5.15 A MODELLING SYSTEM FOR PREDICTING URBAN $PM_{2.5}$ CONCENTRATIONS: NUMERICAL RESULTS AND MODEL EVALUATION AGAINST THE DATA IN HELSINKI

Ari Karppinen[1], Jaakko Kukkonen[1], Jari Härkönen[1], Mari Kauhaniemi[1] Anu Kousa[2] and Tarja Koskentalo[2]
[1] Finnish Meteorological Institute (FMI), Helsinki, Finland
[2] Helsinki Metropolitan Area Council (YTV), Helsinki, Finland

INTRODUCTION

Urban particulate matter originates from local mobile and stationary sources, from regional and long-range transport and from the suspended dust. Previous studies have shown that vehicular traffic is the most important local pollution source of both coarse particles ($PM_{10-2.5}$) and fine particles ($PM_{2.5}$) in the Helsinki Metropolitan Area. Based on data measured at the urban air quality monitoring network, the $PM_{2.5}$ concentrations have shown only moderate variation both spatially and temporally (e.g., Pohjola et al, 2002). It can therefore be anticipated that regionally and long-range transported aerosols (abbreviated here as LRT) are a major contributor to the urban air $PM_{2.5}$ concentrations. However, no model computations have previously been performed regarding the concentrations of fine particulate matter in that area.

MATERIALS AND METHODS

Air quality measurements

The EMEP-stations located nearest to Helsinki are: Utö (59°47' N, 21°23' E), Ähtäri (62°35' N, 24°12 'E), and Virolahti (60°32' N, 27°41' E). The ion concentrations that are measured daily at these EMEP stations are: (i) sulphate, (ii) the sum of nitrate and nitric acid, and (iii) the sum of ammonium and ammonia. The sulphate, nitrate and ammonium ions are in particulate form, while nitrogen acid and ammonia are gaseous compounds in atmospheric conditions.

The above mentioned measurements (i), and (ii) together with (iii) are reported as equivalent masses of sulphur and nitrogen, respectively. We define the so-called ion sum at station i as follows (Karppinen et al., 2004):

$$C_{ion,i} = 3.0 * SO_4^{2-} + 4.4 * (NO_3^- + HNO_3) + 1.3 * (NH_4^+ + NH_3) \qquad (1)$$

As the masses of ions have been given as the equivalent masses of sulphur or nitrogen, these values were converted to equivalent masses of the ions SO_4^{2-}, NO_3^- and NH_4^+, using the conversion factors in (1), respectively. The ion sum was shown to be a suitable proxy variable for LRT by Karppinen et al. (2004), although LRT contains also other compounds, such as elemental and organic carbon.

For comparing the model results with measurements, we utilized the hourly time-series of $PM_{2.5}$ concentrations measured at the urban traffic station of Vallila and the urban background station of Kallio in 2002. Both of these monitoring stations are located in central Helsinki and are part of the municipal air quality monitoring network of the Helsinki Metropolitan Area Council (YTV). The Vallila station is located in a small park, at a distance of 14 m from the nearest street with average traffic volume of 13 000 vehicles/day. The Kallio station is located in a small field, at a distance of 60 m from the nearest road with average traffic

volume of 8 000 vehicles/day. The $PM_{2.5}$ concentrations were measured with the ß-attenuation method (Eberline FH 62 I-R, Thermo Eberline, Santa Fe, NM, USA).

Evaluation of vehicular emissions

The vehicular $PM_{2.5}$ emissions were modelled 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. The contribution of non-exhaust emissions was estimated using results presented in Tiitta et al., 2002. The influence of cold-start and cold driving emissions was modelled by estimating the cold start and cold driving emission separately in 283 areas in the Helsinki Metropolitan Area.

The model for evaluating $PM_{2.5}$ concentrations

The emissions and atmospheric dispersion of vehicular emissions is evaluated using an updated version of a roadside emission and dispersion model, CAR-FMI (Härkönen, 2002). 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 (Tiitta et al., 2002). The total contribution originated from traffic can be written as a sum of in three terms

$$PM^{tr}{}_{2.5} = PM_{2.5}{}^{tr,e} + PM_{2.5}{}^{tr,n\text{-}e} + PM_{2.5}{}^{cs} \qquad (2)$$

The first term $PM_{2.5}{}^{tr,e}$ is the contribution from exhaust (assumed here to be the same as combustion-originated) emissions, while 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 suspended 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 local exhaust vehicular emissions. Clearly, the suspension caused by the traffic flow actually depends on a variety of factors, such as the properties of the road surface and the car fleet, meteorological conditions, and even on the cleaning of sand from road and street surfaces. The last term $PM_{2.5}{}^{cs}$ contains the contribution from cold start and cold driving emissions and is taken into account utilizing the empirically estimated emission fractions cs_t = (cold start emissions /line source emissions) during different seasons and days of week.

The LRT contribution is evaluated on the basis of a statistical model utilising as input values the daily sulphate, nitrate and ammonium ion concentrations at the EMEP stations. We utilized an interpolated value of the ion sum (1), defined as a sum of $C_{ion,i}$, normalized using the inverse value of the distance between the urban measurement location and each of the EMEP stations. In the previous study (*Karppinen et al.*, 2004) the measured urban air $PM_{2.5}$ concentrations at Helsinki area in 1998-2001 were associated with the ion sum values as follows:

$$PM_{2.5} = b\, C_{ion} + C_{loc} \qquad (3)$$

where b and C_{loc} are regression coefficients determined experimentally. The statistical estimation of b indicates the strength of temporal association of the background C_{ion}

concentration with the urban air $PM_{2.5}$ concentration, while C_{loc} indicates the statistically uncorrelated proportion of the $PM_{2.5}$ concentration.

Consequently, the terms bC_{ion} and C_{loc} were interpreted to represent contributions from LRT and all local sources, respectively. The value of the coefficient b was taken to be a weighted average of the coefficients b determined for the stations of Kallio and Vallila during years 1998-2001, leading to a value of b = 1.64 that was used in this study. The influence of the stationary emission sources on the fine particulate matter concentrations in urban air have previously been shown to be negligible in the area.

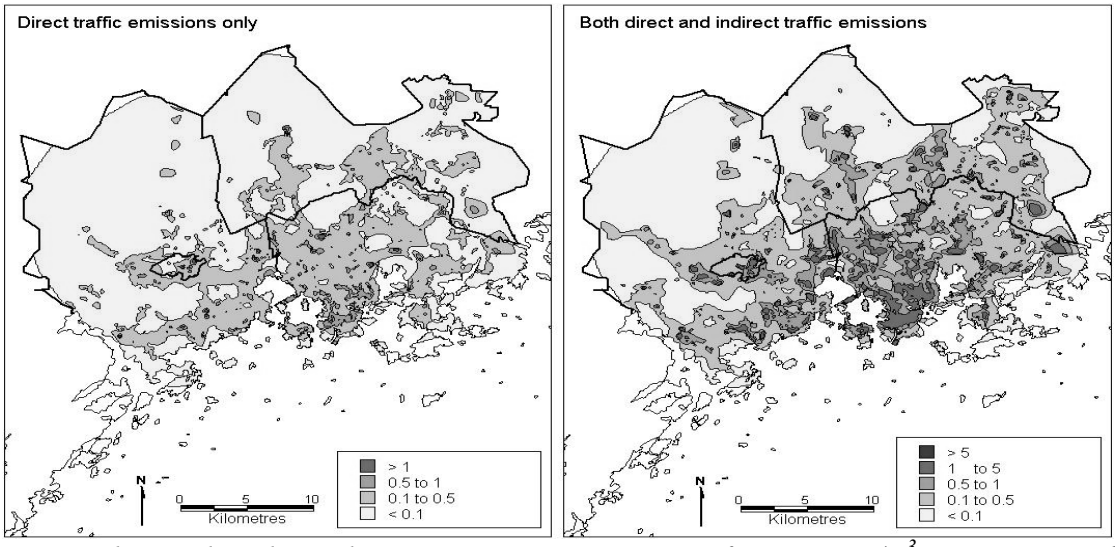
Combining (2) and (3) yields an equation for evaluating the total concentration of $PM_{2.5}$,

$$PM_{2.5} = (1 + a + cs_t)\, PM_{2.5}^{tr,e} + b\, C_{ion} \qquad (4)$$

where a = 1.8, according to the measurements by *Tiitta et al.* (2002), and the coefficients cs_t (varying from 0.17 to 0.48) are estimated separately for the winter and summer seasons, and for working days and weekends.

RESULTS AND DISCUSSION

The predicted spatial concentration distributions in the Helsinki metropolitan area are presented in Figures 1a-b and 2.



Figures 1a-b. Predicted yearly average concentrations of $PM_{2.5}$ [μg/m^3] originating from local traffic in the Helsinki metropolitan area in 2002. The left-hand-side panel presents the concentrations originating solely from exhaust emissions while the right-hand-side panel presents the concentrations originating from all local traffic emissions.

The highest concentrations are located in the centre of Helsinki (the peninsula in the middle of the figures) and in other major urban centres. Comparing Figures 1a and b shows (according to the computations) the substantial proportion of the non-exhaust contribution to local vehicular emissions. Clearly, when the LRT contribution is allowed for, in addition to the contributions originated from local sources, the spatial variation is more moderate (Figure 2).

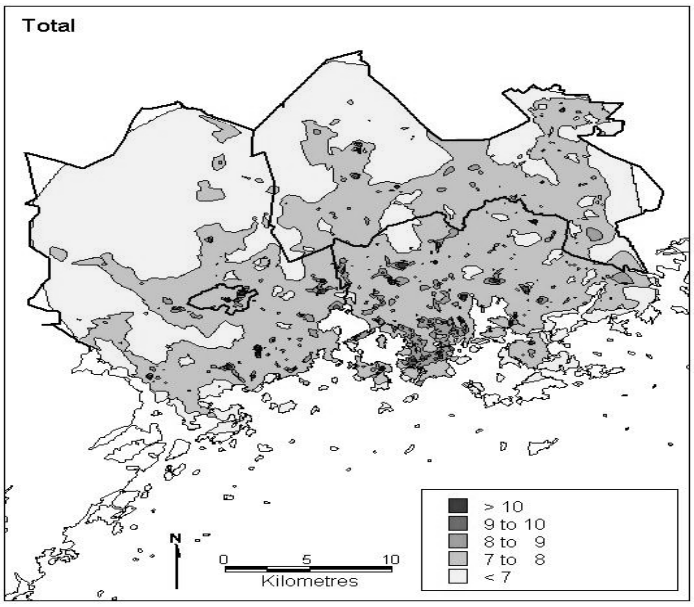


Figure 2. Predicted total yearly average concentrations of $PM_{2.5}$ [µg/m^3] in the Helsinki metropolitan area in 2002. Both the contributions from local vehicular sources and the regionally and long-range transported background have been taken into account.

On an annual basis, the estimated contribution from regionally and long-range transported origin to the observed $PM_{2.5}$ varies from less than 50 % in the centre of Helsinki to more than 90 % in the outskirts of the metropolitan area. The influence of the cold-start and cold driving emissions on the total $PM_{2.5}$ concentrations was found to be substantial. In winter ($T < 0$), cold starts and cold driving increased the amount of the exhaust emissions originated from local traffic approximately by 40 %.

Comparison against measurements

The model performance was evaluated against the $PM_{2.5}$ data of the monitoring network in the Helsinki Metropolitan Area.

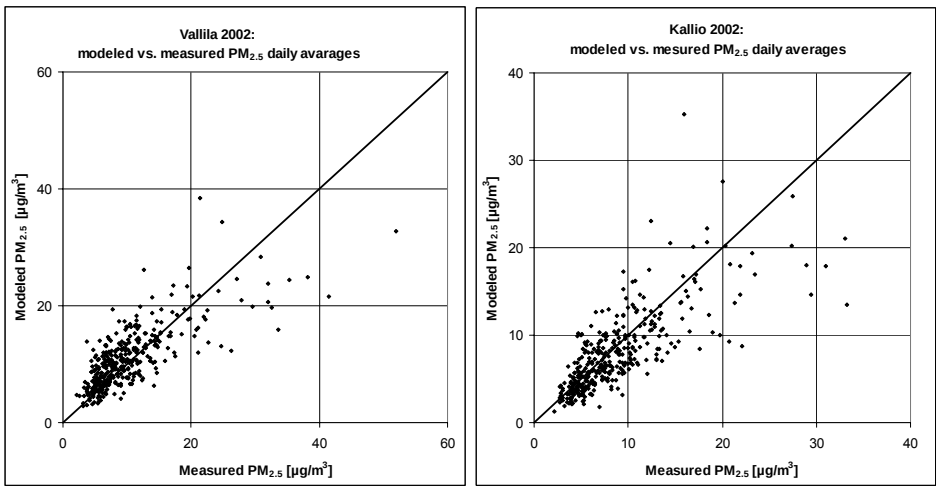


Figure 3. Scatter plots of observed and modelled daily average concentrations of $PM_{2.5}$ at the stations of Vallila and Kallio in 2002. In the model computations, both the contributions from all local sources and the regionally and long-range transported background have been taken into account.

The modelled daily average concentrations of $PM_{2.5}$ have been presented against the measured concentrations in Figure 3 at two monitoring stations. The resulting squared correlation coefficient is $R^2 = 0.60$ (N = 364), and the bias between observed and modelled values was less than 5 % for both stations. The empirical parameters in equation (4) were determined previously against datasets that have not been used in this model evaluation, viz. those in *Karppinen et al.* (2004) and *Tiitta et al.* (2002).

CONCLUSIONS

A model for evaluating the various contributions to measured concentrations of $PM_{2.5}$ originating from local traffic, long-range transport and other sources was presented and evaluated against urban measurements. The preliminary comparison of the daily averaged values with the corresponding measurements showed satisfactory results.

The model presented has inherent limitations. The regional background concentration of $PM_{2.5}$ consists mainly of ammonium nitrate, ammonium sulphate, and carbon compounds. Therefore, the accuracy of the semi-empirical sub-model for evaluating LRT contribution depends on the chemical composition of $PM_{2.5}$, especially the content of carboneous species that are not currently measured at the EMEP stations. Clearly, the evaluation of the contribution from combustion and non-combustion emissions originated from local traffic also causes 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 exhaust local vehicular emissions; however, the suspension caused by the traffic flow actually depends also on numerous other factors.

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