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# REGULATORY DISPERSION MODELLING OF TRAFFIC-ORIGINATED POLLUTION

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ACADEMIC DISSERTATION in physics.

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Abstract

This thesis describes the national finite line source model (CAR-FMI) for the dispersion of traffic-originated pollution from an open road network. CAR-FMI computes the concentrations of carbon monoxide (CO), nitrogen monoxide (NO), nitrogen dioxide ($NO_2$), total of nitrogen oxides ($NO_x$), ozone ($O_3$) and exhaust fine particulate matter ($PM_{2.5}$) in local scale using the Gaussian plume dispersion with dry deposition associated with $PM_{2.5}$. The computed results of statistically analysed hourly concentrations are available in tabular form or presented graphically utilizing the GIS (Geographic Information System) MapInfo.

The modelled $NO_x$ concentrations are compared with the field measurements as well as with the computed results of a Lagrangian model (GRAL). The comparisons show a good agreement between modelled and measured concentrations except in case of weak wind speed conditions when CAR-FMI substantially overestimates the concentrations. The overestimation was the result of the meandering effect, which is not taken into account in the first version of the model. A numerical method for the correction of the influence by meandering is suggested in this work.

A semi-empirical method for the estimation of yearly $PM_{10}$ concentration is presented. Also a statistical method for the estimation of the long-range transported $PM_{2.5}$ concentration is described. Additionally, an indirect statistical method for the estimation of the emission factor of non-exhaust fine particles by the traffic in summertime conditions is suggested in this thesis. All original papers are associated with the different projects of the Atmospheric dispersion modelling group in Air Quality Research (AQR) of the Finnish Meteorological Institute (FMI).

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Tiivistelmä


Tutkimus liittyy viivalähdemallin CAR-FMI kehittelyyn, jota sovelletaan liikenteestä peräisin olevien kaasumaisten yhdisteiden ja hiukkasten leviämisen mallintamiseen. CAR-FMI laskee tieverkoston aiheuttaman hiilimonoksidin (CO), typpimonoksidin (NO), typpidioksidin ($NO_2$), kokonaistypen oksidien ($NO_x$), otsonin ($O_3$) ja polttoaineen palamisesta syntyvien pienhiukkasten ($PM_{2.5}$) pitoisuudet ns. paikallisessa skaalassa. Leviämismalli perustuu gaussimaiseen dispersioyhtälöön sisältäen halkaisijaltaan < 2.5 μm hiukkasten kuivadeposition. Laskennalliset tuntipitoisuudet analysoidaan tilastollisesti ja tulokset esitetään joko taulukkomuodossa tai graafisesti käyttämällä GIS (Geographic Information System) MapInfo-systeemiä.

Mallinnettuja typenoksidipitoisuuksia verrattiin vastaaviin kenttäoloissa mitattuihin pitoisuuksiin. Lisäksi suoritettiin mallivertailu gaussimaisen CAR-FMI ja Lagrange-tyypisen GRAL-mallin välillä. Mitattujen ja mallinnettujen pitoisuuksien välinen yhteensopivuus oli varsin hyvä. Poikkeuksena on heikkotuuliset olosuhteet, jolloin tuulensuunta läheni tien suuntaa. Tällöin CAR-FMI ennustaa selvästi liian korkeita pitoisuuksia. Mallivertailun perusteella yliennustus liittyy tuulensuunnan vaihteluun (nk. meandering-efekti), jonka vaikutuksille työssä esitetään numeerinen korjausehdotus.

Työssä kuvataan myös semiempiirinen malli $PM_{10}$-pitoisuuksien vuosikeskiarvon laskemiseksi kaupunkialueella. Samoin esitetään tilastollinen menetelmä kaukokulkeutuneen $PM_{2.5}$-pitoisuuden arvioimiseksi käyttäen hyväksi EMEP-mittausasemien verkostoa. Lisäksi esitetään tilastollinen epäsuora menetelmä liikenteen (jarrujen, renkaiden, tienpinnan) $PM_{2.5}$-päästökertoimen määrittämiseksi. Tutkimus on suoritettu Ilmanlaadun tutkimuksen mallien kehittelyyn liittyvissä projekteissa Ilmatieteen laitoksella.



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**Contents**

## List of original publications

This thesis is based on six original publications, referred to in the text by bold numbers (**1** – **6**). The papers are reprinted with kind permission of the publishers.

1. Härkönen, J., Valkonen, E., Kukkonen, J., Rantakrans, E., Jalkanen, L. and Lahtinen, K., 1995. An operational dispersion model for predicting pollution from a road. International Journal of Environment and Pollution 5, pp. 602 - 610.

2. Härkönen J., Walden J. and Kukkonen, J., 1997. Comparison of model predictions and measurements near a major road in an urban area. International Journal of Environment and Pollution 8, pp. 761-768.

3. Härkönen, J., Kukkonen, J., Valkonen, E. and Karppinen, A., 1998. The influence of vehicle emission characteristics and meteorological conditions on urban $NO_2$ concentrations. International Journal of Vehicle Design 20, pp. 125-130.

4. Kukkonen, J., Härkönen, J., Walden, J., Karppinen, J. and Lusa, K., 2001. Evaluation of the CAR-FMI model against measurements near a major road. Atmospheric Environment 35, pp. 949-960.

5. Kukkonen, J., Härkönen, J., Karppinen, A., Pohjola, M., Pietarila, H. and Koskentalo, T., 2001. A semi-empirical model for urban $PM_{10}$ concentrations, and its evaluation against data from an urban measurement network. Atmospheric Environment 35, pp. 4433-4442.

6. Karppinen, A., Härkönen, J., Kukkonen, J., Aarnio, P. and Koskentalo, T., 2002. Statistical model for assessing the portion of fine particulate matter transported regionally and long range to urban air. Scandinavian Journal of Work, Environment & Health, in press.

# 1 Introduction

Air pollution is associated with anthropogenic emissions especially since Los Angeles photochemical smog in 1944 and London sulphurous smog in 1952. The term *smog* is derived from the words *smoke* and *fog* (Finlayson-Pitts and Pitts, 1986, p. 5). The assessment of air quality standards in EU countries includes the statistically defined limit values for compounds and less obligatory guideline values. Environmental authorities control the air quality standards using monitoring networks, which produce temporally and locally fixed information of the concentrations. The geometry of emission sources or the network of several sources as well as nearby obstacles affect strong gradients in the spatial concentrations of pollutants. Then the measured concentrations are locally restricted especially in case of short averaging periods when the generalization of the concentration level based on an individual monitor becomes easily misleading.

The dependencies between the atmospheric turbulence and dispersion in the atmospheric boundary layer, revealed during the 20$^{th}$ century, and rapidly increased computational capacity has enabled the air quality modelling. It describes the physics, chemistry and meteorology of pollutants using mathematical and statistical methods. In addition to the air quality control, modelling enables the prediction and testing of future scenarios in environmental planning. This is an implication of computational models combined with large time-series of real meteorological conditions: hourly time-series long enough (e.g. three years) include the meteorological cases of the near future with a great probability. Due to the partly different ranges of application, modelling and monitoring are complementary methods in air quality control and research.

In general, the most realistic models are nonlinear, dynamic and stochastic in nature (Kapur, 1988, p. 9). However, regulatory air quality models are usually static during a certain time interval e.g. one hour and deterministic i.e. non-stochastic. This study deals with regulatory air quality dispersion models of the local scale, which are further classified according to the type of the pollutant source. The evaluation of models and also their validation against good-quality databases, a basic principle of modelling (e.g. Kukkonen, 2000), is emphasized through this study.

Statistical models include a large collection of methods of which those based on simple and multiple linear regressions are commonly applied to the regulatory use. Statistical regulatory models in air quality studies are always locally limited and frequently called empirical or semi-empirical depending on the type of the analysed data or need to emphasize some features of the model. Receptor models, a branch of statistical models applying more sophisticated statistics and the chemical analysis to the monitored data, estimate quantitatively the contribution of different sources at the receptor point. Statistical models in this thesis are used in connection with particulate matter ($PM_x$), in which the lower index represents the upper limit of the particle aerodynamic diameter (µm) in the fraction, concentrations. Instead, chemical reactions on particle surface (e.g. Pohjola et al., 2002) as well as the number concentrations are outside the scope of this study.

## 2 Aims of the study

The aims of the thesis are

- to illustrate the special features involved in the dispersion of pollutants from traffic emissions
- to describe a finite line source model CAR-FMI emphasizing the properties of the refined version
- to present the results of the comparisons between the measured and the predicted values and to show the results of intercomparison of a Gaussian model (CAR-FMI) and a Lagrangian model GRAL
- to present the results and interpretation of the sensitivity analysis of CAR-FMI
- to present a semi-empirical model for computation of the mass concentration of thoracic particle ($PM_{10}$) concentrations
- to present a statistical method applicable for computation of the long-range transported (LRT) background concentration for fine particulate ($PM_{2.5}$) matter
- to present a statistical application of field measurements, CAR-FMI and the statistically determined $PM_{2.5}$ background concentration to estimate an emission factor for non-exhaust emissions from a main road in summertime conditions

## 3 Previous work

An overall description of the road meteorology is presented on the www-pages of Swedish National Road Administration (SNRA), while Moussiopoulos et al. (1996) presents a review of air pollution models. The inventory of regulatory models and their practical properties includes short description of models and general information on contact persons and references (Schazmann et al., 1997). The list of typical features of some regulatory line source models in use (**1**) is updated in Table 1 including more information on particulate matter. It should be noted that exhaust particles (aerodynamic diameter < 1 μm) behave practically like gases, but in case of greater particles and increasing distances from the source the role of dry deposition becomes more significant.

Due to their simplicity and direct applicability for estimates on a local scale, various versions of the Gaussian line source model have been used for dispersion evaluations from a road. Such models include HIWAY-2 (Petersen, 1980), CALINE-4 (Benson, 1984 and 1992), GM (Chock, 1978), GFLSM (Luhar and Patil, 1989) and OMG (Kono and Ito, 1990). The ROADWAY (Eskridge and Catalano, 1987) and MGO (Berlyand et al., 1990) models are based on a K-theory (Eulerian) approach. An obvious advantage of the K-theory models is that they can readily include the interaction of diffusion processes and chemical transformation.

In the HIWAY-2 and CALINE-4 (California line source model) models, the concentrations predicted by a Gaussian line source equation for an arbitrary wind direction are solved by a numerical procedure. This procedure divides the road into a series of elements, from which incremental concentrations are then computed and summed up. Both models allow for a finite mixing height in the computations.

Csanady (1972) presented an analytic solution of the Gaussian equation for a finite line source, for the special case of the wind perpendicular to the road. In the GFLSM (General Finite Line Source Model) model this solution has been extended to allow for any wind direction with respect to the road. The analytic solution in the GFLSM model was originally derived from Gaussian formulae similar to, for instance, the HIWAY-2 and CALINE-4 models, except that the mixing height was assumed to be infinite. The analytical solution is computationally much more economic than the above-mentioned numerical solutions.

The basic dispersion equations of the CAR-FMI model (**1**; Härkönen et al. 1996) are based on the Gaussian finite line source model by Luhar and Patil (1989), while the dispersion equation with dry deposition is based on the analytical solution by Lin and Hildemann (1997). The dispersion parameters in the CAR-FMI are modelled as function of the Obukhov length, the friction velocity and the mixing height.

Table 1. Some features of regulatory models for atmospheric dispersion from a road. The models can be broadly classified as Gaussian numerical and analytic, or K-theory models.

| Model | Dispersion parameters | Plume rise | Chemical transform. | Dispersion of particles | Dry deposit. of particles | References |
|---|---|---|---|---|---|---|
| HIWAY-2 Gaussian numerical | traffic induced and ambient turbulence | no | no | no | no | Petersen, 1980 Rao et al., 1980 |
| CALINE-4 Gaussian numerical | traffic induced and ambient turbulence | no | discrete parcel method | yes PM | no | Benson, 1984 Benson, 1992 |
| OMG Gaussian numerical | eddy diffusion coefficients, volume source | yes | no | no | no | Kono and Ito, 1990 |
| GM Gaussian analytic | traffic induced and ambient turbulence | yes | no | no | no | Chock, 1978 Luhar and Patil, 1989 Rao. et al., 1980 |
| GFLSM Gaussian analytic | traffic induced and ambient turbulence | yes | no | yes PM | no | Luhar and Patil, 1989 |
| CAR-FMI Gaussian partly analytic | traffic induced and ambient turbulence | no | discrete parcel method | yes PM | yes | papers **1** and **4**, Härkönen et al., 1996, 2001 |
| ROADWAY K-theory | eddy diffusion coefficients | no | interactive with diffusion | no | no | Eskridge and Catalano, 1987 Eskridge and Rao, 1986 |
| MGO K-theory | eddy diffusion coefficients | no | interactive with diffusion | no | no | Berlyand et al., 1990 |

Lagrangian dispersion models have become increasingly more feasible, due to the advances in computer technology (e.g., Janicke et al., 1994; Oettl et al. 2001a). Both Eulerian and Lagrangian models are less limited by topographical and meteorological conditions compared to Gaussian plume models, e.g. Oettl et al. (2001b).

# 4 Transportation of pollutants in the atmospheric boundary layer

The released pollutants mix within the atmospheric boundary layer (ABL), where the flows are under the influence of the ground surface. Because of the different mixing properties, the lowest and upper parts of ABL are divided into the surface layer (about 10 % of the ABL depth) and the convective mixed layer or the stable outer layer depending on the atmospheric stability (Kaimal and Finnigan, 1994, p. 21-25). Consequently, the height of the emission source and the horizontal scale are crucial factors in the pollutant dispersion with weak initial plume rise. In case of traffic emissions the plume rise is negligible and the source locates within the first few meters from the ground surface. Moreover, the pollutant concentrations are determined at distances less than ten kilometers from the source in local scale dispersion models indicating the importance of the surface layer.

The surface layer depth in subarctic latitudes changes typically between some tens of meters (> 10 m) and one hundred meters (< 150 m) in stable and unstable atmosphere, respectively. The practical lower and upper limits (in parenthesis) are associated with episodic (inversion) and convective conditions. Surface layer has three important properties:

- Vertical shearing stress is approximately constant.
- The flow is insensitive to the earth’s rotation.
- The wind structure is determined primarily by surface friction and the vertical gradient of temperature.

These properties are combined into one parameter called Obukov length (L) having a characteristic value that depends on the vertical mixing in the surface layer. It has been shown that various atmospheric parameters and statistics, when normalized by appropriate powers of scaling velocity (friction velocity) and scaling temperature, are universal functions of z/L, where z is height from the ground surface.

The similarity theory was initially presented by Monin-Obukhov (M-O) in 1954. It enables the computation of wind speed and stability, the most important factors affecting dispersion of pollutants in the surface layer, at the height of interest in modern regulatory dispersion models. However, according to recent studies M-O similarity fails in the surface layer in case of weak wind speed (e.g. Oettl et al. 2001a). In case of traffic emissions, the road may also cause a significant local perturbation to the vertical stability reaching some tens of meters from the road, because the physical properties of the road differ from the nearby terrain as suggested by Chen et al. (1999).

## 5 Description of the refined modelling system CAR-FMI

Road is treated as a straight line of finite length. The traffic volume of the road during one hour is assumed constant and thus the traffic emissions can be interpreted as a finite line source. CAR-FMI is a Gaussian finite line source dispersion model i.e. a plume model for an open road network (**1**). The model computes an hourly time-series of the pollutant dispersion for CO, NO, $NO_2$, $NO_x$ and exhaust $PM_{2.5}$ concentrations with input information from

- the number and locations of the line sources
- the hourly traffic volumes of the roads
- compounds to be computed and details of statistical interests of the output
- hourly time-series of the meteorology and the background concentration

The meteorological time-series is computed by the meteorological pre-processing model (MPP-FMI), developed at the FMI (Karppinen et al., 1997). The background concentrations of gaseous compounds are interpolated from the measurements of the monitoring network of FMI, while the background concentrations of fine particulate matter can be estimated (**6**), if local measurements are not available. The technical structure of the refined CAR-FMI model (Härkönen et al., 2001) is presented in Figure 1 including several improvements compared to the version 1.0 (**1**; Härkönen, et al., 1996).

The functions of the Graphical User Interface (GUI) CAR-FMI are to allow the user

- to enter the input information
- to save/retrieve input data
- to launch the dispersion model
- to prepare MIF file(s), which are used by MapInfo to display spatial distribution of various variables from the solution of the dispersion equation

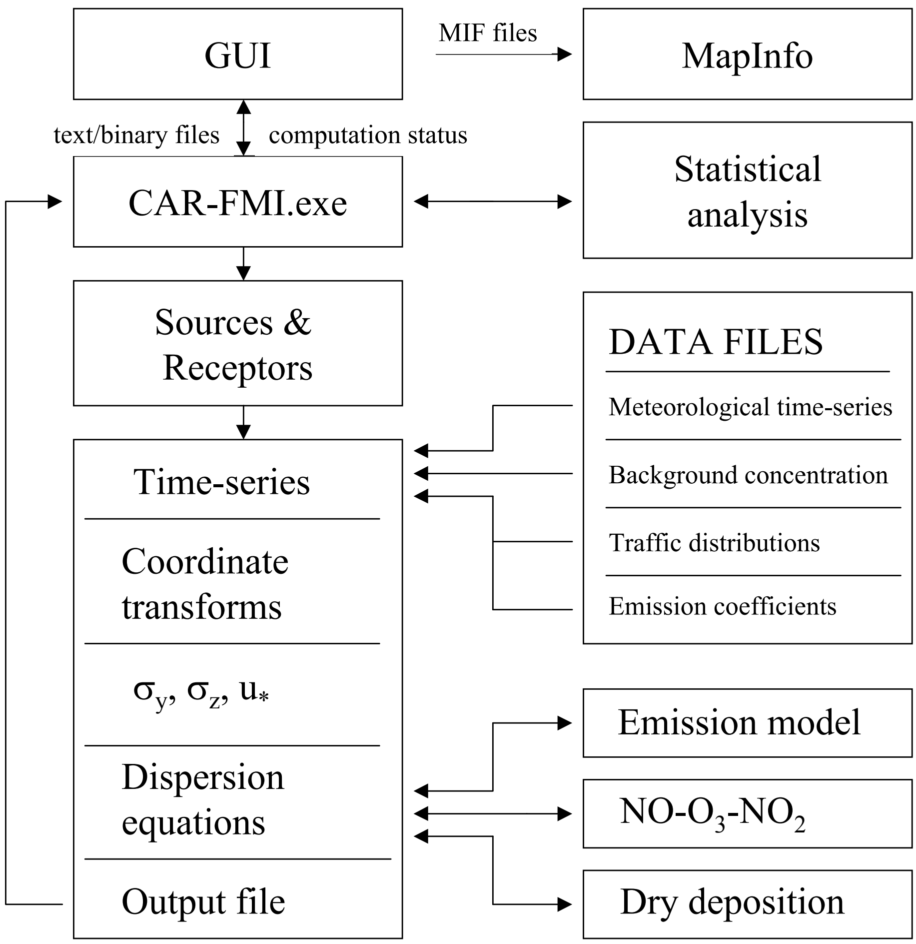


Figure 1. Technical structure of the CAR-FMI model, in which dry deposition is associated with $PM_{2.5}$ concentrations.

After feeding the input information the grid is created and the computation begins by the reading of the hourly meteorological and background concentration time-series and temporal distributions of the traffic volume intensities. The user may change the files in the block “Data files”. The emission coefficients associate with the fitting of the emission factor against the average vehicle travel velocity. The computation continues by coordinate transformations, which depend on wind direction and locations of roads and receptor points.

The computation of dispersion parameters $\sigma_y$ and $\sigma_z$ and the friction velocity $u_*$ is similar to the version 1.0. Also the dispersion equation for gaseous compounds and the chemical conversion of nitrogen oxides are unchanged. On the other hand, the emission model and dispersion equation for particulate matter with dry deposition are new features of the model. The final results of statistically analysed hourly concentrations are available in tabular form or presented graphically utilizing the GIS (Geographic Information System) MapInfo.

### 5.1 Dispersion parameters and the transportation velocity of the plume

The determination of the dispersion parameters in the surface layer is based on Taylor's frozen turbulence hypothesis (Kaimal and Finnigan, 1994, p. 61): the turbulence field is frozen in time and transported horizontally past the observer enabling the conversion of temporal measurements to spatial patterns. The total diffusion is divided into initial diffusion affected by the traffic and the diffusion caused by ambient meteorological conditions according to Figure 2. There are interactions (illustrated by arrows) between the ambient wind speed, exhaust velocities of the tailpipe emissions (Chan et al, 2001) and the traffic wake-induced turbulence (Rao, 2001). The simulated turbulence caused by exhaust velocity is clearly greater during acceleration than in deceleration cycle (Chan et al, 2001). It is meaningful only in stagnant conditions, because otherwise the atmospheric turbulence dominates the process.

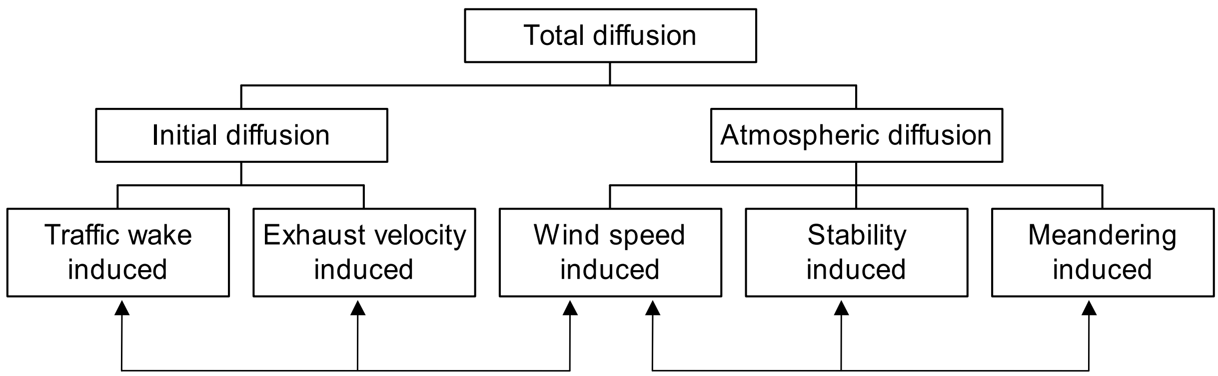


Figure 2. A block diagram of the primary diffusion processes in the plume originated from traffic emissions. The lines with arrows describe the interactions between the blocks.

Total turbulence is a superposition of mechanically and thermally generated eddies, while meandering (e.g. Seinfeld, p. 546) is associated with the size distribution of eddies. The driving force of the dispersion is the total turbulence associated with the mean wind speed. The lateral and vertical dispersion parameters are defined by

$$\begin{aligned} \sigma_y^2(x) &= \sigma_{ya}^2(x) + \sigma_{yo}^2 \\ \sigma_z^2(x) &= \sigma_{za}^2(x) + \sigma_{zo}^2 \end{aligned} \qquad (1)$$

The symbols $\sigma_y$ and $\sigma_z$ mean total lateral and vertical dispersion parameters and the subscripts "a" and "o" refer to atmospheric and traffic-originated turbulences, while x is the distance of the receptor point from the road in wind direction. The lateral dispersion parameter $\sigma_{ya}$ (standard deviation of the concentration perpendicular to the flow) and meandering (e.g. Seinfeld, p. 546) are both proportional to the horizontal wind speed fluctuations (Oettl et al., 2001a). In addition, because meandering is inversely proportional to mean wind speed, the lateral dispersion parameter increases with increasing meandering. If meandering is not accounted for, an underprediction of $\sigma_{ya}$ is expected.

The solution for the influence of traffic wake on the initial dispersion parameter requires 3D numerical model. The fitted solutions based on field experiments are generally used in Gaussian models. The flow pattern near the road is not homogenous (as assumed in Gaussian models), which has been shown in several field experiments e.g. Chock (1977 and 1980) and Rao and Sedefian (1979), and in wind tunnel simulations by Eskridge and Rao (1986) and Rao (2001). If the travel velocity is smooth, as in the GM-experiment (e.g. Petersen et al., 1980), the fitting is valid in a limited travel speed range. However, according to the equation (1) the relative effect of the erroneous initial dispersion parameters on the total dispersion decreases rapidly with the distance from the road.

The determination of initial dispersion parameters (eq. 1) of CAR-FMI is based on fitted results of GM-field experiment in which the traffic wake induced dispersion dominates the initial dispersion processes (Petersen et al., 1980). The main equations are presented in paper (**1**). The influence of meandering is not included in the lateral dispersion parameter.

The modeling of meteorological dispersion parameters (Gryning et al., 1987) is based on M-O similarity. The vertical wind speed profiles applied in CAR-FMI are in accord with the paper of van Ulden and Holtslag (1985), in which the mean wind speed as a function of height is presented using

Obukhov length L, friction velocity $u_*$ and the influence function of momentum flux $\psi_m$. The plume is transported with the wind speed at the emission height.

### 5.2 Emission modelling

The structure of emission model is independent of emitted compounds. The refined emission model (Härkönen et al., 2001) includes motor emissions of gaseous compounds CO and $NO_x$, and fine particulate matter ($PM_{2.5}$). The block diagram of $PM_{2.5}$ emissions is presented in Figure 3. The computed emission factors in the model depend on the vehicle category and travel velocity. The model classifies light-duty vehicles (LDV) in three separate categories: (i) gasoline-powered cars and vans equipped with catalytic converter, (ii) gasoline-powered cars and vans without a catalytic converter, and (iii) and diesel-powered cars and vans. Similarly, the heavy-duty vehicles (HDV) are classified in three categories: (i) diesel-fuelled trucks with a trailer, (ii) diesel-fuelled trucks without a trailer and diesel-fuelled buses, and (iii) natural gas fuelled buses.

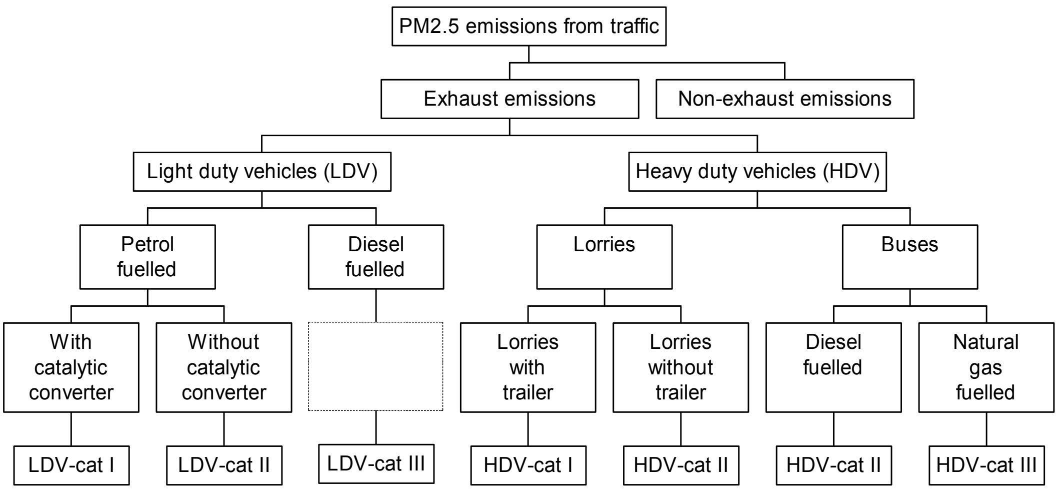


Figure 3. The categories of exhaust $PM_{2.5}$ emissions from traffic. Non-exhaust emissions include emissions from brakes, tyres and resuspension from road surface.

Exhaust emission as a function of the average vehicle travel velocity is fitted separately for each of the above-mentioned six vehicle categories and compounds. The correlations between emissions and velocities are based on the nationally conducted laboratory measurements of vehicle emissions (Laurikko, 1998). The emission factors are polynomial or exponential fittings over the velocity range 1-120 km/h. The emission rate Q (μg $m^{-1}$ $s^{-1}$) of the line source is the product of number of vehicles per hour $n_j$ and emission factor $q_j$ (g $km^{-1}$) summed over the emission categories

$$Q = \sum_j \frac{n_j \, q_j}{3.6} \tag{2}$$

Emission rates are linearly interpolated for the years between 1995 and 2020. The current knowledge of non-exhaust emission factors of fine particulates (including emission from brakes, tyres and road surface) is rather poor. The regression study by Tiitta et al. (2002) suggests that the contribution of non-exhaust emissions is roughly one quarter of the measured $PM_{2.5}$ concentration in the vicinity of the road in summertime conditions. The topic is further discussed in Ch. 7.2.

### 5.3 Dispersion of the gaseous compounds

CAR-FMI uses the general analytical solution of Luhar anf Patil (1989) for the dispersion of gaseous compounds. The general solution is an extension of the special solution of wind perpendicular to the line source (Csanady, 1972). The line source is rotated perpendicular to the wind direction by coordinate transformations i.e. computations are performed in the wind coordinate system as illustrated in Figure 4.

The subscripts “ls” and “w” represent the line source and wind coordinates, respectively. The concentration is computed at the receptor point $R(x_{ls},y_{ls})$ and the physical length of the line source is L. The half-length $p_{ls}$ of the upwind line source depends on the wind direction to the line source and is defined for the receptor points locating in the lee side. The center point O moves to O’ depending on the location of receptor point R and wind direction θ. The displacement of O to O’ causes trigonometric corrections to the $x_w$ and $y_w$ coordinates as well as to the half-length $p_{ls}$. Finally, the physical half-length $p_{ls}$ and the physical emission rate $Q_{ls}$ are transformed to the wind coordinate system, in which the real line source is called virtual.

$$\begin{aligned} p_w &= p_{ls} \left|\sin\theta\right| \\ Q_w &= Q_{ls} / \sin\theta \end{aligned} \tag{3}$$

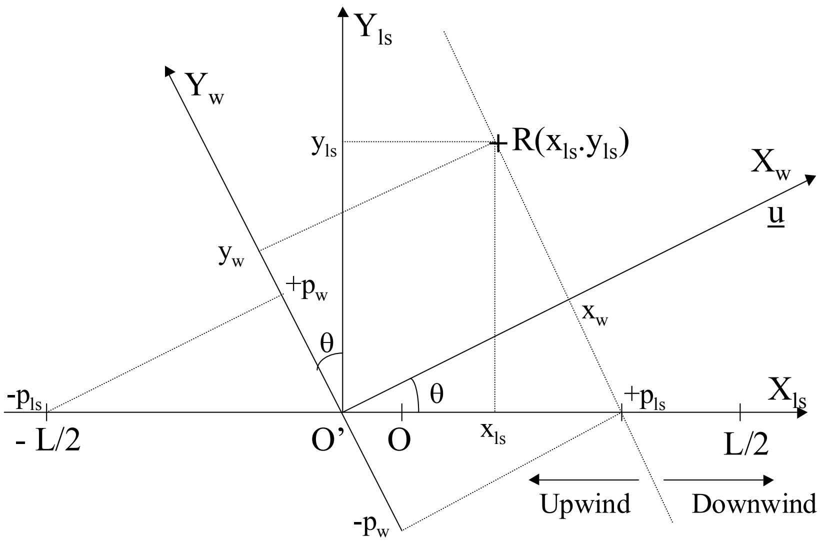


Figure 4. The geometrical orientation of line source L and wind coordinates with subscripts "ls" and "w", respectively. The wind vector u is parallel to the wind coordinate axis $X_w$, while other symbols are defined in the text.

Monin-Obukhov similarity based dispersion parameters $\sigma_y$ and $\sigma_z$ (Gryning et al., 1987) depend on the effective distance d, which is the distance of the line source from the receptor point in wind direction and is defined by

$$d = \min\left(x_w\ ,\ y_{ls}/\sin\theta\ \right) \quad \text{, in which } |\sin\theta| > 0.0876 \qquad (4)$$

The minimum value of the effective distance is 10 m, which is also the minimum perpendicular distance of the receptor points from the centreline of the road. The concept of the effective distance emphasizes the meaning of straight upwind part of the line source with respect to the receptor point. The definition is consistent in case of perpendicular winds and still reasonably good for moderate values of θ, but deteriorates in case of nearly parallel wind directions.

The final result of the concentration at receptor point R is a straightforward result of integration of differential line source elements i.e. Gaussian point source equation in wind coordinate system from $-p_w$ to $+p_w$. The solution has singularities in case of parallel wind direction to the road and wind speed equal to zero, which are avoided by the constraints

- wind direction $|\sin\theta| > 0.08716$

- wind speed $u \geq 0.5$ m/s

Furthermore, the above formulation of the wind direction in the denominator (sinθ-term) is associated with the emission ratio $Q_w$ and the half-length $p_w$ according to equation (4) and not with the wind speed u being always perpendicular to the virtual line source in wind coordinate system. In addition to the coordinate system, this is the main conceptual difference between the Gaussian general dispersion equation of virtual and real line sources, in which the wind parameters of the denominator are associated as the crosswind speed ($u \sin\theta$) being responsible for the dilution.

### 5.3.1 Chemically reactive compounds

The chemistry of nitrogen oxides is closely related to the mixing conditions, because nitrogen monoxide (NO) reacts mainly with ozone ($O_3$), but in inefficient mixing conditions also with oxygen ($O_2$). As a conclusion of the simulations by Galmarini et al. (1995) and recently Chan et al. (2001) the latter reaction is rather usual in the initial mixing zone representing emissions in crossing areas during stagnant conditions. The corresponding reaction is also suggested on the basis the analysis of the urban nitrogen oxide concentrations associated normally with episodic conditions (Harrison et al, 1996).

CAR-FMI uses the cycle NO-$O_3$-$NO_2$, which is solved analytically by Benson (1984). An overview of the processes associated with the expanding Gaussian plume and $NO_x$-chemistry is presented by Hanrahan (1999). The influence of plume dilution is accounted according to the receptor oriented discrete parcel method (Härkönen et al., 1996), which is a modified version of the original discrete parcel method by Benson (1984).

The principal assumptions concerning the chemistry of Gaussian plumes are

- the plume is fully mixed
- there is no interaction between the plumes

The first assumption is partially satisfied due to the contribution from traffic-originated turbulence. However, the assumption becomes questionable in stagnant conditions. A simple approximation of the influence of the overlapping plumes is present in the supercomputer version of the model (Karppinen et al, 2000a), where the input concentrations are accounted for by sorting the computation order from upwind to the downwind direction, though the interactions of separate plumes are treated as chemically independent. The approximation becomes significant in cases of ozone depletion, which is associated with high traffic emissions in urban areas.

The vertical mixing conditions influence the initial concentrations in the chemical reactions of nitrogen oxides. The influence of tailpipe $NO_2/NO_x$ fraction and meteorological conditions on the computed $NO_2$ and $O_3$ concentrations is studied in paper (**3**). The spatial range of pollution is expected to increase in weak mixing conditions, which is typical in sub-arctic regions.

### 5.4 Dispersion of particulate matter with dry deposition

The refined CAR-FMI includes the computation of $PM_{2.5}$ concentration and dry deposition. The size distribution of exhaust particles is determined according to the work of Kerminen et al. (1997) and dry deposition velocity is determined according the work of Nikmo et al. (1997) for three particle size regimes (90 nm, 200 nm and 1000 nm) with weights 0.45, 0.45 and 0.1, respectively. The addition of particles from non-exhaust emissions requires a new regime near to 2000 nm. The influence of relative humidity (RH) on the particle size is accounted for by the semi-empirical formula (Swietlicki et al., 2000), in which the growth factor of particles is an exponential function of RH. The exhaust and non-exhaust emissions by traffic (fig. 3) are combined to line source dispersion model with dry deposition.

Three-dimensional diffusion equation including dry deposition can be separated into a pair of two-dimensional equations (Lin and Hildemann, 1997) as follows

$$C(x,y,z) = Q_w \, G_y(x_w,y_w) \, C_u(x_w,z_w) \,, \tag{5}$$

in which

$C(x,y,z)$ = concentration from a finite line source emissions

$Q_w$ = emission rate of the line source with arbitrary wind direction ($\mu g \, m^{-1} \, s^{-1}$)

$Q$ = emission rate of the line source with perpendicular wind direction ($\mu g \, m^{-1} \, s^{-1}$)

$G_y(x_w, y_w)$ = crosswind dispersion factor for finite line source with arbitrary wind direction

$C_u(x_w,z_w)$ = ambient concentration of contaminant from infinite line source of unit strength with arbitrary wind direction ($m^{-2}$ s).

In case of arbitrary wind direction θ the emission rate is, according to equation (3), $Q_w = Q/\sin\theta$ and the crosswind dispersion factor is

$$G_y(x_w, y_w) = \frac{1}{2}\left[ erf\left( \frac{p_w - y_w}{\sqrt{2}\,\sigma_y(x_w)} \right) + erf\left( \frac{p_w + y_w}{\sqrt{2}\,\sigma_y(x_w)} \right) \right] \tag{6}$$

The constant eddy diffusivity $K_z$ at the effective plume height $h_{eff}$ is

$$K_z = \frac{\kappa\, u_\bullet\, h_{eff}}{\phi_h(\xi)} \tag{7}$$

in which κ is von Karman's constant, $u_*$ is friction velocity and $\phi_h(\xi)$ is the stability function for the heat transfer.

The solution of $C_u(x_w, z_w)$ for a gaussian plume with arbitrary wind direction is

$$\begin{aligned} C_u(x_w, z) = {} & \frac{1}{\sqrt{4\,\pi\, u\, K_z\, x_w}} \left\{ \exp\left[ -\frac{u\,(z - h_{eff})^2}{4\,K_z\,x_w} \right] + \exp\left[ -\frac{u\,(z + h_{eff})^2}{4\,K_z\,x_w} \right] \right\} \\ & - \frac{v_d}{u\,K_z} \exp\left[ \frac{v_d\,(z + h_{eff})}{K_z} + \frac{v_d^2\,x_w}{u\,K_z} \right] erfc\left[ \frac{z + h_{eff} + \dfrac{2\,v_d\,x_w}{u}}{2\sqrt{\dfrac{K_z\,x_w}{u}}} \right] \end{aligned} \tag{8}$$

in which $v_d$ is the dry deposition velocity including the gravitational settling velocity of particles. The emission height is equal to the effective height of the plume $h_{eff}$. It can be shown by dimensional analysis, that in case of Gaussian line source diffusion equation with dry deposition, the relation between diffusivity $K_z$ and dispersion parameter $\sigma_z(x_w)$ is

$$\sigma_z(x_w) = \sqrt{\frac{2\,K_z\,x_w}{u}} \tag{9}$$

Accordingly with the earlier practice, the wind coordinate $x_w$ is replaced by the effective distance d, so that the dispersion equation becomes a physically reasonable approximation of the Gaussian finite line source dispersion equation. The equation of gaseous dispersion in paper (**1**) is a straight consequence of equations (5, 6, 8, 9) when dry deposition is disregarded i.e. $v_d = 0$ and thus the second term in equation (8) represents the correction by dry deposition.

The deposition velocity and particle concentration are solved separately for each size class of particles and the computed concentrations are finally summed up. As a result we can estimate the hourly $PM_{2.5}$ concentration emitted by traffic from the road network.

### 5.5 Statistical analysis of the computed hourly concentrations

Statistical analysis is performed to the computed hourly time-series of CO, NO, $NO_2$, $NO_x$, $O_3$ and $PM_{2.5}$ concentrations at each receptor point. In addition to the statistical parameters used in EU air quality directives, some further analysis is optional in the model. The model computes the highest hourly concentration and the highest daily and monthly means. Also averages of 8-hour, $2^{nd}$-highest daily and yearly concentrations are computed. In addition, the user may select percentiles (1-10 at a time) to be computed (Härkönen et al., 2001). Ozone concentrations in the plume can be computed at the conditions where $NO_2$ has the corresponding statistical parameter, i.e. $99^{th}$ percentile of $O_3$ meaning ozone concentration at conditions with $99^{th}$ percentile of $NO_2$. Also the possibility of the traditional $O_3$ statistics, in which the background $O_3$ concentration in the time-series becomes emphasized, is available for the user.

The output is written out in micrograms per cubic meter at the temperature of 293.1 K in tabular form and in graphical presentation at receptor points in the computing area defined by the user. The results can be analysed and presented utilizing the GIS (Geographic Information System) MapInfo.

## 6 Comparison between predicted and measured concentrations

Predicted and measured gaseous concentrations (NO, $NO_2$, $NO_x$ and $O_3$) in the vicinity of the road were compared in two separate field experiments (**2**; **4**). The hourly means of concentrations in the latter measurement campaign in Elimäki (1995) were monitored simultaneously at three locations at different sides of the road, at three heights (3.5, 6 and 10 m) from the ground surface. The data of traffic volumes (LDV and HDV) as well as on meteorological parameters was based on the on-site measurements. However, the atmospheric stability was determined by the meteorological pre-processing model MPP-FMI (Karppinen, 1997).

The measured and predicted hourly $NO_x$, $NO_2$ and $O_3$ concentrations were analysed statistically using index of agreement (IA), normalized mean square error (NMSE), Pearson's correlation coefficient (COR), fractional bias (FB) and factor of two (F2) at each monitoring level of the monitor locating 34 m from the road. The perfect agreement between measured and predicted concentrations would result in an IA value of 1.0, while, according to Karppinen et al. (2000b), the agreement is perfectly random in case IA is $0.41 \pm 0.01$.

Computed IA values from monitors at distances of 17 and 34 m from the road (N = 587) were 0.83 and 0.82 for $NO_2$ and $NO_x$, respectively, which indicates rather good agreement between measurements and predicted concentrations. The reason for the best agreement (IA = 0.89) between measured and predicted $O_3$ concentrations is due to the emphasizing role of the $O_3$ background concentration in the plume because of low traffic volumes during the measurement campaign. The values of fractional bias of $NO_x$, $NO_2$ and $O_3$ were + 13, - 2 and + 8 %, respectively. The results also suggest that underprediction is greatest at the lowest monitoring level in case of $NO_x$ and $NO_2$.

Sensitivity analysis for the most important parameters was also performed. The contribution of traffic volume to the agreement was insignificant, while the model was sensitive to wind conditions. A detailed study of the influence of wind speed and direction on the computed results by CAR-FMI and a Lagrangian dispersion model GRAL (Oettl et al., 2001a) compared to the Elimäki data is presented by Oettl et al. (2001b). It was observed that CAR-FMI is more sensitive to weak wind speeds and small angles between wind direction and the road than CRAL (fig. 5). This can be expected as the Gaussian finite line source equation indicates the deterioration of robustness of Gaussian models during weak wind speed nearly parallel to the road (**1**). On the other hand, CAR-FMI predicts better than GRAL in case of nearly perpendicular or moderate angles between the road and wind vector. The performance of the CAR-FMI and UDM-FMI modeling system has been evaluated in urban environment by Karppinen et al. (2000b) and Kousa et al. (2001).

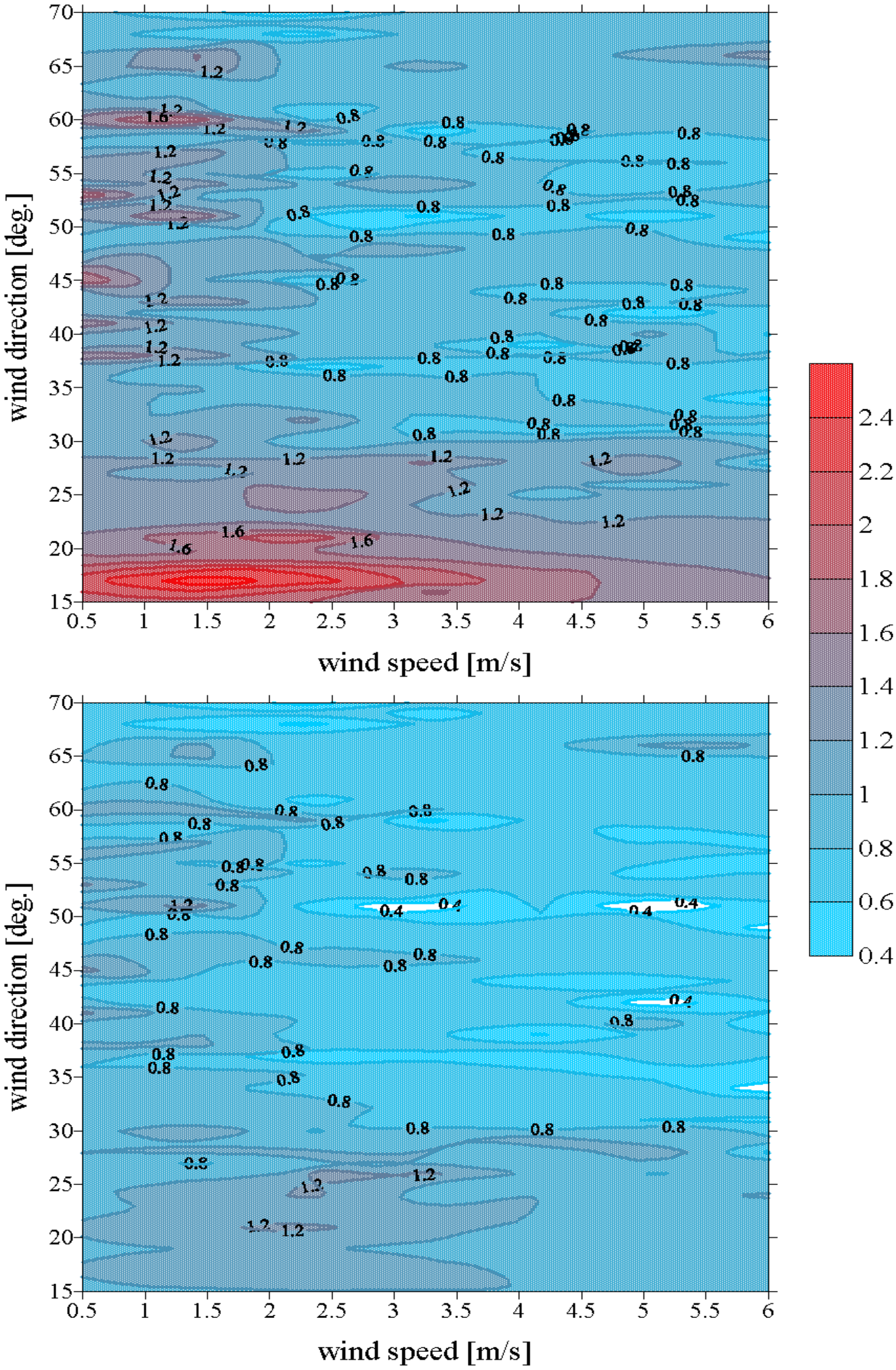


Figure 5. The dependence of the ratio of predicted to observed concentrations (right column) on wind speed and direction for CAR-FMI (top) and GRAL (bottom) models at the measurement site of Elimäki, at the distance of 34 m from the road (After Oettl et al., 2001b).

The meandering may turn the line source temporarily from the upwind to the downwind side with respect to the monitor. Because the Gaussian model assumes a steady state wind direction during one hour, meandering will decrease the monitored concentration compared to predicted values. It is not possible to determine quantitatively the effect of meandering on the performance of the model without fast-response measurements. However, the maximum error can be estimated computationally, if we assume that the lateral wind direction is normally distributed over the hourly mean direction $\theta$ with standard deviation $\sigma_\theta$ and the monitor is located in the lee in case of mean wind direction.

The probability that temporal wind direction $\varphi$ is within the range [a, b] is $P[a \le \varphi \le b] = F(b) - F(a)$, in which F is cumulative normal distribution function. Defining $a = \max(0; \theta - k\,\sigma_\theta)$ and $b = \min(180; \theta + k\,\sigma_\theta)$ in I and II squares of line source coordinate system (see fig. 4), while $a = \max(180; \theta - k\,\sigma_\theta)$ and $b = \min(360; \theta + k\,\sigma_\theta)$ in III and IV squares, respectively. When the value of the parameter $k > 2.54$, the expected wind direction locates within the range [a, b] with probability greater than 99.9 %. The probability plots in I and II squares as a function of mean wind direction $\theta$ with standard deviations 5, 10, 20 and 30 deg are plotted in Figure 6.

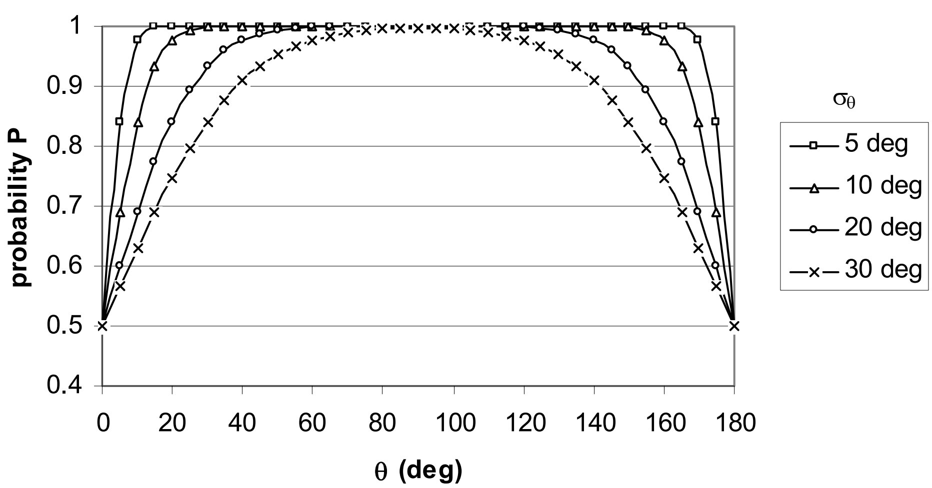


Figure 6. Probability for the downwind location of the monitor in I and II squares, when the lateral wind direction is normally distributed with mean wind direction $\theta$ and the standard deviation $\sigma_\theta$. The 99.9 % confidence intervals are defined in the text.

Standard deviation of lateral wind direction increases with decreasing wind speed and is typically about 25 ± 10 deg in weak wind conditions (< 2 m/s) near the ground surface (e.g. Benson et al., 1986). The probability decreases rapidly in case wind direction turns nearly parallel to the road (0 or 180 deg). Because maximal overprediction becomes proportional to the inverse of probability P, the estimated maximum error is consistent with the results of figure 5 as observed also by Benson et al. (1986). Predicted concentration may be two times higher than measured concentration when wind direction is parallel to the road. As a conclusion, meandering explains substantially the overestimations by CAR-FMI in weak wind conditions with wind vector nearly parallel to the road.

## 7 Studies on particulate matter concentrations

Statistical models are always temporally and spatially limited, which also restricts their use as a predicting tool. Statistical models are used in combination with measurements. The performance of models increases with the averaging time as seen in case of $PM_{10}$ concentration predictions (**5**). The agreement of measured and predicted yearly mean concentrations at the monitoring sites of Helsinki Metropolitan Area Council (YTV) is reasonably good, but deteriorates rapidly towards shorter time averages. This is an indication of the concept of linear regression, in which the best linear fit of $NO_x$ and $PM_{10}$ concentrations (Weingartner et al, 1997) is solved at the expense of individual cases reflecting varying meteorological conditions.

It turned out that addition of meteorological parameters as linear functions to the model (multiple linear regression), does not significantly improve the performance of $PM_{10}$ model. Obviously, most individual processes affecting $PM_{10}$ concentration are nonlinear and independent of current meteorological conditions and traffic. The contribution of traffic to $PM_{10}$ emissions is due to brakes, tyres and abrasion of road surface, but when a threshold wind speed is exceeded, coarse particles (2.5 – 10 μm) are resuspended from all dry surfaces to the air (e.g. Harrison et al., 2001). This is obviously partly the origin of outliers observed when the averaging time is decreased. The contribution of wind speed to resuspension of thoracic particles increases with increasing particle size because of aerodynamics and adhesion between surfaces (e.g. Giess et. al., 1994). As a result, the resuspended fraction of $PM_{10}$ particulates includes mainly coarse particles ($PM_{10-2.5}$).

### 7.1 Estimation of the long-range transported $PM_{2.5}$ concentration

The long-range transported (LRT) background concentration is always needed in local scale evaluations. In case of fine particulates, monitored time-series large enough for modelling purposes are seldom available. A statistical method for estimation of the LRT background concentration of $PM_{2.5}$ is presented in paper (**6**). The method utilizes EMEP (Co-operative programme for monitoring and evaluating of the long-range transmission of air pollutants in Europe) monitoring network and the measured sulfate ($SO_4^{2-}$), sum of nitrate ($NO_3^-$ and nitric acid ($HNO_3$), and the sum of ammonium ($NH_4^+$) and ammonia ($NH_3$) concentrations. The total weighted sum is called the ion sum.

The ion sum (daily mean) is interpolated by inverse distance method from the EMEP stations to the location of $PM_{2.5}$ measurement site. The measured urban (or rural) $PM_{2.5}$ concentrations can be correlated with the ion sum values using linear regression, in which the constant term illustrates the average contribution of all local sources. Consequently, the constant term is expected to change spatially within the same town, while the variation in the slope of ion sum should be small.

The statistical method is tested against time-series (1998-2000) at two monitoring sites of YTV in Helsinki. The first one (Vallila) represents a typical traffic environment located 14 m from a street with average daily traffic volume of 13000 vehicles per day, while the station of Kallio is located 60 m from the nearest street with traffic volume of 7000 vehicles per day, representing an urban background station. The statistical analyses show that the constant term at Vallila representing local sources (like traffic) is clearly greater than at Kallio, while the slopes are the same within 95 % confidence limits.

The modelled long-range transported $PM_{2.5}$ concentrations were compared with the chemically analysed particles at Luukki located 20 km to the NW of Helsinki (Pakkanen et al., 2001). Applying the source apportionment method (Ojanen et al., 1998) the deviation between measured and predicted LRT concentrations were less than 10 %. The measured contributions of LRT component to the total $PM_{2.5}$ concentration at the station of Vallila varies from 60-63 % (Pakkanen et al., 2001), while the corresponding modelled contribution was 64 and 76 % at Vallila and Kallio, respectively (**6**).

The above results indicate that most of the $PM_{2.5}$ concentration even in an urban area is originated by LRT, which is significant in planning of future cost-effective air quality strategies. The method is also important for regulatory model computations, which can utilize the EMEP monitoring network for the background computations of LRT fine particulates in most European countries.

### 7.2 Determining an emission factor for non-exhaust $PM_{2.5}$ emissions

The deterministic regulatory dispersion models require that all emission factors are available. However, it is difficult to obtain those for non-exhaust emissions (including emissions from brakes, tyres and road surface) in case of fine particles. An application of road side measurements, concentrations from exhaust $PM_{2.5}$ emissions by CAR-FMI and the use the statistical models (**5**; **6**) enables the estimation of separate contributions affecting observed $PM_{2.5}$ concentrations by the road side using multiple linear regression (Tiitta et al., 2002). The method includes also a possibility of estimating the non-primary emission factor, which is approximated for a paved road in the summertime conditions.

An estimate for the average non-exhaust emission factor $EF_{ne}$ (g $VKT^{-1}$) in fine particulate region becomes $EF_{ne} = (k - 1)\ EF_e$, where $EF_e$ is a weighted average exhaust emission factor of vehicles, k is the coefficient in multiple regression equation describing the contribution from the traffic caused total emissions and VKT is “vehicle per kilometer travelled”. Correspondingly, the non-primary emission rate is $ER_{ne} = (k - 1)\ ER_e$, where $ER_e$ is the emission rate of tailpipe emissions. Because the emission rate (μg $m^{-1}$ $s^{-1}$) is defined as the product of emission factor and traffic volume (vehicles $h^{-1}$) i.e. ER = EF x TV / 3.6, emission rate of non-exhaust emissions becomes sensitive to the regression coefficient k, primary emission factor and traffic volume.

The monitoring site and measurements and computation of daily average of $PM_{2.5}$ concentrations is described in detail by Tiitta et al. (2002). An overview of the area is also seen in Figure 6, showing the spatial distributions of $PM_{2.5}$ concentrations computed with exhaust emissions factor and the combined emission factor including components from exhaust and non-exhaust emissions in summertime conditions. The monitors used are labeled as A, B, C and D. The abbreviations for the roads are NT (Neulaniementie), ST (Savilahdentie), IT (Iloharjuntie) and VT5 (Valtatie 5 locating about 500 m to the East from the monitors), with mean daily traffic volumes of 2300, 16400, 2400 and 22300, respectively. Savilahdentie contributes about 80 % of the traffic influence on the average $PM_{2.5}$ concentration at the two nearest monitoring points, but increasing distance decreases its weight and increases the relative influence of VT5 from 15 % to 25 % at the most distant site. This is partly a consequence of higher fraction of heavy-duty traffic (15 %) and speed limit 100 km/h on VT5 in spite of greater distance (500 m) compared to Savilahdentie with light controlled travel speed changing from 0 to 50 km/h. Contributions from Neulaniementie and Ilolahdentie are fairly insignificant at the monitoring site with the loads below 5 %.

A weekday average (16 h) of non-exhaust emission factor in summertime conditions is estimated using the two nearest monitors on the downwind side of Savilahdentie (paved two lane road). The average traffic flow of ST was 836 vehicles per hour and the computed average for the emission rate ($ER_{ne}$) and emission factor ($EF_{ne}$) of $PM_{2.5}$ from the road were 26 μg $m^{-1}$ $s^{-1}$ and 0.1 g $VKT^{-1}$, respectively. The reported non-exhaust emission factors for $PM_{10}$ are 3.2-9.3 g $VKT^{-1}$ (Claiborn et al., 1995) and 0.2-3 g $VKT^{-1}$ depending on road type (Venkatram et al., 1999), while the measurements of $PM_{2.5}$ in a road tunnel (Weingartner et al., 1997) suggest that our estimated $EF_{ne}$ for fine particulates would be overestimated by an order of magnitude. The result is expected, because our $EF_{ne}$ includes also emissions from tyres and brakes, which may be a significant contributor near the crossing area.

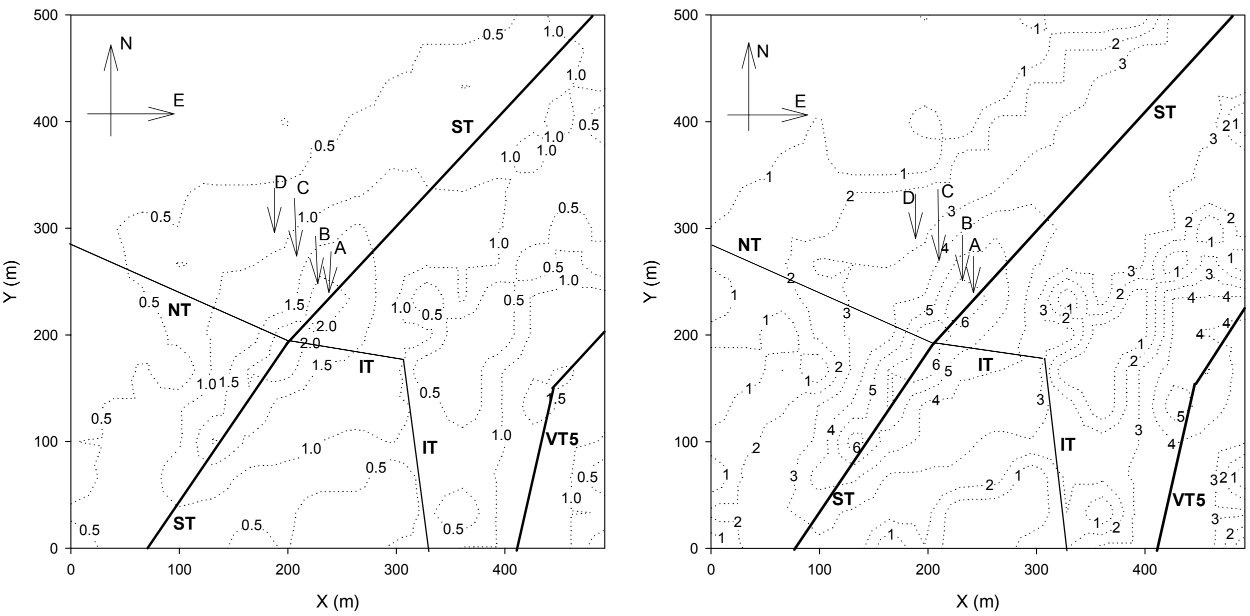


Figure 6. The daily average (16 h) of $PM_{2.5}$ concentrations (μg/$m^3$) by exhaust emissions (left) and by the sum of exhaust and non-exhaust emissions (right) of traffic at the measurement site. The monitors A, B, C and D locate on the line perpendicular to the road (ST) at distances 12, 25, 52 and 87 m from the centreline of the road at 1.8 m height from the ground surface.

## 8 Discussion

The basic principles of the dispersion equations for a finite line source are well documented in connection with several line source models. There are a variety of deterministic model types, which can be used in different environments. Open road dispersion models are based on Lagrangian, Eulerian or Gaussian concepts of atmospheric diffusion and produce reasonably good results in normal terrain for the regulatory use. However, the Gaussian plume model cannot be used in mountainous regions. As a result of the current project, in which CAR-FMI is combined with the street canyon model OSPM (e.g. Hertel and Berkowicz, 1989), a significant improvement in the predictions of downtown areas is expected.

An ideal regulatory open road dispersion model is applicable at the same time to roads and in suburban environment with light controlled crossing areas. The observed deviations in emissions are mainly influenced by driving behavior (De Vlieger et al., 2000), driving cycles (Joumard et al., 2000) and cold start (Laurikko, 1998). However, the statistical uncertainties of modelled emissions in urban areas are within reasonable limits (35 % for $NO_x$) as shown by Kühlwein and Friedrich (2000), while in case of HDV expected deviations are clearly larger (Clark et al., 2002). CAR-FMI applies the widely used method of fitting emissions against the average traveling speed based on laboratory measurements. The procedure excludes the influence of load and obviously under-estimates emissions in suburban environment. The emission factors also depend on the properties of the fuel used (e.g. Clark et al., 2002) and their estimated influence is bound to the year the model run describes, as well as to the average age of the vehicle fleet.

The recent tests, based on the comparison of emissions computed by a traffic simulation model HUTSIM-EMCA (e.g. Höglund and Niittymäki, 1999), suggest that the conditions at the light-controlled crossing can be accounted for in the fitting of traveling speed within reasonable accuracy if the stopping percent is given. Kapur (1988, p.141-4) presents simple solutions for differential equations concerning traveling speed as a function of traffic volume and an empirical parameter, which might be applied to the Gaussian dispersion equation. The information needed for the traffic volume and the unknown parameter is possible to determine by a traffic simulation model.

Open road dispersion models confront further problems when non-exhaust emissions of particulate matter including emissions from brakes, tyres and road surface are to be accounted for. Non-exhaust sources can be analysed separately and fitted against travel speed, or the emissions can be estimated according to the road. However, the monitoring methods used for determination of emission factors in case of resuspension, have not been generally accepted (see e.g. Venkatram, 2000). We proposed an indirect method of estimating non-exhaust component of fine particulate matter from a road network, which is based on the use of modeled traffic contribution (CAR-FMI) and modeled background $PM_{2.5}$ concentration (**6**) and road side measurements (Tiitta et al., 2002).

The basic principles of ambient dispersion have been practically unchanged since M-O scaled dispersion parameters were taken in use. More interest is paid to the influence of traffic on ambient flows i.e. to the initial dispersion parameters as reported in several field experiments (e.g. Chock, 1977; Rao and Sedefian, 1979) and in wind tunnel studies (e.g. Eskridge and Rao, 1986; Rao 2001). According to the works of Postgård and Lindquist (2001) and Borgen et al. (2001) changing weather conditions cause large temperature differences between the road surface and ambient air in Nordic latitudes (58$^{o}$ N) in the layer of lowest few meters from the road surface. Because the physical properties of the road differ from the nearby terrain, the road may cause a significant local perturbation to the vertical stability (Chen et al., 1999) reaching some tens of meters from the road. The duration of perturbation is several hours and so it is a potential explanation of some outliers observed in connection of the model evaluation in the vicinity of the road (**4**).

# 9 Conclusions

This thesis deals with the regulatory modeling of the traffic-originated pollution of inert and reactive gaseous compounds as well as particulate matter. The basic principles of a Gaussian finite line source dispersion model CAR-FMI for dispersion of pollutants from a open road network is described. Also the properties of the refined version of the model are presented, which includes several extensions including the computation of fine particulate matter concentrations with dry deposition. Semi-empirical and statistical models are also presented for thoracic particles ($PM_{10}$) and long-range transported fine particular matter ($PM_{2.5}$), respectively. The CAR-FMI model is applied to study the influence of emission ratio $NO/NO_2$ on ambient air $NO_2$ concentrations.

As an overview of the papers included, the following comments can be made.

- Paper **1** presents the basic structure of the first version (only for gaseous compounds) of the finite line source dispersion model CAR-FMI, while the refined version is described in Ch. 5. The current version of the model includes the prediction of carbon monoxide (CO), nitrogen monoxide (NO), nitrogen dioxide ($NO_2$), nitrogen oxides ($NO_x$), ozone ($O_3$) and fine particulate matter ($PM_{2.5}$) concentrations.
- Papers **2** and **4** describe the evaluation of the model with field measurements. The analysis of the results is continued in Ch. 7.
- Paper **3** is an application of the model and shows the influence of the emission ratio ($NO/NO_2$) on the $NO$-$O_3$-$NO_2$ chemistry in different meteorological conditions. The results suggest that the effect of the emission ratio on $NO_2$ concentration depends in addition to the ambient temperature also on stability and $O_3$ background concentration.
- Paper **5** describes a semi-empirical model for the computation of yearly average of the $PM_{10}$ concentration. It includes the basic equations applied in the estimation of the $PM_{10}$ concentration, description of the model and evaluation against experimental data.
- Paper **6** presents a statistical method for computation of the daily mean of the long-range transported $PM_{2.5}$ concentration. The statistical model in combination with the CAR-FMI model can be used for the estimation of the urban air concentrations of fine particulate matter.

In summary, the paper **1** presents the mathematical structure of the CAR-FMI model, the papers **2** and **4** its evaluation against experimental field data and the paper **3** an application of the model. The papers from **1** to **4** address gaseous pollutants, and papers **5** and **6** particulate matter. Papers **5** and **6** present methods that can be used in combination with the results obtained using the CAR-FMI model.

The new user interface allows the use of own meteorological and background concentration time-series as well as diurnal, daily and seasonal distribution of traffic volumes and emission factors of the vehicle categories. The current emission model is classified according to the vehicle types and the emissions are fitted against the travel velocity. In addition, the statistical treatment of the results is extended so that the user can select any percentile of the concentration. The refined version of the model also includes an option of plume chemistry of the nitrogen oxides for different research purposes. The results are available in the tabular or graphical mode, in which the user interface utilizes 3-D graphics and the Geographic Information System (GIS) MapInfo.

The evaluation of the CAR-FMI model against field experiments is performed at two measurement campaigns. The statistically analyzed results show a considerably good consistency between the measured and predicted concentrations. Also an inter-comparison between CAR-FMI and a Lagrangian model GRAL was performed, which revealed that CAR-FMI is sensitive to weak wind speeds and wind directions nearly parallel to the road. The later is substantially influenced by the meandering effect and can be computationally corrected. The performance of the CAR-FMI and UDM-FMI modeling systems have recently evaluated also in urban environment by Karppinen et al. (2000b) and Kousa et al. (2001).

The first one of the presented statistical models is based on the correlation between $NO_x$ and $PM_{10}$ concentrations. The second statistical method can be used in determining the background concentration of $PM_{2.5}$ in most European countries; it is based on the measured ion concentrations at the EMEP monitoring stations (Co-operative programme for monitoring and evaluating of the long-range transmission of air pollutants in Europe). The combination of roadside measurements, a deterministic line source model CAR-FMI, and the earlier mentioned statistical models (see also Tiitta et al., 2002) is used in connection with an estimation of the emission factor for the non-primary sources of fine particles. However, larger monitored time-series of PM concentrations in different meteorological conditions are needed before the non-exhausted emission factors computed by this indirect method can be reliably applied to the operative use.

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