COMBINATION OF A DISPERSION MODEL AND AN AEROSOL PROCESS MODEL FOR MODELLING ROADSIDE ENVIRONMENT PARTICLES, AND EVALUATION WITH MEASURED DATA

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INTRODUCTION

High number concentrations of fine and ultrafine particles in urban environments, especially in the vicinity of major streets and roads, have raised the interest to study the physical and chemical transformation of PM in the urban environment. This work is a continuation of our effort to analyse particle transformation and dilution in a local scale (Pohjola et al., 2003 and 2004). In this study we analyse fine and ultrafine particle transformation within the distance scale of 200 m (time scale of a couple of minutes) from a major road in Helsinki, and compare the model results with measurement data. The aerosol process model MONO32 is applied for the evaluation of the temporal evolution of number concentration, size distribution and chemical composition of various particle size classes. The dilution rate of particles is obtained from the roadside dispersion model CAR-FMI. Measurement data was obtained from a mobile laboratory campaign in city traffic conditions. In this study, we have focused solely on particles.

MATERIALS AND METHODS

The measuring locations and equipment

The aerosol measurements by a mobile laboratory have been conducted at various locations near Itäväylä, a major road in an urban area of Helsinki, during the LIPIKA campaign in February 17 – 20, 2003 (Pirjola et al., 2005). The mobile laboratory used was constructed into a Volkswagen LT35 diesel vehicle (Pirjola et al., 2004). Particle size distribution in the size range of 0.07 nm – 10 μm (aerodynamic diameter) with 12 channels is measured by the Electrical Low Pressure Impactor (ELPI, Dekati Ltd, Keskinen et al., 1992). The nucleation mode particle size distribution with a high size resolution is measured by the Hauke type Scanning Mobility Particle Sizer (SMPS), where particles are first neutralised, then classified by a DMA based on their electrical mobility, and counted by a CPC 3025 (TSI, Inc.). Measurement size range is 3 – 50 nm (mobility diameter), the number of channels is 20 and the scan-up time mostly 30 s. Additionally, the total number concentration of particles larger than 3 nm is detected by an ultrafine condensation particle counter CPC 3025 (TSI, Inc.). A passive clean air dilution system was installed, with a dilution ratio of 1:3. The SMPS and CPC instruments are operated by the University of Helsinki. The particle measurement inlet is located at the height of 2.4 m. The weather station is located at the roof of the van. Temperature and relative humidity are measured (Model HMP45A, Vaisala) at the height of 3.4 m from the ground. Additionally, a global position system (GPS V, Garmin) saves the van

speed and the driving route and a video camera in the cab records visually the traffic situations. The wind speed and direction used in the modelling was measured at 10 m height at the roof of a cabin located 9 m from the edge of the road.

Modelling methods

The CAR-FMI model includes an emission model, a dispersion model and statistical analysis of the computed time series of concentrations. The CAR-FMI model utilises the meteorological input data evaluated with the meteorological pre-processing model MPP-FMI. For a more detailed description of these models, the reader is referred to Härkönen et al. (2002) and Karppinen et al. (2000 a, b).

The aerosol dynamics model MONO32 is a box model, which includes gas-phase chemistry and aerosol dynamics, and can be applied under clear sky conditions. The model uses monodisperse representation for particle size distribution with an optional number of size modes. In this work we have used six modes: nucleation1 (diameters d<7,5nm), nucleation2 (diameters 7,5nm< d <43,2nm), Aitken (43,2nm <d<0,122μm), accumulation1 (0.122μm<d<0,321μm), accumulation2 (0,321μm<d<2,5μm), and coarse (d >2,5μm). All particles in a mode are characterised by the same size and the same composition. Particles can consist of soluble material such as sulphuric acid, ammonium sulphate, ammonium nitrate and sodium chloride, organic carbon which can be soluble, partly soluble or insoluble, and insoluble material like elemental carbon and mineral dust. 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. For a more detailed description of the MONO32 model and its evaluation against measurement data, the reader is referred to Pirjola et al. (2000, 2001, 2003) and Pohjola et al. (2003).

RESULTS AND DISCUSSION

Model computations

The meteorological data used was from the period February 17 – 20, 2003, at time periods when the wind direction was approximately perpendicular to the road, from the direction of the road towards the measurement sites.

It was found that a combination of number concentration and composition data from literature references was insufficient to describe the vehicular exhaust particles in Helsinki. Therefore we used the particle number concentrations measured in traffic during the campaign in Feb 17-20, 2003 at Itäväylä, combined with the chemical composition data extracted from Kauhaniemi (2003), Norbeck et al. (1998) and Shi et al. (2000). The properties of the particulate modes in urban background air are presented in Table 1.

According to Kauhaniemi (2003), in the Helsinki Metropolitan Area, light duty vehicles constitute about 90% of all traffic, 80% of which are petrol vehicles and 20% diesel light duty vehicles. It was assumed that all heavy duty vehicles are diesel operated. The diameters and the limits of the six modes as well as the percentile portions of the number concentrations in each mode were obtained from the average size distribution measured by the mobile laboratory at Itäväylä. These percentiles were used in calculating the modal emission factors from the emission factors of the total number concentration by Gidhagen et al. (2004) and Yli-Tuomi et al. (2004). The modal emission factors were extended to allow for the temporal variation by using the measured traffic density profiles.

Table 1. The properties of the urban background air particles.

Particle mode	size Dry radius (nm)	Number concen-tration (cm^{-3})	Total mass ($ng\ m^{-3}$)	Mass composition (%)					
				Sulphuric acid	Organic carbon	Elemental carbon	Mineral dust	Ammonium nitrate	Ammonium sulphate
Nucleation1	2.35	1.32e2	1.22e-2	5	90			5	
Nucleation2	9	7.75e3	4.02e+1	5	90			5	
Aitken	36.4	2.63e3	9.03e+2		27	64	7	2	0.3
Accumulation 1	103	5.66e2	4.34e+3		26	63	9	3	0.3
Accumulation 2	226	2.94e1	2.4e+3		25	63	9	3	0.3
Coarse	2020	1.7e1	1.0e+4				100		

We evaluated an average urban background particle size number distribution based on the LIPIKA-measurements at an urban background station, and chemical composition of the background particles was based on the works of Pakkanen et al.(2001 a, b) and Viidanoja et al. (2002) .

The width of the road Itäväylä is approximately 30 m, the width of the three lanes to each direction are 12 m altogether, and there is 6 m wide green belt (mainly grass) in the middle. We have assumed that the height of the initial dilution volume in the MONO32 model is 80 cm. This volume is by definition where exhaust gases have been diluted in a time scale of one second after their discharge from the tailpipe. The smallest size mode of the particles was assumed to be 4.7 nm; this indicates that nucleation of particles has already occurred, due to the temperature decrease immediately after the exhaust. On-road mixing by traffic-induced turbulence has also been omitted (Zhang et al., 2004). The vehicular emissions are assumed to continue as long as the computational volume containing the exhausts is transported and diluted downwind over the a distance of 12 m, after that distance, only transport and dilution occurs over 6 m, then again emission over 12 m and after that only transport and dilution. The numerical power functions for allowing for dilution were calculated for $PM_{2.5}$ by the FMI-CAR model; the road was modelled as a set of two line sources that were located in the middle of the three lanes in both directions. We have assumed that the dilution rate is the same also for the particle number concentrations. The emission and dilution rates were taken into account in the aerosol process model by appropriately modifying the differential equations of the number and mass concentrations of particles.

Comparison of modelled and measured concentrations

As an example, the comparison of modelled and measured total number concentration of particles smaller than 2.5 μm at 10 to 11 a.m. on February 18, 2003 is presented in Figure 1.

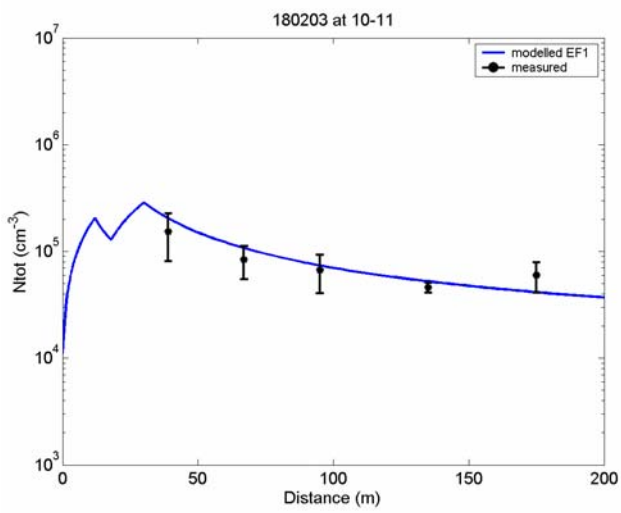

Figure 1. The modelled and measured total number concentrations of particles as a function of distance from the edge of the road Itäväylä at 10 to 11 a.m. on February 18, 2003. EF1 refers to emission factor by Gidhagen et al.(2004). The error bars show the estimated uncertainty of the experimental techniques.

We will also evaluate the evolution of particle size distribution and chemical modal composition, assuming various concentrations of condensable vapours.

ACKNOWLEDGEMENTS

This work has been funded by the Maj and Tor Nessling Foundation. We also wish to thank all the people in the LIPIKA project for their careful work in producing the mobile laboratory measurement data, the projects KOPRA, OSCAR and SAPPHIRE, and the CLEAR cluster of EU-funded projects.

REFERENCES

Gidhagen, L., 2004: Emissions, dynamics and dispersion of particles in polluted air. Doctoral dissertation, Department of Meteorology, Stockholm University, SE-106 91 Stockholm, Sweden.140 p.

Härkönen, J., 2002: Regulatory dispersion modelling of traffic-originated pollution. Finnish Meteorological Institute, Contributions No. **38**, FMI-CONT-38, ISSN 0782-6117, University Press, Helsinki, 103 p.

Karppinen, A, J. Kukkonen, T. Elolähde, M. Konttinen, T. Koskentalo and E. Rantakrans, 2000a: A modelling system for predicting urban air pollution, Model description and applications in the Helsinki metropolitan area. *Atmos. Environ.* **34-22**, 3723-3733.

Karppinen, A, J. Kukkonen, T. Elolähde, M. Konttinen and T. Koskentalo, 2000b: A modelling system for predicting urban air pollution, Comparison of model predictions with the data of an urban measurement network. *Atmos. Environ.* **34-22**, 3735-3743.

Kauhaniemi Mari, 2003: Usability of the Air Quality Model CAR-FMI in City Planning. Master's Thesis, University of Oulu, Department of Process and Environmental Engineering, Control Engineering Laboratory, 87+7 (13) p.

Keskinen, J., Pietarinen, K. and Lehtimäki, M., 1992: Electrical Low Pressure Impactor., *J. Aerosol Sci.*, **23**, 353-360.

Norbeck, J.M., Durbin, T.D., Truex, T.J. 1998: Measurement of Primary Particulate Matter Emissions from Light Duty Motor Vehicles. CRC Project No: E-24-2 Final Report.

University of California, College of Engineering, Center for Environmental Research and Technology. 56 p.

Pakkanen, Kerminen, Korhonen, Hillamo, Aarnio, Koskentalo & Maenhaut, 2001: Urban and rural ultrafine (PM0.1) particles in the Helsinki Area. *Atmos. Environ.* **35**, 4593-4607.

Pakkanen,T.A., Loukkola, K., Korhonen, C.H., Aurela, M., Mäkelä,T., Hillamo, R.E., Aarnio,P., Koskentalo, T., Kousa, A., Maenhaut, W., 2001: Sources and chemical composition of atmospheric fine and coarse particles in the Helsinki area. *Atmos. Environ.* **35**, 5381–5391.

Pirjola, L. and Kulmala, M., 2000: Aerosol dynamical model MULTIMONO. *Boreal Environment Research* **5**, 361-374.

Pirjola, L., Tsyro, S., Tarrason, L. and Kulmala, M., 2003: A monodisperse aerosol dynamics module – a promising candidate for use in the Eulerian long-range transport model. *Journal of Geophysical Research*, **108**, (D9), p. 4258. doi:10.1029/2002JD002867.

Pirjola, L., Parviainen, H., Hussein, T., Valli, A., Hämeri, K., Aalto, P., Virtanen, A., Keskinen, J., Pakkanen, T.A., Mäkelä, T., Hillamo, R.E. 2004: "Sniffer" – a novel tool for chasing vehicles and measuring traffic pollutants. *Atmospheric Environment*, **38,** 22, 3625-3635.

Pirjola L., Paasonen, P., Parviainen H., Hämeri K. and Hussein T., 2005: Mobile laboratory measurements in the urban environment in Helsinki. Abstracts of the 5th International Conference on Urban Air Quality, Valencia, Spain, 29-31 March, 2005. p. 154.

Pohjola, M A, Pirjola, L, Kukkonen, J, Kulmala, M. 2003: Modelling of the influence of aerosol processes for the dispersion of vehicular exhaust plumes in street environment. *Atmos. Environ.*, **37**, 3. 339-351.

Pohjola, M., Pirjola, L., Kukkonen, J., Karppinen, A., Härkönen, J., 2004: The influence of aerosol processes in vehicular exhaust plumes: model evaluation against the data from a roadside measurement campaign. In: Proceeding of the 9th International Conference on Harmonisation within Atmospheric Dispersion Modelling for Regulatory Purposes, 1–4 June 2004, Garmisch-Partenkirchen, Germany. Vol. **2**,142–146.

Shi, J.P., Mark, D., Harrison, R.M. 2000: Characterization of Particles from a Current Technology Heavy-Duty Diesel Engine. Environ. Sci. Technol., **34**, 748-755.

Yli-Tuomi, T., Aarnio, P., Pirjola, L., Mäkelä, T., Hillamo, R. and Jantunen, M., 2004: Emission from on-road vehicles in Finland. *Submitted to Atmos. Environ.*

Zhang, K.M., Wexler, A.S., 2004: Evolution of particle number distribution near roadways – Part I: analysis of aerosol dynamics and its implications for engine emission measurement. *Atmos. Environ.*, **38**, 6643-6653.

Viidanoja, J., Sillanpää, M., Laakia, J., Kerminen, V.-M., Hillamo, R., Aarnio, P., Koskentalo, T., 2002: Organic and black carbon in PM2.5 and PM10:1 year of data from an urban site in Helsinki, Finland. *Atmos. Environ.* **36**, 19. p. 3183-3193.