coefficient squared (R^2) values of the predicted and observed daily average $PM_{2.5}$ concentrations in 2002, for the monitoring stations at Vallila (a) and Kallio (b).

Predicted total annual average concentration of $PM_{2.5}$ in the Helsinki metropolitan area and Helsinki city centre are presented in Figures 3a and b, respectively. On a yearly basis, the estimated contribution from regional and long-range transported origin to the observed $PM_{2.5}$ varies from 40 % at the most trafficked areas in Helsinki to nearly 100 % in the outskirts of the metropolitan area.

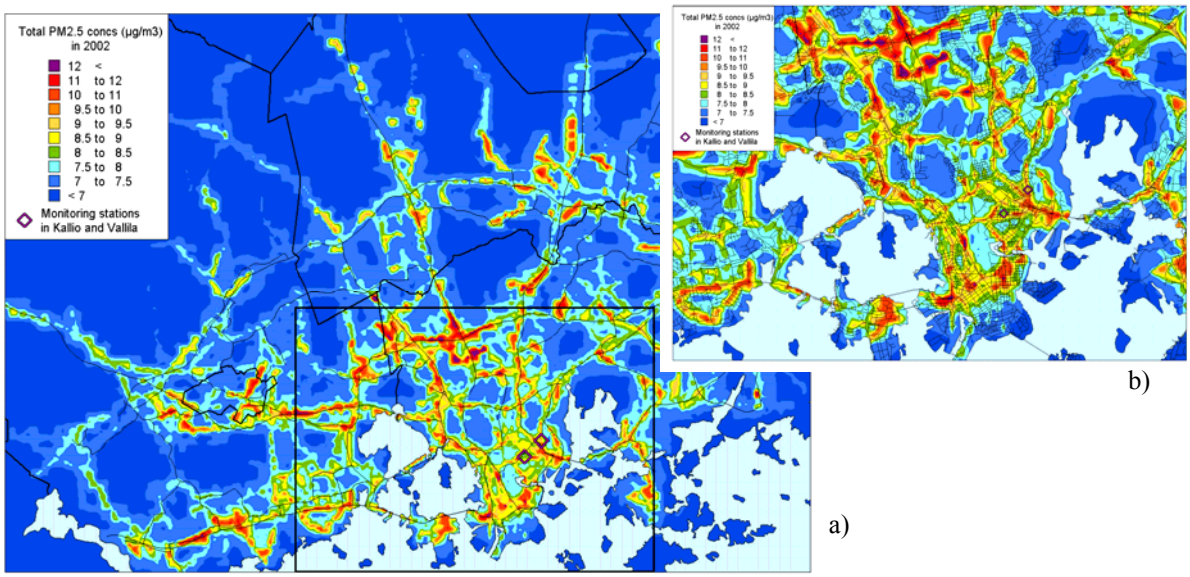

Figure 3. Predicted total annual average $PM_{2.5}$ concentrations [µg/m^3] in 2002, in the Helsinki metropolitan area (a) and in the centre of Helsinki (b). The main road and street network, and the monitoring stations of $PM_{2.5}$ are also presented in the figure.

Conclusions

The comparison of the modelled daily averaged values with the corresponding measurements showed a fairly good agreement. The results show that the statistical LRT model is a useful and simple tool for the assessment of LTR'ed $PM_{2.5}$ that is applicable within a fairly good accuracy. Clearly, the model also has inherent limitations. The accuracy of the model presented depends on the chemical composition of $PM_{2.5}$, especially the content of carbonaceous species; however, measurements of these species at the EMEP stations have not been published. The ion sum parameter defined also contains in part the measurements of two gaseous substances (HNO_3, NH_3). If their concentrations were high, compared to the concentrations of the corresponding compounds in particulate form, there could be substantial inaccuracies in the model predictions.

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Regional scale dispersion modeling of fine particulate matter (SILAM)

[Sofiev M., Jourden E., Pirjola L., Kangas L., Karvosenoja N., Karppinen, A. and Kukkonen J, 2007. Dispersion modelling of the concentrations of fine particulate matter in Europe, *Proceedings ot the 28th NATO/CCMS, ITM on Air pollution Modelling and its Applications,* May 15-19, 2006, Leipzig, Germany]

Introduction

The results presented are based on a research project "An integrated model for evaluating the emissions, atmospheric dispersion and risks caused by ambient air fine particulate matter" (KOPRA) of Finnish Finding Agency for Technology and Innovation (TEKES). An overall goal of the project was to evaluate the whole cycle of aerosol air pollution in Finland including emission of particulate matter and its pre-cursors, their dispersion, transformation and deposition, resulting contamination patterns and their impact to public health, as well as possible ways to reduce the aerosol atmospheric concentrations. In this paper, we concentrate of dispersion simulations performed with the Finnish emergency and air quality model SILAM at Finnish Meteorological Institute. The main goal of these simulations was to assess a link between the Finnish and European emissions of various gaseous and particulate species and resulting aerosol contamination of northern Europe. A specific goal was to build as complete budget for the aerosol composition over Finland as possible and compare the resulting bulk values with the observations. Separate verification was performed for some specific substances, such as aerosol precursors, primary PM, sea salt, etc.

Input data and modelling tools

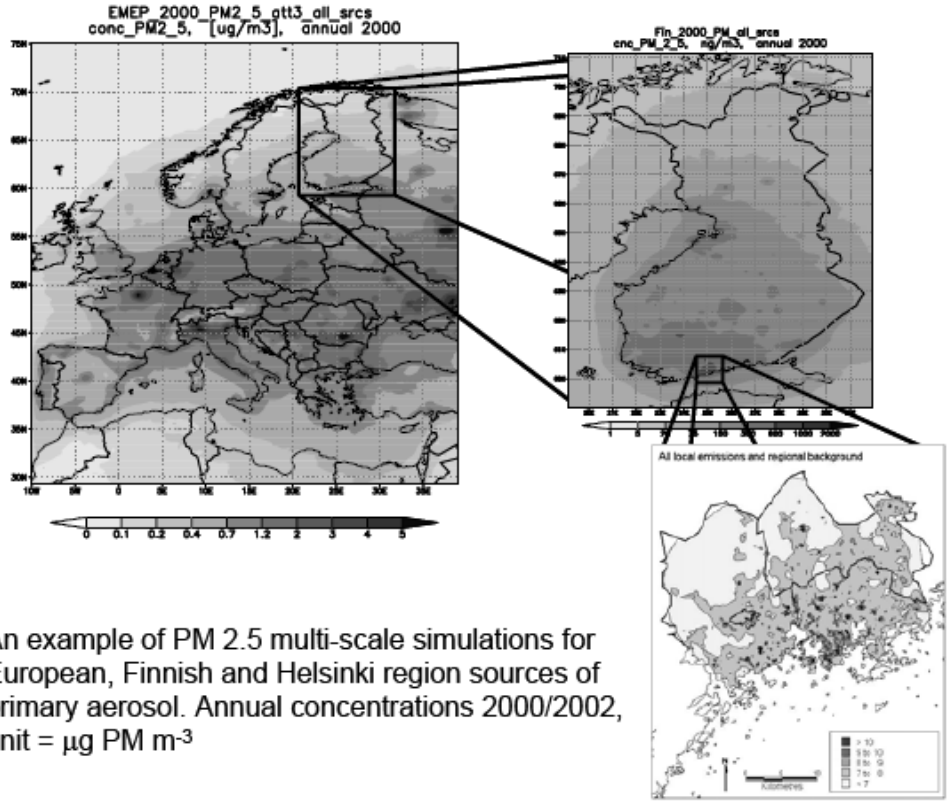
Input data for the simulations were combined from several sources. European-wide anthropogenic emission of particulate matter (with a split to PM 2.5 and PM 2.5-10), as well as of sulphur oxides, was adopted from the WebDab database of European Monitoring and Evaluation Programme EMEP (www.emep.int). The information was available at annual level and with spatial resolution of 50km. Temporal disaggregating to hourly fluxes was made on a country-by-country basis using the results of EUROTRACGENEMIS project (Lennart *et al.*, 1997). Vertical distribution of emission followed the EMEP methodology based on 11 emission source categories and characteristic injection heights for each of them (Simpson *et al*., 2003). The European information was complemented with a high-resolution Finnish national emission inventory with detailed chemical and size-segregation splits: emission of primary particles was considered for 5 size classes (PM 0.1, PM 0.1-1, PM 1-2.5, PM 2.5-10, PM 10-TPM) and included separate estimates for the following compounds: black carbon, organic carbon, dust, sulphates, and total PM. Emission of pre-cursors included SO2, NOx, NH3, and anthropogenic VOC. The dataset included spatially distributed emission with a resolution of 1 km and over 250 point sources with physical stack characteristics. Temporal disaggregating was performed following the same GENEMIS methodology. Vertical injection height for area sources was assumed to be within the lowest 100 m, while the plumes from point sources were parameterised using actual stack heights though made independent from actual meteorological conditions to reduce the computation costs. Meteorological information and necessary geophysical and land cover maps were taken from the FMI-HIRLAM and ECMWF meteorological models. All input

data covered the period of 2000-2002 and the simulations were also targeting this time interval. The main modelling tool used for regional- and meso-scale simulations was the Finnish Emergency and Qir Quality Modelling System SILAM (Sofiev *et al*, 2006). It is a lagrangian particle model with Monte-Carlo random-walk mechanism representing the vertical and horizontal turbulent diffusion. The system includes a sophisticated meteorological pre-processor for evaluation of basic features of boundary layer and free troposphere using the meteorological fields provided by NWP models. In implementation, SILAM assumes well-mixed boundary layer and fixed turbulent diffusion coefficients in free troposphere. Exchange between them is mainly driven by temporal variation of the top of boundary layer. A physico-chemical module of SILAM covers up to 496 radioactive nuclides, sulphur oxides, primary particles of various types as well as probability (used for evaluation of area of risk and for solution of inverse-problems). The system accepts an arbitrary definition of the particle size spectrum described in the current study via a set of bins. Chemical transformations of SOx follow the scheme of DMAT model (Sofiev, 2000). A local-scale model CAR-FMI (Kukkonen *et al*., 2001a,b) was used for evaluation of Helsinki city-scale pollution levels. Evaluation of the influence of aerosol dynamics is done using the aerosol dynamics model MONO32 (Pirjola and Kulmala, 2000; Pirjola *et al.*, 2003). MONO32 is a box model covering gas-phase chemistry and aerosol dynamics. The model uses monodisperse representation for particle size distribution with an optional number of size modes. In this work, five modes are used: nucleation, Aitken, accumulation, and two coarse modes. All particles in a certain mode are characterised by the same size and composition, and they can consist of sulphuric acid, ammonium sulphate, ammonium nitrate, organic carbon, elemental carbon, sodium chloride, and mineral dust. Water content of aerosols is calculated separately. Particles can be emitted as primary particles or formed in the atmosphere by nucleation. Size and composition of particles in any class can change due to multicomponent condensation of sulphuric acid and organic vapours as well as due to coagulation between particles. A specific model, simulation setup, and input data were needed for evaluation of the desert dust pollution. Due to highly episodic character of this phenomenon as well as its strong inter-annual variability, computations over a single or a few years would be insufficient to catch even an order of magnitude of its contribution to aerosol concentrations over Finland. Therefore, we utilised a simplified but computationally efficient model DMAT (Sofiev, 2000), which was forced by pre-processed NCAR 22-years long meteorological re-analysis over the Northern Hemisphere. More information on this study can be found in (Hongisto & Sofiev, 2004).

Results and discussion

Following the strategy outlines above, four sets of simulations have been performed: at the European-scale – for primary PM 2.5, PM 2.5-10, sea salt ,and SOx; at regional scale – for primary PM 0.1, PM 0.1-1, PM 1-2.5, PM 2.5-10, PM-coarse (over 10μm size), and SOx; for Helsinki area – PM 2.5, PM 10 and NOx; finally, the wind-blown dust was computed for the Northern Hemisphere. European-scale resolution was 30 km with daily averaging, regional simulations provided 5km daily output fields while hemispheric runs were made with 150km grid and provided monthly-mean values. The reference year was 2000; most of simulations were also performed for 2001 and 2002; hemispheric

simulations were made for the period 1967-1988 in order to obtain a climatologically representative dataset. There were also a few case studies in the adjacent years (1999, 2003) made for the periods of observational campaigns. An example of the simulation results is presented in Figure 1 for primary PM 2.5, which characteristic level in 2000 was about 1 μg m-3 over large areas of Europe with several highconcentration areas. This is in a good agreement with the aerosol model simulations by EMEP Western Centre (Kanhert & Tarrason (eds), 2003). However, nearly twice better spatial resolution of current simulations allowed for more detailed patterns over strongly polluted areas, such as Po Valley, Scandinavian capitals, etc. Improved resolution over Finland and, further, over Helsinki area also highlighted local-scale distributions – both as urban vs regional background (visible at 5-km map) and over specific parts of the city (local-scale map). It should be pointed out that the 5km map in Figure 1 is presented without the European background, which would bring the total level of primary PM 2.5 concentrations over southern Finland to the level of about 1-2 μg PM m-3. Direct comparison of primary particles with observations was not performed because in 2000 there were practically no representative observations resolving the chemical composition of aerosol and thus capable of separating the primary PM from secondary inorganic aerosol (sulphates, nitrates and ammonia), sea salt and mineral dust. The second part of aerosol budget constitutes from the secondary inorganic aerosol.



An example of PM 2.5 multi-scale simulations for European, Finnish and Helsinki region sources of primary aerosol. Annual concentrations 2000/2002, unit = μg PM m-3

Figure 1. An example of three-scale off-line nested simulations for primary PM 2.5.

Comparison of sulphates and SO2 with airborne measurements of e.g. EMEP network as well as with other models is quite straightforward. According to that, the SILAM

simulations have "standard" accuracy: the model tends to slightly underestimate sulphates being otherwise within a factor of two from the most of observations. Simulations for two more components of the atmospheric aerosol are – sea salt and windblown dust – are shown in Figure 2a and 2b, respectively. Near-surface concentrations of the wind-blown dust do not make much sense over Scandinavia because most of such aerosol flies over thousands of kilometres before reaching the region, which implies a wide and often uneven distribution along the vertical of the arrived masses. Therefore, the only representative parameter for that component is the vertically integrated column burden. Observations of the sea salt are quite scarce and can be performed either via comparison of vertically integrated aerosol optical depth observed by satellites or by comparing some chemical components specific for this type of aerosol, for example, Na+, which constitutes about 30% of the sea salt mass. Comparison with Na+ observations at Mace Head showed that the model tends to under-estimate the overall level of concentrations by a factor of 2-3, closely capturing the summer low-salt periods and being significantly lower than measurements over winter periods when strong storms inflate the salt concentrations by 3-5 times. This seasonality in the model is less pronounced. The present sea salt emission module in SILAM is based on improved scheme of Monahan *et* al (1986). It is known to somewhat under-produce the sub-micron particles, which have the longest transport distance. The other feature of the scheme – strong over-production of particles smaller than 0.05 μm was corrected during implementation. Finally, SILAM emission module neglects the mechanism of production of coarse particles (over 10 μm) as they have no impact on regional and long-range transport. However, they can still significantly affect observations at Mace Head.

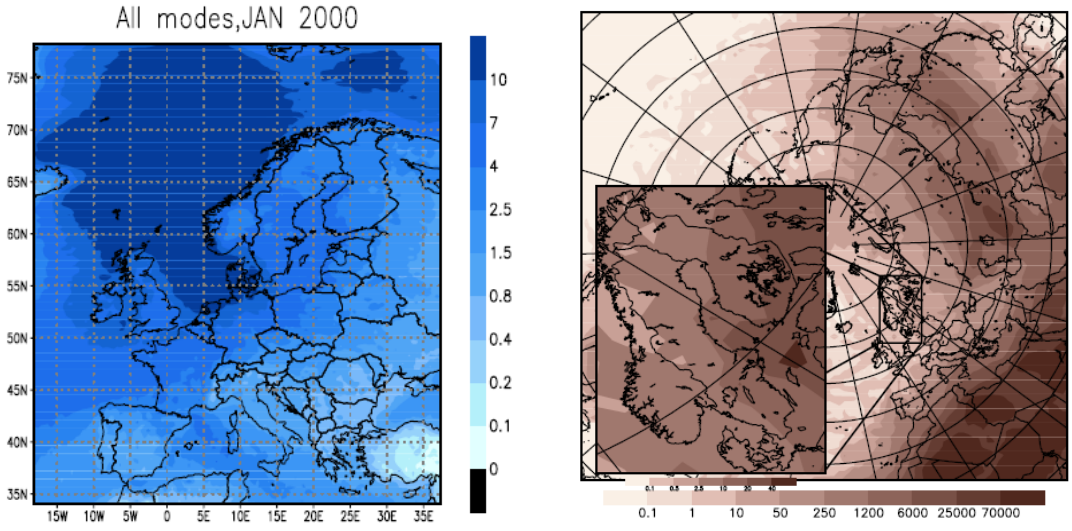


Figure 2 a) Mean Jan 2000 concentration of sea salt (all size classes). Unit μg PM m^{-3}. b) Mean 1967-1988 vertically integrated wind-blown dust concentration in air column. Unit: mg PM m^{-2}. Obs different scales!

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