REGULATORY MODELLING OF PM - A HYBRID APPROACH FOR MODELLING THE PARTICLE TOTAL NUMBER CONCENTRATIONS

Härkönen J (a,), Karppinen A (a), Pakkanen T (a), Hussein T (b), Hämeri K (b, c)*

a) Finnish Meteorological Institute, Air Quality Research, Helsinki, FIN-00880, Finland
b) Department of Physical Sciences, Division of Atmospheric Sciences, P.O. Box 64, FIN-00014, University of Helsinki, Finland
c) Finnish Institute of Occupational Health, Topeliuksenkatu 41 A, Helsinki FIN-00250, Finland
*) Jari.Harkonen@fmi.fi , +358-9-1929 5452, Air Quality Research, FIN-00880, Finland

INTRODUCTION

We have studied the dependence of measured ultrafine (UFP) and accumulation (ACP) particle number concentrations on meteorological conditions and predicted fine particle mass concentrations (PM$_{2.5}$) in Viikki, Helsinki during May 15 to June 30 in 2000.The objective of this work is to relate the regulatory predictions of PM$_{2.5}$ mass concentrations on long range transportation (LRT), city background concentration and local line sources with the measured UFP and ACP near a motorway using the General Linear Model (GLM). We derive a statistical parameter "predictor effect" for estimating the influences of different predictors on UFP and ACP, which allows us to illustrate relatios of the predictors and particle number concentrations within and between the size classes (Härkönen et al., 2005).

MATERIALS AND METHODS

The measurement site and measurements

The measurement site at Viikki is located about 5 km north from downtown of Helsinki. It is a typical suburban background area influenced only weakly by other local sources except the traffic. The distance to one of the major highways in the Helsinki area was less than 100 m. This highway (Lahdenväylä) provides by far the most significant, temporally variable local source of aerosol particles. Another main road, located about 700 m to the south, approached the main highway and crossed it at a distance approximately 1 km to the southwest. A circular highway crossed the other main highways about 2.8 km northeast of the measurement site.

The particle number concentrations were measured about 1 m above the ground surface from May 15 to June 30, 2000. The aerosol measurements included measurements of number size distribution. The fine particle number size distributions were measured with a differential mobility particle sizer (DMPS) assembled at the University of Helsinki according to the principles described by Aalto et al. (2001). The sampling flow rate was 1 liter/min. The particle diameter range covered with the DMPS setup was from 7 to 600 nm. The instrument was calibrated prior to the measurement campaign.

Modelling methods

The regression modeling was performed utilizing GLM (SYSTAT 11, SYSTAT Software, Inc., 2004). The dependent variables are the measured total number concentrations and the predictors are a combination of measured (meteorological parameters) and deterministically or statistically modeled variables (like the contribution of LRT and traffic to PM2.5 mass concentrations) at the monitoring site of Viikki.

UFP and ACP are the dependent variables in the multiple linear regression model. UFP is defined as a sum of number concentrations in the nucleation and Aitken modes. Although the log-transformed UFP was observed not to be normally distributed, the distributions of ACP and the ratio UFP/ACP of the Viikki data fulfilled the normality criteria of multiple linear regression. The interaction terms were estimated to be insignificant.

We write regression equations (1a,b) for the hourly time-series, where the predictor variable X_i is a meteorological parameter or variable associated with modeled values: statistically determined regional, LRT or urban traffic mass concentrations and a deterministic estimate for the concentration originating from the nearest line sources. The number of predictors is denoted by n1 and n2. The terms in equation (1) are vector elements but the case-index is left out for simplicity.

$$Log\left(ACP\right) = \sum_{i=1}^{n1} k_i^{(1)}\ X_i + const1 \tag{1a}$$

$$Log\left(UFP / ACP\right) = \sum_{j=1}^{n2} k_j^{(2)}\ X_j + const2 \tag{1b}$$

The ion sum C_{ion} (Karppinen et al., 2004) is defined by equation (2), where the daily means of ion and gas concentrations are monitored at three EMEP stations and the ion sum that is interpolated (inverse distance) to the measurement site. The subscripts S and N denote that the mass has been given as the equivalent mass of sulphur or nitrogen. Hourly values are fitted from the time-series of daily means by the cubic spline method and finally the inverse distance weighting (χ_i) is applied to hourly ion concentration (3) of the station i.

$$C_{ion,i} = 3.0\left(SO_4^{2-}\right)_S + 4.4\left(NO_3^- + HNO_3\right)_N + 1.3\left(NH_4^+ + NH_3\right)_N \tag{2}$$

$$C_{ion} = \sum_{i=1}^{3} \chi_i\ C_{ion,i} \tag{3}$$

The predictor for local traffic emissions from the nearby roads and the motorway is estimated by a finite line source dispersion model CAR-FMI (Härkönen, 2002) by computing $PM_{2.5}$ concentration originating from line sources within a distance of one kilometer from the monitoring site. The urban background number concentrations are assumed to be related to the mean hourly traffic profile (TRP) in Helsinki describing the normalized hourly traffic volumes (number between [0, 1]).

The total mass concentration originating from traffic (TR) at the monitoring site is modeled as a superposition of local and urban effects of traffic emissions according to equation (4). Assuming that the diurnal variation of the local and urban traffic is similar the average ratio (r) of computed $PM_{2.5}$ to TRP is used as a scaling parameter to transform hourly urban TRP into the same scale and dimension as the local $PM_{2.5}$ mass concentration computed by CAR-FMI. The first term in equation (4) represents the proxy of urban traffic and depends on human activities i.e. diurnal time. Hence it may interact with meteorological variables as wind speed and stability. The second term represents the deterministic estimates from the nearest line sources.

$$TR = r * TRP + PM_{2.5}\text{, where } r = (PM_{2.5}/TRP)_{ave} \tag{4}$$

For estimating the influences of the separate predictors on the dependents, we rewrite equations (1a,b) into a product of ratios presented in equations (5a,b) separately for ACP and

UFP. We call the ratios on the right hand side the predictor effects (PE). The coefficient K_j in equation (5b) is the sum of the coefficients from equations (1) i.e. $K_j = k_i^{(1)} + k_j^{(2)}$ of the same predictor and CONST = const1 + const2. If n1 < n2, then coefficients and constants of missing ACP predictors equals zero in the model for UFP. The logarithmic form of PE used in this work follows directly from equations (5) and (1).

$$\prod_{i=1}^{n1} PE_i^{(ACP)} = \prod_{i=1}^{n1} \frac{Exp(k_i^{(1)}\ X_i)}{\left[{}^{ACP}\!/\!{}_{Exp(const1)} \right]^{1/n1}} = 1 \qquad (5a)$$

$$\prod_{j=1}^{n2} PE_j^{(UFP)} = \prod_{j=1}^{n2} \frac{Exp(K_j\ X_j)}{\left[{}^{UFP}\!/\!{}_{Exp(CONST)} \right]^{1/n2}} = 1 \qquad (5b)$$

RESULTS AND DISCUSSION

A summary of the results of the linear regression (eq.1) is presented in Table 1. The regression coefficients are mostly very significant ($p<0.001$) or significant ($0.01>p>0.001$) for the predictors ion sum C_{ion}(μgm^{-3}), traffic emissions TR(μgm^{-3}), wind speed U(ms^{-1}), relative humidity RH(%) and inverse of Monin-Obukhov length L(m). Gidhagen et al. (2005) and Hussein et al. (2005) found also a clear dependence of particle number concentration on ambient temperature. In our study temperature was not found to be a significant predictor, which is only to be expected, taking into account the relatively short duration of the measurement campaign.

The model of Log(ACP) includes four predictors, while the regression of Log(UFP/ACP) includes also stability (L^{-1}). Based on the values of the adjusted squared multiple correlation (R^2) the models explain 77 % and 67 % of the variance in Log(ACP) and Log(UFP/ACP) regimes including 291 cases, respectively.

Table 1. The regression coefficients (coeff) with lower and upper 95 % confidence limits (L95% and U95%) and p-values for significance of the predictors (see text). N is the number of cases and R^2 is the adjusted squared multiple correlation.

	Log(ACP)				Log(UFP/ACP)			
N	291				291			
R^2	0.765				0.668			
	coeff	L 95%	U 95%	p	coeff	L 95%	U 95%	P
Const	6.215	5.978	6.451	0.000	3.511	3.208	3.813	0.000
C_{ion}	0.175	0.149	0.200	0.000	-0.319	-0.345	-0.278	0.000
TR	0.104	0.084	0.125	0.000	0.039	0.012	0.065	0.004
U	-0.116	-0.140	-0.092	0.000	0.047	0.017	0.078	0.002
RH	0.009	0.007	0.011	0.000	-0.006	-0.009	-0.003	0.000
1/L					2.099	1.246	2.953	0.000

Predictor effects

We applied the regression coefficients of Table 1 to equations (5) to compute the logarithm of the predictor effects for ACP and UFP regimes in all cases (N = 291). The slopes illustrate the influence of predictor on the total number concentrations, while the intercepts represent the average influence of other predictors. Because the influence of the number of predictors (n1 = 4, n2 = 5) on the predictor effect of ACP is only 3.6 % ± 3.4 % lower than on UFP, the results can be reasonably compared within and between the different size classes.

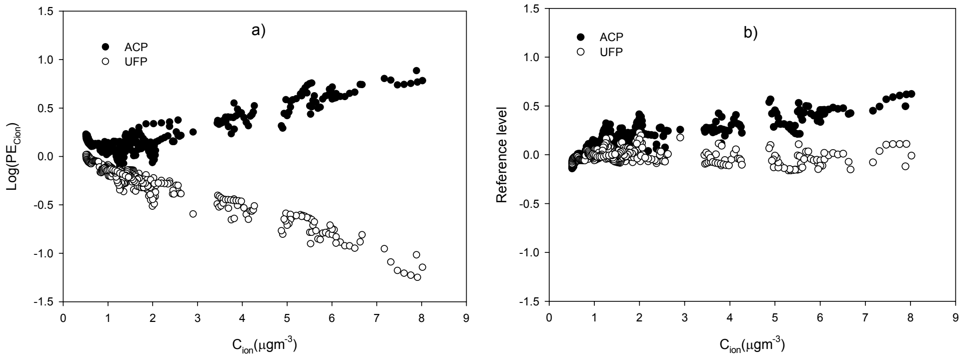

Figure 1. Logarithm of predictor effects PE (a) and corresponding reference levels (b), computed by equation (5) against predictor values in observed ACP and UFP regimes (N = 291), are illustrated.

When interpreting the plotted results we recall that log(PE) of a case is the difference between influences of an individual predictor (k_iX_i) and average of all predictors (reference levels). The influences of reference levels are illustrated in Figure 1 (right). Positive value of log(PE) indicates greater influence of primary predictor, while negative value represents the reverse and zero indicates equality. Applying these principles to Figure 1, we conclude that the predictor ionsum (C_{ion}) has a strong influence in ACP and weak in UFP regime. Physical processes like the influence of polluted background air and scavenging of fresh UFP by larger particles (e.g. Zhang and Wexler, 2004) may explain the result.

We also observed (not shown) that the predictor “traffic” (TR) has strong influence for both size classes, but affects more UFP. Wind speed (U) affects clearly more UFP than ACP, while relative humidity (RH) has more affect on ACP. Finally the influence of stability (L^{-1}) is positive on UFP with increasing stability, though neutral stratification is dominant during the study period.

CONCLUSIONS

We constructed a regression model (GLM) for predicting the particle number concentrations in two size classes (UFP, ACP) utilizing measured particle number concentrations, meteorological parameters and results from regulatory mass concentration modeling. With just 4-5 explaining variables we could satisfactorily predict the observed particle number concentrations.

Interpretation of the modeled GLM-parameters and predictor effects also indicate that UFP is mainly associated with nearby motorway emissions and in a less extent with the average local traffic in the city. The main part of ACP was based on LRT, but also the traffic component of the city background had some influence on it. The increasing LRT mass concentration contributes to UFP and ACP reversely.

High values of explained variance suggest that this type of model could be used successfully also in other similar urban locations, however, this hypothesis needs still to be tested with independent data.

REFERENCES

Härkönen., J., Hussein, T. Karppinen, A., 2005. Development of a hybrid model for predicting particle number concentrations. In Abstract 20B-1 of the 2005 AAAR PM Supersites Conference in Atlanta, Feb 7-11, GA.

Aalto, P., Hämeri, K., Becker, E., Weber, R., Salm, J., Mäkelä, J., Hoell, C., O'Dowd, C., Karlsson, H., Väkevä, M., Koponen, I.K., Buzorius, G., Kulmala, M., 2001. Physical characterization of aerosol particles during nucleation events. *Tellus* **53B**, 344-358.

Karppinen A., Härkönen, J., Kukkonen J., Aarnio P., Koskentalo T., 2004. Statistical model for assessing the portion of fine particulate matter transported regionally and long range to urban air. *Scand J Work Environ Health,* **30**, no. 2, p. 47-53.

Härkönen, J., 2002. Regulatory dispersion modeling of traffic-originated pollution. Finnish Meteorological Institute, Contributions No. 38, FMI-CONT-38. University Press, Helsinki, 103 p.

Gidhagen, L., Johansson, C., Langner, J., Foltescu, V.L., 2005. Urban scale modelling of particle number concentration in Stockholm. *Atmospheric Environment* **39**, 1711-1725.

Hussein, T. Hämeri, K., Aalto, P.P., Paatero, P., Kulmala, M., 2005. Modal structure and spatial-temporal variations of urban and suburban aerosols in Helsinki—Finland. *Atmospheric Environment* **39**, 1655-1668.

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