# A MODEL FOR EVALUATING THE CONCENTRATIONS OF $PM_{2.5}$ IN URBAN AREAS

*Jaakko Kukkonen[1], Ari Karppinen[1], Jari Härkönen[1], Päivi Aarnio[2], Tarja Koskentalo[2],*
Petri Tiitta[3] and Taisto Raunemaa[3]
[1]Finnish Meteorological Institute (FMI), Air Quality Research, Helsinki, Finland
[2]Helsinki Metropolitan Area Council, Helsinki, Finland
[3]University of Kuopio, Finland

## INTRODUCTION

Good-quality experimental field data specifically produced for model validation and evaluation purposes are scarce, with only a few datasets being available for local-scale models. No appropriate field measurement campaign datasets have been presented to date for the evaluation of roadside emission and dispersion models in the case of $PM_{10}$ or $PM_{2.5}$. Such information is required in evaluating the exposure of the population to particulate matter in urban and roadside environments.

We have developed a simple model for predicting the concentrations of $PM_{2.5}$ in urban areas, and evaluated the model predictions against available datasets. For a more detailed description of this study, the reader is referred to Tiitta et al. (2002) and Karppinen et al. (2002).

## THE MODEL

### Roadside dispersion modelling

The vehicular emissions and dispersion from the local road network was evaluated with the Gaussian finite-line source model CAR-FMI (Contaminants in the Air from a Road – Finnish Meteorological Institute). An overview of the model, together with evaluation against experimental field-scale data, has recently been presented by Kukkonen et al. (2001a).

The CAR-FMI model allows for the emissions and dry deposition of particulate matter. The vehicular $PM_{2.5}$ emissions were modeled to be dependent on vehicle travel velocity (ranging from 0 to 120 km $h^{-1}$), separately for the main vehicle categories. Light-duty vehicles were classified into three categories: (i) gasoline-powered cars and vans without a catalytic converter; (ii) gasoline-powered cars and vans equipped with a catalytic converter; and (iii) and diesel-powered cars and vans. Similarly, heavy-duty vehicles were classified into four categories: (i) diesel-powered trucks with a trailer; (ii) diesel-powered trucks without a trailer; (iii) diesel-powered buses; and (iv) buses powered by natural gas.

We fitted numerical correlations of PM emissions in terms of vehicle travel velocity, separately for each of the above-mentioned seven vehicle categories. These correlations are based on nationally-conducted vehicle emission measurements. The dry deposition of PM was evaluated using an analytical solution for the diffusion equation from a finite line source, as presented by Lin and Hildemann (1997).

### Regionally and long-range transported particulate matter

The only measurements of $PM_{2.5}$ in Finland available in 1999 were conducted in the Helsinki Metropolitan Area (Kukkonen et al., 2001b). The ratio of $PM_{2.5}$ and $PM_{10}$ concentrations varies substantially depending on the season of the year and the synoptic scale circulation patterns that determine the origin of the air masses (Pohjola et al., 2002).

It was therefore necessary to evaluate indirectly the regionally and long-range transported (abbreviated in the following as LRT) PM contribution. We developed a simple semi-empirical, statistical model that utilises as input values the daily measurements at the nearest EMEP (Co-operative programme for monitoring and evaluation of the long-range transmission of air pollutants in Europe) stations. The following quantities 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) (Leinonen et al., 2001). The sulphate, nitrate and ammonium ions are in particulate form, while nitrogen acid and ammonia are gaseous compounds in atmospheric conditions. These variables can be treated as proxy variables for the LRT PM; although LRT aerosol also contains other compounds, such as elemental and organic carbon.

Instead of using three separate proxy variables, it is convenient to define a single variable PM formed on the basis of the above-mentioned concentrations. These concentrations are reported as the mass of sulphur (i) or nitrogen ((ii) and (iii)). These were first converted to the mass of sulphate (i), nitrate (ii) and ammonium (iii), by multiplying using conversion factors in the temperature of 293.1 K that are 3.0, 4.4 and 1.3, respectively, and these masses were then added together. This conversion is necessary in order to treat concentration variables in comparable form. The results variable will be termed "the sum of ions" in the following.

We utilised a combination of data from several EMEP stations, in order to smooth out any disturbances that may have been caused by local emission sources. We utilised an interpolated value of the ion sum ($C_{ion}$), defined as

$$C_{ion} = \sum_{i=1}^{n} \chi_i \, C_{ion,i} \tag{1}$$

where the subscript $i$ refers to the EMEP stations considered, $n$ is the total number of stations, $\chi_i$ is the weight coefficient, and $C_{ion,i}$ is the ion sum at a specific site. The weight coefficient was defined separately for each daily value, and it is a normalised inverse value of a distance variable, that between the measurement location and the EMEP station.

**Combining the contributions from various sources**

Let us consider the contributions to measured concentrations of $PM_{2.5}$ at a roadside measurement location originating from various source categories. The total measured concentration of $PM_{2.5}$ can be written as (following Kukkonen et al., 2001b):

$$PM_{2.5} = PM_{2.5}^{tr,e} + PM_{2.5}^{tr,n\text{-}e} + PM_{2.5}^{st} + PM_{2.5}^{bg,urb} + PM_{2.5}^{bg,lrt} + PM_{2.5}^{res,o} \tag{2}$$

where the superscripts '*tr,e*' and *'tr,n-e*' refer to the primary (exhaust) and non-exhaust contributions of vehicular traffic from the nearest roads and streets, respectively. The superscript '*st*' refers to stationary sources, and the superscripts '*bg,urb*' and '*bg,lrt*' refer to the urban and LRT background. The superscript '*res,o*' refers to the resuspension of PM from various surfaces caused by other sources and mechanisms except for the traffic flow.

The term $PM_{2.5}^{tr,n\text{-}e}$ contains the contribution of non-exhaust PM emissions that originate from the vehicles, such as material from brakes and catalytic converters, and the resuspended particulate matter from street surfaces caused by the local traffic flow. This term was estimated

here simply to be directly proportional to the concentrations that originated from primary local vehicular emissions. It was not possible to evaluate separately the term $PM_{2.5}^{res,o}$, so we considered this term together with the terms $PM_{2.5}^{st}$ and $PM_{2.5}^{bg,urb}$,

$$PM_{2.5} = (1 + a)\, PM_{2.5}^{tr,e} + PM_{2.5}^{bg,,lrt} + (PM_{2.5}^{st} + PM_{2.5}^{bg,,urb} + PM_{2.5}^{res,o}) \quad (3)$$

where $a$ is a coefficient that has to be determined experimentally. The first term on the right-hand side of equation (3) represents the contribution from local traffic, the second term the LRT background, and the third term the contribution from all other sources. We have applied a modelling system that consists of an atmospheric boundary layer scaling model, a roadside dispersion model (for the evaluation of the term $PM_{2.5}^{tr,e}$), and a model for evaluating the regionally and long-range transported PM (the term $PM_{2.5}^{bg,lrt}$). The above-mentioned ion sum is assumed to be directly proportional to the LRT background concentration,

$$PM_{2.5}^{bg,lrt} = b\, C_{ion} \quad (4)$$

where $b$ is a coefficient that has to be determined experimentally. Substituting equation (4) into equation (3) results in a simple semi-empirical parameterization for evaluating the roadside concentration of $PM_{2.5}$,

$$PM_{2.5} = (1 + a)\, PM_{2.5}^{tr,e} + b\, C_{ion} + c \quad (5)$$

where the term $c$ denotes the contribution of all other sources, except for local traffic and the LRT background. After computing the terms $PM_{2.5}^{tr,e}$ and $C_{ion}$, the right-hand-side of equation (5) can be correlated statistically with the measured data of $PM_{2.5}$; this procedure can be used in evaluating the experimental coefficients $a$, $b$ and $c$. The validity of the resulting model, i.e., equation (5), can subsequently be evaluated by comparing its predictions against an independent dataset.

The third term on the right-hand-side of equation (5) contains contributions originated from various pollutant source categories. In cases where the influence of stationary sources is substantial, the term $PM_{2.5}^{st}$ should be computed separately.

**TESTING OF THE MODEL**

A particle measurement campaign was conducted in a suburban environment near a major road in Kuopio, Central Finland from August 3 to September 9, 1999. The mass concentrations of fine particles ($PM_{2.5}$) were measured simultaneously at distances of 12, 25, 52 and 87 m from the centre of a major road at a height of 1.8 m, using identical samplers. The concentration measurements were conducted during 16 daytime hours (from 6.00 a.m. to 10.00 p.m.) for 27 days. Traffic flows and relevant meteorological parameters were measured on-site; meteorological measurements from a nearby synoptic weather station were also utilised.

**RESULTS AND DISCUSSION**

We evaluated the term $PM_{2.5}^{tr,e}$ in equation (5) using the roadside dispersion model CAR-FMI. We utilised the measured traffic flow values, and the evaluated vehicle classifications in computing the PM emissions. The above-mentioned semi-empirical model based on EMEP measurements was applied to evaluate the term $PM_{2.5}^{bg,lrt}$ in equation (4). We utilised a combination of measurements from the EMEP stations located at Ähtäri and Virolahti. The right-hand-side of equation (5) was subsequently correlated statistically with the measured $PM_{2.5}$ concentration data. We applied a multiple regression analysis of the 16-hourly averaged

concentration values. All measured data was pooled in the analysis in order to obtain as good statistical reliability as possible. The average contribution of the stationary sources was found to be approximately only 1 % of the total measured concentration.

The values of these concentrations averaged over the whole measurement campaign, and the corresponding predicted concentrations, are presented in Figure 6 classified into various wind direction sectors. Clearly, the concentrations are highest in the immediate vicinity of the road (at distances of 12 and 25 m). Considering the whole dataset (including both upwind and downwind cases), traffic emissions in the road caused an increase in concentrations of approximately 30 % from the nearest to the largest distances. The wind directions are classified into 90° sectors: NE (from 0° to 90°), SE, SW and NW. The sectors NE and SW include the cases in which the wind direction deviates at most 45° from the direction of the road, and SE and NW are the downwind and upwind 90° sectors. As expected, the measured concentrations are clearly higher for the downwind sector than for the upwind sector. However, these values correspond to different meteorological conditions, and it is therefore not feasible to compare these quantitatively.

The differences of model predictions and data are largest for the downwind cases. This is caused by the relatively large inaccuracy in evaluating both the combustion and non-combustion emissions originated from local traffic.

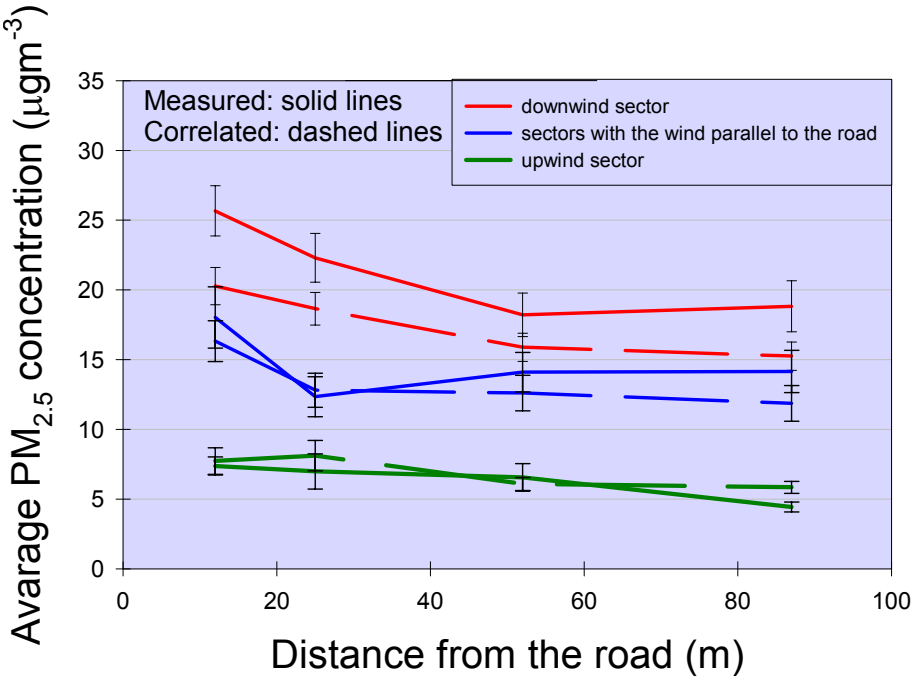


*Figure 1. Measured and correlated average $PM_{2.5}$ concentrations against distance from the centre of the road during the measurement campaign, classified into various wind direction sectors (Tiitta et al., 2002). The standard errors of the mean concentrations are also presented.*

## CONCLUSIONS

A novel mathematical model was suggested for evaluating the various contributions to measured concentrations originating from local traffic, long-range transport and other sources. However, the model has several inherent limitations. The evaluation of the contribution from combustion and non-combustion emissions originated from local traffic probably causes the largest uncertainties to the model predictions. The term representing the contribution of non-exhaust PM emissions was estimated here simply to be directly proportional to the concentrations that originate from primary local vehicular emissions; however, the resuspension caused by the traffic flow actually depends also on numerous other factors.

We conducted a measurement campaign in the vicinity of a major road in order to investigate the source contributions, and the spatial and temporal variation in fine PM. The model presented includes three numerical parameters that need to be empirically determined. We therefore statistically correlated the mathematical terms predicted by the model with the measured data, and evaluated the numerical values of these parameters. Clearly, this study cannot be considered to be a validation of the model presented; the model could be validated against another independent dataset by using the model parameter values determined in this study.

The model predictions could also be critically tested by comparing these with the corresponding results obtained with source apportionment techniques. The model presented could also be evaluated and probably applied in other European cities for predicting the $PM_{2.5}$ concentrations and analysing the source contributions to measured concentrations.

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