Dispersion and emission modelling of traffic induced road dust

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1 Introduction

1.1 Background and problem statement

Road traffic has become increasingly important in many urban environments and large cities due to the enlargement of infrastructures resulting from the increase in the world's population. According to a report by the UK Department for Transport, the road traffic in terms of vehicle kilometres was 11 times higher in 2007 than in 1949 in Great Britain (DfT, 2008). However, although road traffic is an important part of people's life, especially in the western world, it acts as an important source of many pollutants having adverse effects on both the environment and health; these pollutants include nitrogen oxides, (NO_x), carbon monoxide (CO), volatile organic compounds (VOC) and particulate matter (PM). According to the Data Warehouse provided by the UK National Atmospheric Emissions Inventory (NAEI), road traffic contributed to ~30%, ~37% and ~18% of the total NO_x, CO and the PM_{10} (particulate matter with diameter smaller than 10 μm) emission levels, respectively, in 2007. The health effects range from airway irritation to more serious effects such as cardiovascular and respiratory diseases and even cancer (Laden et al., 2000; Lippmann, 2007, Health Canada).

As a concequence of the increased pollutant levels resulting from road traffic, industry, wood burning and other urban and rural activities, the levels of NO_x, SO_2, CO, CO_2, PM_{10}, O_3, Pb, etc. are currently legislated according to the European directives 2008/50/EC and 2004/107/EC (EC, 2008; EC, 2005a). In 2015 limit values for $PM_{2.5}$ will also be introduced. However, in many cases these limit values are not met (EEA report, 2005). As a concequence, efforts have been carried out to reduce exhaust emissions, including the introduction of particle filters in diesel vehicles, the use of biodiesel in the diesel mixture, as well as the introduction of diesel/hybrid cars. In addition, European emission standards exist, defined in a set of EU directives, defining acceptable limits for exhaust emissions of NO_x, hydrocarbons (HC), CO and PM of new vehicles sold in the EU. These emission standards are lowered with time and the stages are referred to as "EURO" followed by a number denoting the acceptable emission standard.

Road traffic is of particular importance since it causes emissions both in urban and rural areas, as well as covering both exhaust and road dust emissions. The first mainly covers emissions of NO_x, CO, VOC as well as fine and ultrafine partculate matter ($PM_{2.5}$), while the latter covers direct emission and resuspension of road dust due to road surface wear in addition to the wear of tyres, brakes, clutches, chassis, etc., leading to emissions of PM in the size range 1-70 μm (Myran, 1985). Resuspension is the process where particles that have been deposited onto the road surface, e.g. particles worn off the road, tyres and brakes, as well as sanding and salting particles, become emitted later due to traffic induced turbulence. Several studies have reported that PM emissions due to the mehcanical wear of the road, tyres and brakes as well as resuspension in many cases are equally or more important than exhaust PM emissions (Lenschow et al., 2001; Luhana et al., 2004; Forsberg et al., 2005). The estimation of road dust emissions and exhaust emissions is very different since the processes controlling them are so diverse. For example, NO_x emission data from traffic is considered to be quite certain since the data needed to estimate these emissions are related to vehicle characteristics such as fuel type, vehicle category and traffic density that is relatively straight forward information to obtain. With regard to road dust emissions, however, the processes and physics governing these emissions are many, complex and challenging to model. Other than the vehicle characteristics that govern the exhaust emissions, road surface characteristics and meteorological factors are also parameters that play a significant role. The efforts that have been carried out to reduce the exhaust emissions have little to no effect on road dust emissions, and the sources of road dust are at present unregulated. Therefore, since the sources of road dust contribute significantly to the ambient PM levels, as well as not being well understood, these sources need our attention.

1.2 The objectives of this study

As indicated above, understanding the effect of meteorology, road and traffic characteristics, as well as the effect of abatement measures on emissions and concentration levels is crucial with regard to traffic originated pollutants due to their importance in terms of health and environmental effects. This requries access to up-to-date air quality models that are robust and able to reproduce the real world in a satisfying manner. To achieve this, model development is an important action; we must strive to increase the robustness and quality of all types of air quality models. This thesis is concerned with the evaluation and inter-comparison of line source dispersion models as well as the development and evaluation of a new generalised road dust emission model. The former study is concerned with the analysis of modelled and measured concentration levels at near-road stations, where we have used NO_x as a tracer, since it can be considered inert in the short time scales associated with the relatively short distances from the road. With regard to the latter study, models currently being developed to predict road dust emissions suffer from a lack of important information that is required for their useful implementation. Hence, we have developed and tested a more generalised road dust emission model, containing thorough process descriptions of the most important parameters involved.

Emissions and concentrations of pollutants are closely linked; a good emission model can aid the development of a dispersion model, and vice versa. Furthermore, since the quality of the model can never exceed the quality of the measurements, it is crucial to conduct measurement campaigns with good observations of the most important pollutants as well as of parameters describing meteorology, traffic and pavement characteristics. Then, the understanding of what controls the emissions and concentration levels becomes clearer and analysis and model deveopment can be conducted. In the following, we go deeper into the theory of dispersion of traffic originated pollutants (section 2) and road dust emissions (section 3), along with small reviews of some developed traffic dispersion models (section 2.2) and road dust emission models (section 3.4).

2 Dispersion of pollutants from open road networks

Traffic exhaust and road dust emissions are the most predominant sources of air pollution, especially in urban environments. In the 1970's, the General Motors (GM) were in the earliest field of collecting experimental data to understand the processes controlling dispersion and nearby concentration levels due to pollutant emissions from traffic (Cadle et al., 1976). From that time onwards many studies have been concerned with analysis of experimental data, as well as with modelling of air pollutant levels at receptor points relatively close to the road. The models range from simple Gaussian line source models to more complex models in two and three dimensions. Some extensive reviews have been carried out in regard to modelling of vehicular exhaust and line source dispersion models (e.g. Sharma and Khare, 2001; Sharma et al., 2004; Nagendra and Khare, 2002). In the following sections, some theoretical aspects regarding the spatial and temporal distribution of any pollutant released into the atmosphere are given, with emphasis on the Gaussian plume equation, followed by a small review of operational traffic dispersion models.

2.1 Approaches to describe the dispersion of traffic originated pollutants and turbulence

2.1.1 The Gaussian plume equation

A widely used equation is the Gaussian plume equation, giving the concentration at (x, y, z) on time t of a pollutant emitted from a point source:

$$C = \frac{Q}{2\pi u_h \sigma_y \sigma_z} \cdot f \cdot (g_1 + g_2 + g_3) \tag{1}$$

where C (µg m^{-3}) is the concentration of the pollutant at the receptor point located at x m downwind from the road, y m crosswind from the emission source point and z m above ground level; Q (µg s^{-1}) is the source emission rate, u_h (m s^{-1}) is the horizontal wind velocity along the plume centerline and σ_y and σ_z (m) are the horizontal and vertical standard deviations of the emission distribution, respectively. $f = \exp\left(-y^2/2\sigma_y^2\right)$ and $g = g_1 + g_2 + g_3$ are the crosswind and vertical dispersion parameter, respectively;

$g_1 = \exp\left(-\frac{(z-H)^2}{2\sigma_z^2}\right)$ is the vertical dispersion with no reflection,

$g_2 = \exp\left(-\frac{(z+H)^2}{2\sigma_z^2}\right)$ is the vertical dispersion due to reflection from the ground and

$$g_3 = \sum_{m=1}^{\infty}\left\{\exp\left(-\frac{(z-H-2mL)^2}{2\sigma_z^2}\right) + \exp\left(-\frac{(z+H+2mL)^2}{2\sigma_z^2}\right)\right.$$

$\left. + \exp\left(-\frac{(z+H-2mL)^2}{2\sigma_z^2}\right) + \exp\left(-\frac{(z-H+2mL)^2}{2\sigma_z^2}\right)\right\}$ is the vertical dispersion due to reflection from inversion lid aloft. H (m) is the emission plume centerline above ground level and L (m) is the distance from ground level to the bottom of the inversion aloft. For a road of finite length (i.e. line source), eq. 1 must be integrated over the length of the road. The conditions under which the Gaussian plume equation is valid are highly idealized due to the assumptions of homogeneous and stationary turbulent flow, no vertical profile in turbulence and wind speed, as well as the assumption that the spread of each puff/eddy is small compared to the downwind distance it has travelled (slender plume approximation). Turbulent flow is seldom fully homogeneous and stationary, and the wind speed varies a lot with height near the surface. Hence, the ability of the model to reproduce the measured concentrations of some pollutant in a satisfactory manner rests on the estimation of the disperson coefficients (the standard deviations of the emission distribution), σ_y and σ_z, and how the effective dilution velocity, u_h, is modelled. The dispersion coefficients are important parameters as they determine the amount of dispersion and hence, the concentration at a given receptor point. Furthermore, other than the natural atmospheric turbulence, the turbulence created by the moving vehicles on the road is highly important close to the source. Farther away from the source, the atmospheric dispersion plays an increasingly bigger role relative to the traffic produced turbulence.

The wind speed, u_h, in eq. 1 represents the wind speed at the height of the emission source; hence, we call it the effective dilution velocity. However, since the observed wind speed usually is measured at a higher level than the height of the source, h, u_h has to be estimated. This can be carried out using the mean wind speed or applying Monin-Obukhov similarity theory represented by the logarithmic wind speed profile to extrapolate the measured wind speed down to the height of the emission source.

Eq. 1 has been deduced following a Lagrangian approach, i.e. the concentration changes are described relative to a moving fluid. However, when following the Eulerian approach, in which the behaviour of a fluid particle is described relative to a fixed coordinate system, K-theory (also called mixing length theory) is used, in which constant eddy diffusivities, K_{yy} and K_{zz}, are applied:

$$C = \frac{Q}{4\pi\left(K_{yy}K_{zz}\right)^{1/2}x}\cdot\exp\left(-\frac{u_h}{4x}\left(\frac{y^2}{K_{yy}}\right)\right)\cdot\left(g_1' + g_2' + g_3'\right) \tag{2}$$

where K_{yy} and K_{zz} are the horisontal and vertical eddy diffusivities, respectively, and g_1', g_2' and g_3' are as in eq. 1, but with $\sigma_{y,z}^2 = 2K_{yy,zz}t$.

More details on the derivation of eqs. 1 and 2 are well described in Seinfeld and Pandis (1998).

2.1.2 Lagrangian dispersion technique

In Lagrangian dispersion models, the dispersion is calculated by computing the statistics of the trajectories of an ensemble of particles. An important parameter in Lagrangian dispersion models is the autocorrelation function, also called the Lagrangian turbulent velocity correlation coefficient. Using this function allows calculating the dispersion parameters. In most cases, the following exponential form of the autocorrelation function, $\rho_{Li}(\tau)$ is used:

$$\rho_{L_i}(\tau) = \exp\left(-\frac{\tau}{\left(m^2+1\right)T_{Li}}\right)\cos\left(-\frac{m\tau}{\left(m^2+1\right)T_{Li}}\right) \tag{3}$$

where τ is the time lag, m is the loop parameter controlling the meandering oscillation frequency associated with the horizontal wind, T_{Li} is the Lagrangian time scale and i = 1,2 and 3 are the components in the x, y and z directions.

2.1.3 Large eddy simulation technique

Large eddy simulation (LES) is a numerical technique used to solve the partial differential equations governing a turbulent flow. Based on the theory of self similarity (Kolmogorov, 1941), which states that large eddies are dependent on the flow geometry, while the smaller ones are self similar, large scale motions are calculated, while the effect of smaller scales are modelled using a sub-grid scale model.

2.2 Modelling concentration and dispersion of traffic originated pollutants

The types of air pollutant emission sources are commonly characterized as point, line, area or volume sources. Eq. 1 can be applied with respect to all these kinds of sources; however, with regard to the modelling of traffic originated pollutants, the most common approach is to consider the road as a line source, since the road can be considered as a linear, one-dimensional line. Furthermore, differences in the local terrain imply different approaches to describe dispersion. Generally, in urban environments with high buildings surrounding the road (street canyons) the concentration of the traffic originated pollutants are higher than in open areas; the high buildings prevent the dispersion of pollutants outside the urban environment. In rural areas with fewer buildings or other obstacles surrounding the road, usually associated with highways, there are few obstacles preventing dispersion of the pollutants, and the concentration at a specific location is lower than in a street canyon. In this study, we have focused on open road line source modelling in rural environments. In the following, a review of some operational open road line source models is given.

2.1.4 Review of some traffic dispersion models

Table 1 contains a summary of some operational open road line source models and their characteristics.

Table 1: *Summary of a selection of operational open road line source models and their characteristics.*

	Model type,	**Parameterisation of σ_y and σ_z**	**Parameterisation of traffic produced turbulence**	**Parameterisation of u_h**	**Reference**
OML-Highway	Semi-empirical, analytical for crosswind, empirical for along-wind directions	Pasquill-Gifford dispersion curves (Gifford, 1961)	Based on traffic produced turbulent kinetic energy, traffic density, traffic speed and vehicle size	Monin-Obukhov similarity theory	Jensen et al. (2004), Olesen et al. (2007)
The HIWAY models	Numerical, Richardson extrapolation	Pasquill-Gifford dispersion curves (Gifford, 1961)	Dependent on aerodynamic drag, U_c (Petersen, 1980)	Monin-Obukhov similarity theory	Petersen (1980), Rao and Keenan (1980)
CAR-FMI	Analytical	Pasquill-Gifford dispersion curves (Gifford, 1961)	Dependent on aerodynamic drag, U_c (Petersen, 1980)	Monin-Obukhov similarity theory	Härkönen (2002), Kukkonen et al. (2001)
The CALINE models	Semi-empirical, separates between crosswind and along-wind directions	Pasquill-Gifford dispersion curves (Gifford, 1961)	Dependent on aerodynamic drag, U_c (Petersen, 1980)	Monin-Obukhov similarity theory	Beaton et al. (1972), Benson et al. (1986)
WORM	Numerical, Gaussian quadrature	Choice between CAR-FMI, HIWAY2 or OML-Highway	Choice between CAR-FMI, HIWAY2 or OML-Highway	Monin-Obukhov similarity theory	S.E. Walker, personal communicat ion
ROADW AY	Empirical, K-theory	Pasquill-Gifford dispersion curves (Gifford, 1961)	-	Monin-Obukhov similarity theory	Eskridge and Thompson (1982)

Based on the General Motors (GM) experimental data, a number of traffic dispersion models were tested and developed in the USA; The California model (CALINE) is a Gaussian based model developed in the 1970's (Beaton et al., 1972), the latest version being CALINE 4 (Benson et al., 1986). It separates between crosswind directions and wind directions parallel to the road and uses a semi-empirical solution to eq. 1 which can be used for several road conditions, including intersection, bridge and depression. The model treats the road as a series of finite line sources located normal to the wind direction. In the later versions the model was developed with regard to dispersion of inert pollutants from more complex road environments. σ_y is calculated using the standard deviation of the wind direction, σ_θ (Draxler, 1976), while σ_z is calculated using traffic produced, thermal and atmospheric turbulence.

Another Gaussian based model developed at the US EPA is the HIWAY models, of which the first version was developed in the 1970's (Zimmermann and Thompson, 1975). The model assumes that for "at-grade" highway each lane is comprised out of several finite line sources with uniform emission rate, and the calculation of the downwind concentration depends on stability conditions. For a "cut section" the top of the cut is modelled as an area source, divided into ten line sources of equal strength. In the 1980's HIWAY underwent several revisions, i.e. HIWAY2 (Petersen, 1980), HIWAY3 (Rao and Keenan, 1980) and HIWAY4. σ_y and σ_z are calculated as a combination of atmospheric and traffic produced turbulence; during the development of the model, new expressions for traffic produced turbulence were introduced:

$$\sigma_{z0} = 3.57 - 0.53U_c \tag{4}$$

$$\sigma_{y0} = 2\sigma_{z0} \tag{5}$$

where σ_{y0} and σ_{z0} represent the traffic produced turbulence in the lateral and vertical direction, respectively, and U_c is the aerodynamic drag depending on the wind speed and the angle between the wind speed vector and the road. Also, a modification of the Pasquill-Gifford curves (Gifford, 1961) is applied, using the stability regimes stable, unstable and neutral conditions. In order to integrate eq. 1 along the line source, the trapezoidal rule together with Richardson extrapolation is used. Another model based on the GM experimental data is ROADWAY (Eskridge and Thompson, 1982), which is a model finite difference model using K-theory (eq. 2). The model emphasizes the importance of traffic turbulence using vehicle wake theory in which the surface layer is described by surface layer similarity theory.

Other than the models developed in the USA, several other models have more or less successfully described traffic dispersion and pollutant levels near trafficked roads; among these are CAR-FMI, OML-Highway, WORM and GRAL. CAR-FMI (Contaminants in the Air from a Road) (Härkönen, 2002; Kukkonen et al., 2001) has been developed at the Finnish Meteorological Institute (FMI). It consists of an emission model, a dispersion model and a statistical analysis of the estimated time series of the concentrations. In the dispersion model, in order to calculate the pollutant levels due to road traffic, eq. 1 is used, i.e. the model regards the road as a finite line source and eq. 1 is then integrated along the length of the line source as is the case with the HIWAY model. However, CAR-FMI assumes total reflection from the ground, ignores reflection at the mixing height and allows any wind direction with respect to the line source (Härkönen, 2002). The atmospheric dispersion coefficients are modelled using Pasquill-Gifford curves, while traffic produced turbulence is modelled according to eqs. 4 and 5.

OML-Highway model is a part of the OML model system, which is a Gaussian atmospheric dispersion model developed at the National Environmental Research Institute, Denmark, in the 1990's (Jensen et al., 2004; Olesen et al., 2007). Several model versions have been developed to assess pollutant emissions from point, area and line sources, such as industrial activity, domestic heating, traffic, as well as the assessment of ammonia depostition and regulation of odour. OML underwent a review in 2005/2006, where the model performance was evaluated using more experimental data than previously. To estimate dispersion and concentration levels due to highway traffic a version called OML-Highway has been developed. It calculates the concentration at a receptor point using a double integral in the crosswind and along-road wind directions. Eq. 1 is integrated along the line source for crosswind directions. For wind directions parallel to the road it uses the Romberg integration technique (Press et al., 1992) with Richardson's extrapolation of the trapezoidal rule. The main difference from other Gaussian line source models is that it bases the parameterisation of traffic produced turbulence on traffic produced turbulent energy, e (m^2 s^{-2}), which is a function of traffic number, speed and vehicle size. The traffic produced turbulence, here denoted σ_0, decays in a exponential manner with distance from the source:

$$\sigma_0(t) = \sigma_{initial} + u_{TPT}\tau\left[1 - \exp\left(-\frac{t}{\tau}\right)\right] \tag{6}$$

where $\sigma_{initial}$ = 3.2 m is the initial dispersion near the source, $u_{TPT} = e^{1/2}$ (m s^{-1}) is a velocity parameter related to traffic produced turbulence (TPT), τ (s) is the time scale for decay of σ_0 and t (s) is the transport time (Jensen et al., 2004). This parameterisation has proved to work well even for low wind speed conditions (Jensen et al., 2004; Ketzel, M., personal communication).

The WORM model (Weak Worm Open Road Model) is a newly developed model at the Norwegian Institute for Air Research (S.E. Walker, personal communication). Based on an inter-comparison between four open road line source models in the Nordic countries conducted by Berger et al. (2010), the model now contains parameterisations from the models OML-Highway, HIWAY2 and CAR-FMI. The objective is then to choose a set of parameterisations for a given meteorological condition and traffic situation, to obtain the best result with regard to dispersion and observations. In addition, WORM integrates eq. 1 using the Gaussian quadrature method (Kythe and Schäferkotter, 2005), which is also accurate for wind directions parallel to the road.

GRAL (GRAz Lagrange model) (Oettl et al., 2001a) is a Lagrangian dispersion model making use of the autocorrelation function for the horizontal wind component according to the model of Wang and Stock (1992); the new positions for a particle at time $t + \Delta t_h$ are then given by

$$x(t + \Delta t_h) = x(t) + [\overline{u} + u'(t + \Delta t_h)]\Delta t_h \qquad (7)$$

$$y(t + \Delta t_h) = y(t) + [\overline{v} + v'(t + \Delta t_h)]\Delta t_h \qquad (8)$$

where

$$u'(t + \Delta t_h) = \rho_u u'(t) + \sigma_u (1 - \rho_u^2)^{1/2} \chi \qquad (9)$$

$$v'(t + \Delta t_h) = \rho_v v'(t) + \sigma_v (1 - \rho_v^2)^{1/2} \chi \qquad (10)$$

where x and y are the particle position at time t and $t + \Delta t_h$, $\overline{u}$ and $\overline{v}$ are the mean components of the wind vector in the x and y directions, respectively, u' and v' are the velocity fluctuations, σ_u and σ_v are the standard deviations of the velocity components, χ are random numbers with zero mean and a standard deviation equal to 1, ρ_u and ρ_v are the intercorrelation parameters and Δt_h is a random time step for which the horizontal velocity fluctuations remain constant. Vertical dispersion is treated according to Franzese et al. (1999), which satisfies the well-mixed criterion under stationary and horizontally homogeneous turbulence:

$$w(t+\Delta t_v) = a(w,z)\Delta t_v + [C_0 \varepsilon(z)]^{1/2}\, dW + w(t) \tag{11}$$

$$z(t+\Delta t_v) = z(t) + w(t+\Delta t_v)\Delta t_v \tag{12}$$

where w is the vertical velocity of a particle, $C_0 = 2.1$ is a universal constant, $\varepsilon(z)$ is the ensemble-average rate of dissipation of turbulent kinetic energy, dW are the increments of a Wiener process with zero mean and variance Δt_v. Since the model takes into account enhanced horizontal meandering, it has proved to perform well under low winds (Oettl et al., 2001b). For a more detailed description of this model, the reader is referred to Oettl et al. (2001a).

In order to improve traffic dispersion models, proper evaluation against several good quality experimental data as well as comparisons with other similar models are crucial actions. Marmur et al. (2003) compared CALINE4 and HIWAY2 based on their application on an "at-grade" and a "cut/depressed" road section in Israel; when applied to "at-grade" road sections, their performance was similar with respect to the measurements. For "cut/depressed" sections, however, HIWAY2 performed better during unstable conditions, while CALINE4 better predicted peak concentrations. The main problem with the Gaussian approach of describing dispersion and near-road pollutant levels from an open road network, is the disability to accurately reproduce the dispersion characteristics and concentration during low wind speed conditions and/or for wind directions parallel to the road. A regular problem during low wind speed conditions is overestimation of the pollutant levels and hence, underestimation of the dispersion. This mainly happens because of unsufficient or lack of proper description and parameterisation of traffic produced turbulence, which has proved to by important, especially near the source. Levitin et al. (2005) evaluated and compared CALINE4 and CAR-FMI against measurements near a major road in Finland. They found that both models correlated well with measurements, although the performance was better at a larger distance from the road. However, for low wind speed conditions and wind directions close to parallel to the road, the performance of both models deterioated, i.e. the concentrations were overestimated. In Oettl et al. (2001b) CAR-FMI was compared with the Lagrangian dispersion model GRAL using the same dataset as that in Levitin et al. (2005).

For low wind speeds and wind directions parallel to the road, where the performance of CAR-FMI deterioated, GRAL managed to reproduce the measurements better. This was found to be due to GRALs treatment of flow meandering due to traffic produced turbulence, based on the formulation of Wang and Stock (1992), which leads to increased dispersion during low wind speed conditions.

3 Emission of suspended PM from traffic

The study of airborne particulate matter (PM) is an area of interest due to the implications on health and climate. Emissions from non-exhaust sources (road wear, break and tyre wear) is one of the major contributors to poor air quality, particularly in countries where traction control methods such as the use of studded tyres, sanding and salting are applied during winter time (Kupiainen, 2007; Norman and Johansson, 2006; Gustafsson et al., 2008a,b). The health effects related to PM_{10} include respiratory morbidity (wheeze, reduced lung function) and mortality, cardiovascular morbidity and mortality, and cancer (Downs et al., 2007; McCreanor et al., 2007; Laden et al., 2000). According to Clean Air For Europe (EC, 2005b), long-term exposures to $PM_{2.5}$ were associated with ~350 000 premature annual deaths in 2000. Predicting road dust emissions, through the use of models, is a major challenge and has only been addressed to a limited extent. In addition, although the emissions of exhaust particles are regulated by law, the sources of road dust are at present unregulated. As mentioned, significant efforts have been made over the years in order to reduce exhaust emissions. As a result, road dust emissions are at present almost equally, or in some cases, more important than exhaust emissions. According to Luhana et al. (2004), in the Hatfield tunnel in the UK, diesel and petrol exhaust accounted for ~47% of the net PM_{10} concentrations, while the resuspension of road dust, and emissions due road, tyre and brake wear together amounted to ~46% of the net PM_{10} concentrations. Lenschow et al. (2001) reported that ~50% of the PM_{10} emissions caused by traffic are attributable to road dust resuspension, and ~50% attributable to exhaust and emissions due to tyre wear at a traffic site in Berlin. In addition, the use of studded tyres increase the PM levels dramatically; according to Forsberg et al. (2005), due to the use of studded tyres, ~90% of the PM_{10} levels during springtime in Stockholm are mechanically generated particles due to road wear and resuspension. Furthermore, the existing limit values for PM_{10} are often exceeded. According to the European directives 2008/50/EC (EC, 2008), daily averages cannot exceed 50 $\mu g\ m^{-3}$ more than 35 times a year, and the annual average cannot exceed 40 $\mu g\ m^{-3}$. In many cities in Europe these limit values are not met. For example, in a case study in Stockholm the daily averages of the three streets involved exceeded the limit value during all the years from 1999-2004 (Norman and Johansson, 2006). The same has been the case in Trondheim, Norway, in the years 1999-2007, although the PM_{10} levels show a downward trend due to a reduction in the share of vehicles using studded tyres, as well as development of more durable pavements (Snilsberg, 2008).

This section is concerned with the physical processes and parameters governing road dust emissions, followed by a review of regulatory road dust emission models including their characteristics and shortcomings.

3.1 What is road dust?

Road dust is dust generated as a result of the mechanical wear of the road surface. In addition, tyre, brake and clutch wear, as well as corrosion of vehicle components and street furniture, de-icing salt, grit from traction sand, etc are important non-exhaust emission sources. In this thesis we use the term "road dust" since we do not cover the other non-exhaust sources in any great detail. Road dust emissions cover both direct emissions due to mechanical wear and resuspension of dust on the road surface and shoulder. While exhaust particles mainly lie in the nucleation or accumulation size mode, i.e. particles with diameters $d < 50$ nm and $0.05\ \mu m < d < 2.5\ \mu m$, respectively, particles attributed to mechanical wear that become emitted as TSP, typically lie in the size range 1-70 µm (Myran, 1985), with a peak at the coarse size mode, i.e. $2.5\ \mu m < d < 10\ \mu m$ (Thorpe et al., 2007). Particles with diameters up to 100 µm can become suspended, but $d \approx 20\ \mu m$ is considered a size limit for particles if they are to remain airborne for longer periods of time, as these particles typically have settling velocities smaller than the vertical velocities in the turbulent boundary layer (Kupiainen, 2007).

The composition of road dust varies with location, season and other parameters. Typically, it is composed of 60% sand, 20% fine sand and 20% silt (Luhana et al., 2004). However, traces of lead, copper, calsium and zink from tyre and brake wear are also present. In a test study conducted by Kupiainen et al. (2005), where traction sand was applied on the pavement, 90% of the TSP (total suspended particles) were aluminosilicates, which are found in hornblende, a tracer for pavements, in addition to quartz and K-feldspar, which serve as tracers for anti-skid aggregates. Hence, the formed dust consisted of dust from both the pavement and the traction sand, with the amount of quartz and K-feldspar increasing with increasing load of traction sand. Gustafsson et al. (2008b) reported that as a result of studded tyre use, mineral particles from the pavements (Al, Si and K) totally dominate the wear, regardless of pavement type.

3.2 Sources and formation of dust

The factors and processes affecting road dust emissions are many and complex. Fig. 1 presents a simplified diagram describing the main sources and processes controlling road dust and other non-exhaust emissions, as well as their dependencies on vehicle, traffic and pavement characteristics and meteorology; particles generated from road, tyre and brake wear, as well as deposition of any external sources as sanding, salting and exhaust will either become directly emitted, determined by the function f_{direct}, or deposited onto the surface, determined by the function ($1-f_{direct}$). A fraction of the resulting dust load on the road surface and shoulder will eventually become resuspended, controlled by the functions g_{road} and $g_{shoulder}$, respectively, while some particles will be removed due to runoff, controlled by the function g_{runoff}. The direct and resuspended emissions, as well as PM from any other sources (long-range transport from industry, wood burning, other road environments, etc.) all contribute to the total airborne PM (TSP). The main factors controlling road dust emissions are also presented in the diagram. In the following the processes controlling road, tyre and brake wear, as well as road dust resuspension will be discussed.

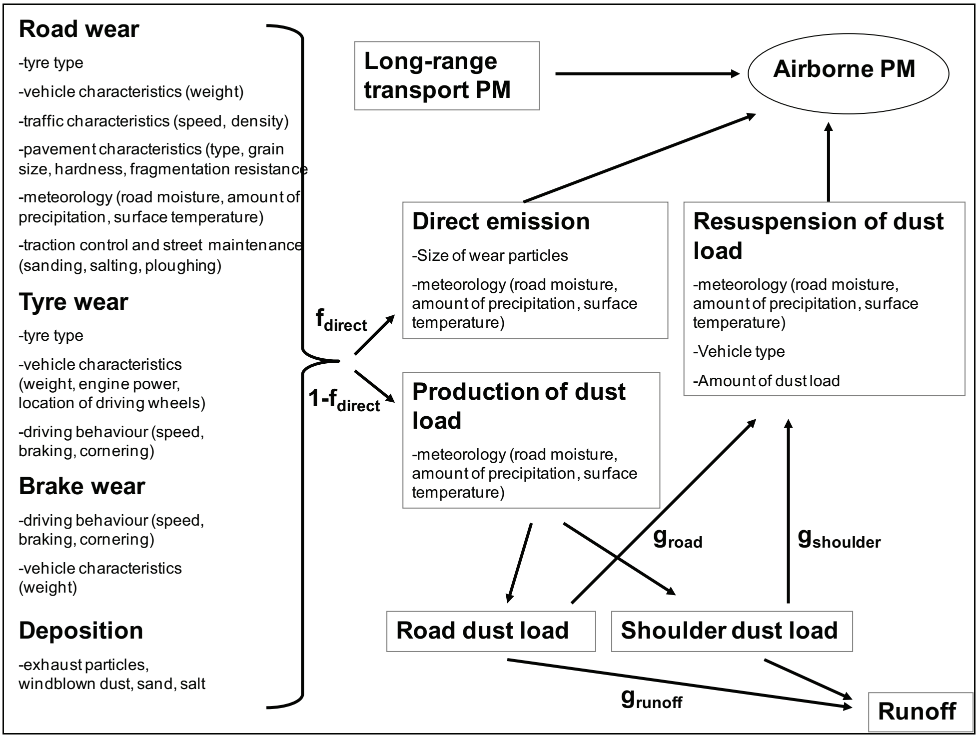

Figure 1: *Main sources and factors affecting road dust emissions.*

3.2.1 Road, tyre and brake wear

Road wear is closely linked to tyre wear as it occurs as a result of the tyre-road-interaction. The wear rates depend on numerous factors such as tyre type, vehicle type, vehicle speed, road geometry, surface properties, driving behaviour and meteorology. A range of road wear rates have been reported in the literature; 3.8 mg veh^{-1} km^{-1} (Muschack, 1990), 90-440 mg veh^{-1} km^{-1} in New Zealand (Kennedy et al., 2002) and 7.9-38 mg veh^{-1} km^{-1} depending on vehicle category (Lükewille et al., 2001). However, in the Nordic countries, North America and Japan where traction control methods such as studded tyres and/or dispersion of traction sand or de-icing material on the road are applied during the winter to increase traffic safety, the road wear is much more pronounced. In Norway (Oslo) and Sweden (southern parts) the share of studded tyre use during winter time is ~20% and ~40%, respectively (Norwegian Public Roads Administration and Omstedt et al., 2005), while in Finland the use rate is ~80% (Kupiainen, 2007). According to the Norwegian Institute for Air Research (NILU, 1996), studded tyres wore off ~250 000 tonnes of asphalt each year in Norway in the mid 1990's. Lindgren (1996) reports a wear rate of 24 g veh^{-1} km^{-1} for studded tyres during winter time in

Sweden. Another traction control method widely used in e.g. the Nordic countries is traction sand/anti-skid aggregate which increases the wear of the pavement when the tyres interact with the sand on the road surface; according to Kupiainen et al. (2005), loose particles on the road surface act as studs when tyres interact with the surface, leading to increased road wear. In addition, the “road-sand-tyre” interaction may cause breakage of the larger particles into smaller resuspendable sizes. This process is called the sandpaper effect. Kupiainen (2007) studied this phenomenon and reported that aggregates with high resistance to fragmentation resulted in high relative contributions from the pavement. In the same study the PM concentrations increased with increasing amount of anti-skid aggregate. According to Tervahattu et al. (2006), the abundance of particles from the pavement is highly dependent on the fragmentation resistance and grain size of the sanding aggregates, as well as the hardness of the pavement. In Kupiainen et al. (2005) the use of traction sand and studded tyres gave the highest particle concentrations, followed by friction tyres and the same traction sand. Furthermore, studies have shown the impact vehicle speed has on road wear; Kupiainen (2007) carried out indoor road simulator tests, and found that at a speed of 15 km h^{-1} studded tyres resulted in an average PM_{10} concentration that was four times higher compared with non-studded winter tyres and that the effect became more pronounced at higher vehicle speeds due to the increased tyre and road wear. Gustafsson et al. (2008b) found that the use of studded tyres resulted in particle concentrations 60-100 times higher (at speeds of 30, 50 and 70 km h^{-1}) than when friction tyres were used. Road moisture also increases the road wear; according to Folkeson (1992), the road wear is two to six times higher for a wet surface than for a dry surface. This is mainly because under wet conditions, particles attach more easily to the surface, rather than becoming emitted, leading to an increase of the road dust depot. Hence, due to the sandpaper effect a wet surface may cause enhanced wear.

Tyre wear occurs as a result of the interaction and frictional energy developed at the road-tyre interface (Veith, 1995), and the wear rate is dependent on numerous factors such as tyre characteristics (size, tread depth, tyre pressure and temperature), vehicle characteristics (weight, engine power and location of driving wheels), road surface characteristics and driving behaviour. In general, tyre wear is enhanced during acceleration, braking and cornering. A new tyre on an average European car will loose ~1-1.5 kg of its weight before it must be replaced. This corresponds to a service lifetime of three years or 50 000-60 000 vehicle km (Boulter, 2005). A range of values have been reported in the literature; e.g. Legret and Pagotto (1999) reported a tyre wear rate of 68-136 mg veh^{-1} km^{-1}, depending on vehicle category, while 16 mg veh^{-1} km^{-1} has been reported by Lee et al., 1997). According to Luhana et al. (2004), the tyre wear decreases as the mean trip speed increases, mainly because of more frequent braking and cornering in urban areas than on highways where the vehicle speed is higher and more constant.

Brake wear occurs during forced deceleration due to large frictional heat generation (Luhana et al., 2004), and hence increases in areas where braking is needed such as crossings and slopes. In addition, some factors affecting brake wear are type of brakes (disc brakes or drum brakes), composition of the friction material and driving behavior. Under normal usage, it is expected that front disc and rear brakes last for ~56 000 and 112 000 vehicle km, respectively (Garg et al., 2000). Some brake wear rates reported in the literature range from 11-29 mg veh^{-1} km^{-1}, depending on vehicle category (Garg et al, 2000) and 20-47 mg veh^{-1} km^{-1}, depending on vehicle category (Legret and Pagotto, 1999) and 8.8 mg veh^{-1} km^{-1} (Luhana et al., 2004). As with tyre wear, Luhana et al. (2004) reported a decreasing pattern of brake wear with increasing mean trip speed because of less braking on highways where the speed limits are higher than in urban areas.

The values reported here show that road wear is ~100 times larger when studded tyres are used as compared to tyre and brake wear, while summer tyres cause road wear in the same size order as tyre and brake wear.

3.2.2 Resuspension of road dust

As indicated in fig. 1, resuspension is the process where particles that have been formed earlier due to wear of the road, tyres, brakes, road surface, etc. deposit onto the road surface, and become resuspended later mainly due to mechanical emission through interaction between the tyres and the surface, and vehicle induced turbulence. The factors influencing resuspension are many and complex. The most important ones are road dust on the road surface and shoulder (road dust depot or dust/silt loading), meteorological factors (precipitation, humidity, temperature, wind speed), traction control methods, amount of traffic, vehicle type, vehicle speed, road cleaning and pavement properties. In addition, the separation between direct emissions due to road, tyre and brake wear and road dust resuspension is a challenging task, and has only been addressed to a limited extent; according to Thorpe et al. (2007), resuspension comprises 20-22% of the total PM_{10} emissions. Lenschow et al. (2001), however, reported that resuspension is the predominant source of road dust PM_{10} near roads. Furthermore, Harrison et al. (2001) reported that approximately half of the roadside PM_{10} levels originated from resuspended road dust, while the other half originated from exhaust particle emissions. Even higher shares are reported by e.g. Forsberg et al. (2005). The main reason for the different results is the use of studded tyres during winter time in some countries; during snowy winters the particles formed due to the enhanced road wear, as well as tyre and brake wear, deposit onto snow piles or wet road surface. When the spring arrives the surface dries up and the large dust depot resuspends, causing high peaks in the PM_{10} concentrations (Omstedt et al, 2005; Ketzel et al., 2007).

The effect of tyre type, road moisture, vehicle category and vehicle speed on resuspension has been adressed in a number of studies; according to Johansson (2007), the resuspension was measured to 16-43% and >87% when using studded and non-studded winter tyres, respectively. With regard to surface moisture, studies have shown that when the surface is wet the road dust emissions are low; Thai et al. (2008) showed that PM_{3-10} and PM_{10} were negatively correlated with precipitation. Furthermore, Nicholson and Branson (1990) showed that resuspension is greatest immediately after the road has dried up. This is supported by Kuhns et al. (2003) who reported that the emission potential was closely related to the period since the last rainfall. With regard to vehicle type, studies have shown that HDV (heavy duty vehicles) contributes significantly to resuspension; larger vehicles create more aerodynamic drag in the wake of the vehicle and thereby cause enhanced traffic induced. Thorpe et al. (2007) reported that $PM_{2.5-10}$ emission factors range from ~171 to ~183 mg km^{-1} for HDV and from ~1.1 to ~5.1 mg km^{-1} for LDV (light duty vehicles) for one set of background data. Abu-Allaban et al. (2003) found that the resuspension emission factors for HDV are 8 times larger than for LDV. Vehicle speed is also an important factor as several studies have shown that the PM emissions increase with vehicle speed. In addition, the rolling tyres squeeze the air beneath the tyres and generate a shear due to tyre rotation. Hussein et al. (2008) showed that the particle mass concentrations behind the studded tyre at a vehicle speed of 100 km h^{-1} were ~10 times higher than that at 20 km h^{-1}. Hagen et al. (2005) reported results from a measurement campaign in Oslo where the speed limit on a major road in Oslo was reduced from 80 to 60 km h^{-1}. As a result the average vehicle speed was reduced with ~12.5%, and the resulting reduction in net concentration of $PM_{2.5-10}$ was 30-35%.

3.3 Efforts to reduce road dust emissions

Several measures have been introduced to reduce road dust emissions. Among these measures are the reduction of the share of studded tyres which has proved to have a large beneficial effect; according to Norman and Johansson (2006), a 10% reduction in the share of vehicles using studded tyres resulted in a reduction of the weekly averaged PM_{10} levels (due to local road wear) of 10 μg m^{-3}. In order to reduce the studded tyre share, taxational measures have been introduced in the larger cities of Norway. In some countries (e.g. Belgium, Japan, Germany, United Kingdom) the use of studded tyres is totally prohibited. Furthermore, several regulations with respect to the use of studded tyres have been introduced since they were first introduced in the early 1960s. The regulations deal with composition of the stud to reduce stud protrusion, stud weight and number of studs per tyre; for example, according to Angerinos et al. (1999), a reduction of the weight of the stud of 1.7-1.9 g to 1.1 g will potentially reduce the the road wear by ~30%.

A number of studies have studied the effects of application of dust-binding material onto the road and road cleaning; Aldrin et al. (2008) used a generalised additive model to assess the effect of salting with magnesium chloride, as well as sweeping and washing of the road on the PM concentration in a road tunnel. The analysis revealed no clear effect of sweeping and washing, while the estimated effect of salting immediately after the salting took place was a 70% reduction compared to the corresponding PM level without salting. With regard to sweeping and washing, similar results were reported by Kuhns et al. (2003) and Norman and Johansson (2006). The latter study also assessed the effect of applying calcium magnesium acetate (CMA) on streets in the city of Stockholm and reported a 35% reduction of the daily PM_{10} under dry conditions. The reported minimal effect of road cleaning may be due to use of less efficient cleaning equipments or that the duration of the measurements are too short to reveal the true effect. It may have an effect on the long term due to the removal of large particles that otherwise would be crushed into smaller resuspendable sizes do to the sandpaper effect.

Many studies have also revealed the effect vehicle speed has on road wear and the PM levels (e.g. Hussein et al., 2008; Etyemezian et al., 2003a; Kupiainen et al., 2005). As a concequence, on some large streets in Oslo, Norway, the speed limit is reduced from 80 km h^{-1} to 60 km h^{-1} during the studded tyre season. Hagen et al. (2005) reported a net reduction in the PM_{10} and $PM_{2.5-10}$ levels of 30-40% due to the reduction in speed limit (the share of studded tyres was ~10% lower during the season with reduced speed limit than the previous season).

3.4 Model concepts and critical summary

This section describes some model concepts that have been developed in order to describe road dust emissions, followed by a critical summary of these models. A common concept in such models is 'emission factors', *EF*, which is usually defined as the mass of PM_x per vehicle kilometre travelled (mg veh km^{-1}) relating the quantity of a released pollutant with the activity that causes the release. A wide range of emission factors for road, tyre and brake wear, as well as for resuspension, are available from literature, e.g. Gehrig et al. (2004), Abu-Allaban et al. (2003), Thorpe et al. (2007), Etyemezian et al. (2003a,b). However, due to differences in ambient meteorological conditions, duration of measurement campaigns, properties of pavements, tyre type, etc, large differences between emission factors for a particular road dust source usually occur.

3.4.1 Road dust emission models

Omstedt et al. (2005) developed a model for vehicle-induced non-tailpipe emissions taking specifically into account the effect of road surface moisture content on the road dust layers. They make use of the fact that the dust load increases due to sanding and road wear, which is strongly dependent on the use of studded tyres, and decreases due to runoff and resuspension:

$$E = N \cdot \left(EF_{direct} + EF_{resuspension}\right) \tag{13}$$

where E (g m^{-1} s^{-1}) is the total emission of particles from the road, N (veh s^{-1}) is the number of vehicles, and EF_{direct} *and* $EF_{resuspension}$ (g veh^{-1} m^{-1}) are the emission factors for direct and resuspended emissions, respectively. Furthermore,

$$EF_{resuspension} = f_q \cdot l \cdot EF_{ref,winter} \tag{14}$$

$$EF_{resuspension} = f_q \cdot EF_{ref,summer} \tag{15}$$

where f_q is a reduction factor related to moisture content, l is the dust load (normalised to the number of sanding days during winter time), depending on runoff, suspension due to road wear and the share of studded tyres, and $EF_{ref,winter}$ and $EF_{ref,summer}$ (g veh^{-1} m^{-1}) are reference emission factors estimated using local measurements of NO_x. $EF_{resuspension}$ is separated into a summer and a winter part because the dust layer is not so important during the summer relative to the winter, especially in Nordic conditions where traction control methods are used during winter.

The EMEP CORINAIR emission model (EMEP CORINAIR, 2003) depends on vehicle category and vehicle speed; in order to calculate PM_{10} emissions from road, tyre and brake wear, the following expressions are used:

$$E_{R,m,k} = N_k \cdot M_k \cdot (EF_R)_k \cdot f_{R,m}$$

$$E_{T,m,k} = N_k \cdot M_k \cdot (EF_T)_k \cdot f_{T,m} \cdot S_T(V) \tag{16}$$

$$E_{B,m,k} = N_k \cdot M_k \cdot (EF_B)_k \cdot f_{B,m} \cdot S_B(V)$$

where the subscripts $i = R$, T, B correspond to road, tyre and brake wear, respectively, and m and k correspond to size class and vehicle class, respectively. $E_{i,m,k}$ (g) is the total PM emissions due to wear source i size class m and vehicle class k, N_k (veh s^{-1}) is the number of vehicles in vehicle class k, M_k (km) is the average mileage driven per vehicle in vehicle class k, $EF_{i,k}$ (g km^{-1}) is the mass emission factor from wear source i and vehicle class k, $f_{i,m}$ is the mass fraction of wear particles from wear source i that can be attributed to size class m and $S_i(V)$ is a wear correction factor for a mean travelling speed V. The expressions for $S_i(V)$ are based on the work of Luhana et al. (2004) who found a decreasing trend of tyre and brake wear for increasing vehicle speed, as mentioned in section 3.2.1. No speed correction are found for road wear due to little information on airborne emission rates from pavement wear. Particle emissions from each of the three sources using relevant emission factors are given in EMEP CORINAIR (2003).

The Norwegian Institute for Air Research (NILU) in Norway has developed a road dust emission model in which the resuspension of $PM_{2.5-10}$ is related to the share of studded tyres, s (%) and heavy traffic, TT (%), traffic speed V (m s^{-1}), vehicle numbers and road wetness, RW (Tønnesen, 2000). The emission of PM_{10}, $E(PM_{10})$ (g veh^{-1} km^{-1}), is expressed as

$$E(PM_{10}) = E(PM_{2.5}) + E(PM_{10-2.5}) \tag{17}$$

where

$$E(PM_{10-2.5}) = E_{ref}(PM_{2.5}) \cdot (a \cdot TT + b) \cdot \left(\frac{V}{V_{ref}} \right)^2 \cdot RP \cdot RW \tag{18}$$

$$E(PM_{2.5}) = E(EP) \cdot (1 + 0.69 \cdot RP \cdot RW), \tag{19}$$

where $E_{ref}(PM_{2.5})$ (g veh^{-1} km^{-1}) is the emission of $PM_{2.5}$ in the reference situation, *a* and *b* are empirical resuspension values specific for each site, V_{ref} (m s^{-1}) is a reference speed related to the speed limit at the site, *RP* is a factor related to the use of studded tyres and *E(EP)* (g veh^{-1} km^{-1}) is emission of exhaust particles. *RP* is expressed as

$$RP = 0.98 \cdot s + 0.02 \tag{20}$$

where *s* (%) is the share of studded tyres.

The USEPA AP-42 model (US EPA, 2006) calculates emissions from brake wear, tyre wear and "fugitive dust" emissions from paved and unpaved roads:

$$E_{daily} = k \cdot \left(\frac{l_{75}}{2}\right)^{0.65} \cdot \left(\frac{m}{3}\right)^{1.5} \cdot \left(1 - \frac{P}{4 \cdot N}\right) \tag{21}$$

$$E_{hourly} = k \cdot \left(\frac{l_{75}}{2}\right)^{0.65} \cdot \left(\frac{m}{3}\right)^{1.5} \cdot \left(1 - \frac{1.2 \cdot P}{N}\right) \tag{22}$$

where E_{daily} and E_{hourly} (g veh^{-1} km^{-1}) are the daily and hourly emission factor for particles, respectively, *k* (g veh^{-1} km^{-1}) is a base emission factor dependent on particle size, l_{75} (g m^{-2}) is the dust load less than or equal to 75 µm, *m* (tonnes), is the average weight of the vehicles and *P* is the number of days with at least 0.254 mm precipitation. The silt loading is determined by vacuuming the travelled portion of the paved road. The model was developed using a larger dataset consisting of about 60 data points for a variety of roads and the term in the parenthesis was then determined by regression analysis of the data. A modified version of this model is in use in Germany, where the modifications involved separation of exhaust and road dust contributions and adjustments to the constant values (Rauterberg-Wulff, 2000, Gamez et al., 2001). However, this model overpredicted the emissions at highways, and the whole concept of AP-42 was abandoned in favour of providing emission factors as functions of "traffic situations", e.g. motorway, city main roads, etc. (Ketzel et al., 2007). A more detailed description of USEPA AP-42 can be found in Venkatram (2000) and Boulter (2005).

3.4.2 Critical summary of road dust emission models

In general, the majority of the road dust emission models described above apply local air quality measurements or empirical constants specific to the one or very few road environments; e.g. the *a* and *b* values in the model of Tønnesen (2000) (eq. 18), specific for roads in Oslo, Norway, and the factor *k* in the AP-42 model (eqs. 21 and 22). In particular, in the model of Omstedt et al. (2005), which is developed to handle the seasonal variations in PM typical for countries using studded tyres during winter time, the reference emissions factors for PM, $EF_{ref,winter}$ and $EF_{ref,summer}$, are determined by using the tracer method with NO_x as tracer; hence, the model becomes tuned towards a specific site, although the other terms are more general. The tracer method assumes that the ratio between the net observed concentration of NO_x, $C_{NOx,net}$, and the emission factor of NO_x, EF_{NOx}, (both considered to be relatively certain) is equal to the ratio between the net observed concentration of PM, $C_{PMx,net}$, and the emission factor of PM, EF_{ref}:

$$\frac{C_{PM_{10},net}}{EF_{ref}} = \frac{C_{NO_x,net}}{EF_{NO_x}}. \tag{23}$$

The EMEP CORINAIR, on the other hand, does not take into account resuspension and hence, cannot take into account seasonal variation and reproduce the spring time peaks in PM_{10} in countries where traction control methods are used. Furthermore, no speed correction is included for road wear, although many studies reveal the importance of vehicle speed on road wear (Gustafsson et al., 2008a, Kupiainen et al., 2005). In addition, more detailed information is required with regard to the relative effects of different tyre and road surface combinations, road wear emission factors and resuspension (EMEP CORINAIR, 2003). The model of Tønnesen (2000) is critizised for not taking into consideration different pavement types, and for relating the resuspension to modelled emissions of exhaust particles, the latter being emphasized by Bringfelt et al. (1997), who stated the emission factor for resuspended particles increases with increasing vehicle speed, while this is not the case for exhaust particles. In addition, the model does not include any explicit description of resuspension. The AP-42 model has been critizised for using silt loading (*l*) as input, which can be quite ambiguous. Venkatram (2000) concluded that the estimates of PM_{10} emissons from paved roads were not reliable.

Model inter-comparison studies are important to develop road dust emission models. Ketzel et al. (2007) compared the model of Omstedt et al. (2005) with several other models, among them being the tracer method (eq. 23). The models were applied onto datasets from streets in Stockholm, Sweden, and Copenhagen, Denmark. It is interesting to note that when applied upon the Danish measurements, the model of Omstedt et al. (2005) did not perform better than the tracer method. This was mainly because there is less seasonal variation in

Copenhagen in terms of PM levels, as studded tyres are not allowed. Other than indicating that the emissions are dependent on external factors not described by the model of Omstedt et al. (2005), this also illustrates the general importance of comparing models and applying then upon several other datasets other than the ones upon which they are developed.

In order to aid the development of more general model frameworks with process descriptions of the parameters involved, detailed measurement campaigns should be conducted in the future in order to better understand the mechanisms controlling direct emissions and resuspension. The campaigns should include measurements of surface conditions such as surface temperature, wetness, freeze and drainage, as well as measurements of road wear and dust depot on the road surface and shoulder with analysis of source apportionment and size distributions.

4 Research summary and main conclusions

Road traffic is a highly important source of a range of pollutants having adverse health and environmental effects. Hence, efforts to reduce traffic related emissions are beneficial for both health and the environment in the long term. Development of models describing traffic related emissions and dispersion of pollutants will in the end lead to robust models that well reproduce available air quality measurements. Furthermore, such models will aid the analysis of existing mitigation measures, as well as developing new ones, to reduce the emissions of traffic related pollutants. With this background, the main objective in this thesis is to evaluate and develop models related to the emissions and dispersion of traffic induced road dust. This thesis emphasizes the importance of using models as tools for increasing our general understanding of traffic related emissions and dispersion.

The first study deals with the modelling of the dispersion and resulting concentration levels of NO_x at distances up to 100 m from major roads in rural areas, using four Gaussian open road line source models developed in Norway, Denmark and Finland. These models were applied to datasets from measurement campaigns conducted in rural areas near major roads in the mentioned countries. NO_x was considered since it was measured at all sites, its emissions are well known and since it can be treated as a tracer for the short time scales involved. When comparing the models with the measurements we found that the results are sensitive to the parameterisation of traffic produced turbulence (TPT), especially at distances close to the road in combination with low wind speeds. The Danish model OML-Highway performed best at all sites due to its parameterisation of TPT based on turbulent kinetic energy. Future models should also be aware of their description of effective transport velocity, as well as Lagrangian time scales.

The second study is concerned with the development of a more generalised model framework to describe road dust emissions, i.e. emissions of particulate matter (PM) from the road surface due to road surface wear as well as resuspension of deposited material on the road surface and road shoulders. There is a need for a more generalised road dust emission model since the majority of already developed road dust emission models contain empirical constants or functions related to local air quality measurements. As such, the applicability of these models on other road environments is limited and they cannot be used for analysis of mitigation measures related to road dust emissions. The model concept described in this study accounts for the main processes controlling road dust emissions and does not depend on local measurements; it is based on measurements of road, tyre and brake wear to obtain the relevant emission factors. A mass balance concept is used for describing the variation in dust load on the road surface and shoulders. Furthermore, the model separates the direct emissions and resuspension and treats the road surface and shoulder as two individual sources. When applying the model onto two datasets from measurement campaigns conducted at major roads in and outside Oslo, Norway, during the studded tyre season, it performed well during warm periods and less well for temperatures close to or below 0 ºC in combination with precipitation. In particular, it overestimated the PM_{10} concentrations under heavy precipitation events, since it does not take amount of precipitation into account, and underestimated the PM_{10} levels during periods in which salting occurred; the model does not include salting as an additional mass source. As such, refinements of the parameterisations of road surface conditions are needed and measurement campaigns with the aim of understanding the effect of road surface conditions on road dust emissions should be conducted. In spite of the current limitations, the model provides a well described conceptual framework and describes processes that no model has ever done before. The model will in the future provide the potential for good air quality planning.

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Summary of papers

Paper 1: Berger, J., Walker, S.E., Denby, B., Berkowicz, R., Løfstrøm, P., Ketzel, M., Härkönen, J., Nikmo, J., Karppinen, A., 2010. Evaluation and inter-comparison of open road line source models currently in use in the Nordic countries. Boreal Environmental Research 15: 00-00 (preprint). Will be published 2010.

In this paper, a modified version of the US EPA model HIWAY2, the Finnish CAR-FMI model, the Danish OML-Highway model and the Norwegian WORM model are inter-compared and evaluated. All these models are Gaussian open road line source models. The overall aim is to determine under which conditions the models perform well or poorly. The models are applied onto three datasets from measurement campaigns conducted at open road environments in Oslo, Norway, Helsinki, Finland, and Copenhagen, Denmark. We assess the results with regard to normalisation, wind speed, wind direction, horizontal profiles and atmospheric stability. We use NO_x in the model simulations, since its emissions are well known and it can be treated as a tracer for the short time scales involved. In general, a decrease of the correlation between model estimates and observations is evident when normalising the data with NO_x emissions, due to the significant positive correlation between observed concentrations and emissions. Furthermore, a reduction of bias is evident when normalising the Norwegian and Danish data, due to overestimation of the dispersion at lower emission values. In general, OML-Highway performs best, particularly for higher emission values when the influences of traffic density and vehicle speed on traffic produced turbulence are higher. This is due to a more advanced parameterisation of traffic produced turbulence based on turbulent kinetic energy. With regard to horizontal profiles, the relative bias for CAR-FMI increases with increasing distance from the road, indicating that the Lagrangian time scales are too short. In the future, OML-Highway's parameterisation of traffic produced turbulence should be implemented in open road line source models. In addition, it is important that the effective transport velocity and Lagrangian time scales are well described.

Paper 2: Berger, J., Denby, B., 2010. A generalised model for traffic induced road dust emissions. Part 1: concept and model description. Submitted to Atmospheric Environment.

This is the first paper in a two part series of studies concerning the development and evaluation of a generalised road dust emission model. Most of today's road dust emission models are based on local measurements and/or contain empirical emission factors that are specific for a given road environment. Hence, they are less applicable to other road environments with different meteorological conditions and traffic characteristics. The aim with this study is to develop a more generalised model using process descriptions of the most important parameters involved, preferably without any tuning towards the dataset upon which it has been developed. The model framework presented in this paper uses road, tyre and brake wear rates to determine the emission factors for direct road dust emissions. The mass balance concept is used to estimate the dust load on the road surface and road shoulder. Furthermore, the model separates the road dust emissions into a direct part and a resuspension part, and treats the road surface and road shoulder as two different sources. In this paper we discuss the parameterisations that are needed to complete the model and a review of results reported in the literature is given. We analyse the model under highly idealized conditions to see the connection between the different parts of the model (i.e. the production of dust load on the road surface and shoulder, the functions controlling the amount of dust particles becoming directly emitted or resuspended, as well as the dust load on the road surface and shoulder and their time scales).

It is highly challenging to obtain full information on all parameters that may affect road dust emissions. Nevertheless, estimates of the time scales for the build up of road dust on the road surface and road shoulder have been obtained; the former is less than one hour under the majority of traffic conditions while the latter ranges from weeks to months. The model also manages to reproduce the observed increase in road dust emissions directly after drying due to build up of mass on the road surface and shoulder during precipitation events. The model enables a broader conceptual view of the phenomenon of road dust and other non-exhaust emissions and provides a framework on which to base further studies.

Paper 3: Berger, J., Denby, B., 2010. A generalised model for traffic induced road dust emissions. Part 2: model evaluation. Submitted to Atmospheric Environment.

This is the second paper in a two-part series of studies concerning the development and evaluation of a generalised road dust emission model. In this paper the model is compared with PM_{10} measurements from two major roads; Aker Hospital in Oslo, Norway, and Nordbysletta, 20 km outside Oslo. Both measurement campaigns were conducted during the studded tyre season. Analysis of the results showed that the model gave excellent results under warm conditions, and needs refinement of the road surface condition parameterisations under other conditions. In particular, the model underestimated the net PM_{10} levels for temperatures around 0 ºC mainly due to the observed salt contribution; the model currently does not include salting as an additional mass source. In general, the model outputs as well as the PM_{10} concentrations are highly sensitive upon the effect of precipitation. Since the model does not take the amount of precipitation into account, it overestimates the observations under relatively high precipitation events.

In spite of the current limitations of the model, the model reflects a number of processes that no other model has ever done before, e.g. with regard to the particle redistribution rates (*f*- and *g*-functions), the treatment the road shoulder as an individual source and giving as output the time scales for build up of road dust on the road surface and shoulders. We have indeed developed a generalised road dust emission model that does not rely on local measurements; the basic emission factors are adjusted to road wear information that can be more easily obtained than air quality data from road network measurements. Overall, the model presents a generalised conceptual framework for further development and can lay the course for future measurement campaigns. As more monitoring data is obtained and specific parameterisations are refined, the model will in the future enable improved estimates of road dust emissions and provide the potential for good air quality planning and management.

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Evaluation and inter-comparison of open road line source models currently in use in the Nordic countries

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The aim of this study was to compare and evaluate operational open-road Gaussian line-source models currently in use in Norway, Denmark and Finland. Four models, HIWAY2-AQ, OML-Highway, CAR-FMI and WORM, were applied to datasets from three measurement campaigns from each of the mentioned countries. The results were assessed through analysis with regard to normalisation, wind speed, wind direction, horizontal profiles and stability. Generally, the correlation between model estimates and observations decreased when normalising with emissions, due to the significant positive correlation between observed concentrations and emissions. Furthermore, we found a reduction of bias when normalising the Norwegian and Danish data, caused by overestimation of the dispersion at lower emission values. Due to OML-Highway's more advanced parameterisation of traffic-produced turbulence, this model performed best at higher emission values when the influence of traffic density and vehicle speed on traffic produced turbulence was higher. With regard to horizontal profiles, the relative bias for CAR-FMI increased as a function of distance from the road, indicating that the Lagrangian time scales are too short.

Introduction

Model comparison studies provide a robust basis for evaluation, development and improvement of models. When several models are applied to the same dataset, we obtain insight and knowledge on specific differences between the models, and on the parts of the models that perform well or poorly. Over the years, a number of operational open-road or highway dispersion models have been developed, e.g. the HIWAY models (1 to 4), the CALINE models (1 to 4), GM and ROADWAY. A review of these models can be found in Sharma *et al*. (2004). Gaussian models, however, typically perform poorly under low wind-speed conditions, or when the wind direction is close to parallel to the road, as described in Benson (1992). In Oettl *et al*. (2001), the Gaussian line-source model CAR-FMI was compared with the Lagrangian dispersion model GRAL, with special emphasis on low wind-speed conditions. It was shown that CAR-FMI tended to overestimate the NO_x concentrations, while GRAL underestimated them, which was mainly due to GRAL's special treatment of enhanced horizontal dispersion (meandering flows) under low winds. Similar comparison studies were performed by e.g. Levitin *et al*. (2005) where CAR-FMI and the Gaussian line-source model CALINE-4 were applied to a Finnish dataset. The results showed that both models performed better at a greater distance from the road, and poorest performance was seen for low winds and parallel wind directions.

In this study, we applied a modified version of the HIWAY-2 model called HIWAY2-AQ, the Danish OML-Highway model, the Finnish CAR-FMI model and the new Norwegian WORM model, to three datasets from Norway, Denmark and Finland, respectively. The study addressed only roadside environments at rural sites that were not influenced by any building obstacle; hence, we denote these environments rural and open. The comparison was aimed at analysing the variability and quality of these open-road line-source (ORLS) models. More specific aims were to (i) determine the traffic-related and meteorological conditions, for which further model development is needed, and (ii) to evaluate each model performance against datasets.

Model descriptions

Four models were applied in this study. Three of these are operational at the institutions involved, while the WORM model is still under development at the Norwegian Institute of Air Research (NILU). Each model calculates concentrations at various receptor points by integrating concentrations from a set of infinitesimal point sources defined along each line-source using the Gaussian plume equation as a basis (Seinfeld and Pandis 1998):

$$C = \frac{Q}{u_h}\int_0^D f d\ell, \tag{1}$$

where C (g m^{-3}) is the concentration at the receptor point, Q (g m^{-1} s^{-1}) is the line source emission strength (assumed constant along the line source), u_h (m s^{-1}) is the effective transport velocity, D (m) is the length of the line source, f (m^{-2}) is the plume dispersion function and ℓ (m) is an arbitrary line. The function f is given as:

$$f = \frac{1}{2\pi\sigma_y\sigma_z}\exp\left(-\frac{y^2}{2\sigma_y^2}\right) \times\left[\exp\left(-\frac{(z-h)^2}{2\sigma_z^2}\right)+\exp\left(-\frac{(z+h)^2}{2\sigma_z^2}\right)\right], \tag{2}$$

where σ_y and σ_z (m) are the Gaussian horizontal and vertical dispersion parameters, respectively, y and z (m) are coordinates and h (m) is the effective source height. The above formulation does not include internal reflections from the top of the boundary layer. Extra terms are included in some of the models to account for this. The effective transport velocity, u_h, transports the pollutants away from the source. Since the observed wind speeds in each dataset are measured at a higher level than h, u_h must be calculated. Although each model uses different methods in order to calculate u_h, they all apply Monin-Obukhov similarity theory to extrapolate the measured wind speed down to the transport height:

$$u_h = u(z_1)\frac{\ln\left(\frac{h}{z_0}\right)-\psi_m\left(\frac{h}{L}\right)+\psi_m\left(\frac{z_0}{L}\right)}{\ln\left(\frac{z_1}{z_0}\right)-\psi_m\left(\frac{z_1}{L}\right)+\psi_m\left(\frac{z_0}{L}\right)}, \tag{3}$$

where $u(z_1)$ (m s^{-1}) is the measured wind speed at the measurement height $z_1 > h$, z_0 and L (m) are the surface roughness and Monin-Obukhov lengths, respectively, and ψ_m is the stability correction function (Businger *et al*. 1971). The explicit forms of this function can be found e.g. in Paulson (1970).

A common assumption in all the models is that the total Gaussian dispersion parameter, $\sigma_{y,z}$, can be represented as a combination of atmospheric turbulence, σ_{y_a,z_a}, and traffic-produced turbulence (TPT), σ_{y_0,z_0}, as

$$\sigma_{y,z}^2 = \sigma_{y_a,z_a}^2 + \sigma_{y_0,z_0}^2. \tag{4}$$

All models except form OML-Highway base their formulation of TPT on the formulation in the HIWAY-2 model (Petersen 1980), which is a semi-empirical treatment based on the General Motors experiments (Cadle *et al*. 1976):

$$\sigma_{z,\mathrm{initial}} = 3.57 - 0.53U_{\mathrm{c}}, \quad (5)$$

$$\sigma_{y,\mathrm{initial}} = 2\sigma_{z,\mathrm{initial}}. \quad (6)$$

Equations 5 and 6 represent the initial vertical and horizontal dispersions in the near vicinity of the source caused by turbulent mixing due to car wakes. The smallest allowable value of $\sigma_{z,\mathrm{initial}}$ is 1.5 m. The model includes a factor U_c called the aerodynamic drag, which accounts for the initial dilution of the pollutant on the roadway, and allows the model to make reasonable concentration estimates during low wind-speed conditions. An analysis of the General Motors data showed that U_c, must depend on u_h and the horizontal angle of the wind to the roadway, θ (deg):

$$U_c = 1.85u_h^{0.164} \cos^2 \theta. \quad (7)$$

Note that Eqs. 5 and 7 are numerical equations, and the units do not fit. For further details, *see* Petersen (1980).

The HIWAY2-AQ model (NILU version of HIWAY-2)

The HIWAY2-AQ model is a modified version of the US EPA HIWAY-2 model (Petersen 1980), and is currently used as the operational sub-grid scale line source model in NILU's Air Quality Information and Management System (AirQUIS). It is a steady-state Gaussian model in which it is assumed that each lane of the highway is a continuous, finite line-source with a uniform emission rate.

In order to calculate downwind concentrations, three conditions are considered with regard to stability: in stable conditions, or if the mixing height $h_{\mathrm{mix}} > 5000$ m, Eq. 1 is used. In neutral or unstable conditions, if $\sigma_z > 1.6h_{\mathrm{mix}}$, the distribution below the mixing height is uniform with height regardless of the source and receptor height, provided both are smaller than the mixing height. For all other unstable or neutral conditions multiple reflections from the mixing height and surface are also considered up to N reflections. However, due to the low level of the traffic sources and the close proximity of the measurements to the road, the mixing height did not play a significant role in the model calculations carried out it this study.

Integration method

The trapezoidal rule together with the Richardson extrapolation is used to evaluate Eq. 1. This is based on the concept that a weighted average of two different estimates of the same value can be more accurate than either of the estimates, provided the weights are chosen appropriately to cancel the errors. The integration is iteratively solved for 9 iterations or until a predefined accuracy of 2% is achieved.

Parameterisation of turbulence parameters

The HIWAY-2 model calculates the dispersion parameters, σ_y and σ_z using Pasquill-Gifford curves and stability classes (Turner 1969). However, in HIWAY2-AQ, only two stability conditions are used; classes A, B, C, representing unstable and D, representing neutral conditions, are all treated as neutral, i.e. class D, whilst classes E and F are both treated as lightly stable, i.e. class E. In Eq. 4, σ_{z_a} is of the form:

$$\sigma_{z_a} = ax^b, \quad (8)$$

where x (km) is the downwind distance from the source, and a and b are empirical factors depending on the atmospheric stability. On the other hand, σ_{y_a} is dependent on x and on the half angle of the horizontal spreading of the plume to the road, θ_{p} (deg):

$$\sigma_{y_a} = \left(\frac{1000}{2.15}\right) x \tan \theta_{\mathrm{p}}, \quad (9)$$

The value 2.15 is the number of standard deviations from the centreline of the Gaussian distribution to the point where the distribution falls to 10% of the centreline value.

TPT is modelled according to Eqs. 5–7.

The OML-Highway model

The National Environmental Research Institute (NERI) in Denmark has developed the OML-Highway model. It is a local-scale Gaussian air pollution model based on boundary layer scaling, which estimates dispersion from point sources and area sources. It comprises two versions; OML-Point 2.1, which is applicable to a single source, and OML-Multi 5.0, which allows multiple point and area sources. These underwent a revision in 2005–2006, but in this study we have used the Multi 5.0 version before the revision. OML-Highway has a meteorological pre-processor, which applies Monin-Obukhov similarity theory using synoptic, sonic and radiosonde data to calculate the required turbulent parameters.

Integration method

OML-Highway treats the traffic lanes as area sources. It calculates the concentration at a receptor point by a double integral in the crosswind and the along-road wind directions. When the receptor is inside an area source, OML-Highway only integrates the upwind part of the area source. For crosswind directions it treats the road as a finite line source and applies the analytic solution of Eq. 1 with error functions similar to Eq. 11. However, for along-road wind directions, OML-Highway uses the Romberg integration technique (Press *et al*. 1992) with Richardson's extrapolation of the trapezoidal rule. At greater distances from the road, the numerical integration is replaced by a single line source for faster calculations. A more detailed description of the integration procedure can be found in Olesen *et al*. (2007).

Calculation of effective dilution velocity, u_h

In order to estimate u_h for stable conditions, OML-Highway calculates an average wind speed between the ground and the emission height by integrating the wind profile (Eq. 3) between these heights. The minimum height at which the wind speed is calculated is $z_{\min} = \max\{z, 10z_0\}$ (m). For unstable conditions, OML-Highway does not use average wind speeds but applies Eq. 3 to estimate the $u_{h,\text{unstable}}$ at the emission height, h. In all conditions, a minimum wind speed given by $u_{\min} = \max\{0.2, 0.6w^*\}$ m s^{-1} is used, where w^* is the convective velocity scale ($w^* = 0$ m s^{-1} for neutral and stable conditions).

Parameterisation of turbulence parameters

The parameterisation of σ_0 is based on the formulation in the OSPM model (Berkowicz 2000), but is slightly modified with regard to highways in open environments, where traffic produced turbulent kinetic energy, e (m^2 s^{-2}), is represented as a function of the number, the size and the speed of the vehicles. The assumption in OSPM of a constant TPT is not applicable to an open highway, as the concentrations are calculated at a greater distance from the road. It is therefore assumed that the velocity parameter, given as $u_{\text{TPT}} = \sqrt{e}$ (m s^{-1}) decays in an exponential manner with respect to distance from the source:

$$\sigma_0(t) = \sigma_{\text{initial}} + u_{\text{TPT}}\tau\left[1-\exp\left(-\frac{t}{\tau}\right)\right], \quad (10)$$

where t is the transport time (s), τ is the time scale for the decay of TPT (s) and $\sigma_{\text{initial}} = 3.2$ m is the initial dispersion. These parameters have been determined empirically based on analysis of the Danish data used in this study.

The CAR-FMI model

The CAR-FMI model (Contaminants in the Air from a Road) has been developed by the Finnish Meteorological Institute (FMI). It consists of an emission model, a dispersion model and a statistical analysis of the computed time-series of the concentrations. A more complete description of the model is presented in e.g. Härkönen *et al*. (1996). In this particular study, we applied the version that was used in the OSCAR (**O**ptimised Expert **S**ystem for **C**onducting Environmental **A**ssessment of Urban **R**oad Traffic) project. The meteorological pre-processor, MPP-FMI, is based on the method developed by van Ulden and Holtslag (1985). This method evaluates the turbulent heat and momentum fluxes in the atmospheric boundary layer (ABL) from observations. The parameterisation of the ABL height is based on the boundary layer scaling and meteorological sounding data (Karppinen *et al*. 1998), yielding hourly values of turbulence parameters, such as the Monin-Obukhov length scale, friction velocity and convective velocity scale, and boundary layer height.

Integration method

The pollutant concentration is estimated from the analytical solution of Eq. 1, which is integrated over a finite line source in the lateral direction (Luhar and Patil 1989). Furthermore, the model assumes a total reflection from the ground, ignores reflection at the mixing height (h_{mix}), and allows any wind direction with respect to the line source:

$$C = \frac{Q}{2\sqrt{2\pi}\sigma_z\left(u\sin\theta + u_0\right)} \times\left[\exp\left(-\frac{(z-h)^2}{2\sigma_z^2}\right)+\exp\left(-\frac{(z+h)^2}{2\sigma_z^2}\right)\right] \times\left[\operatorname{erf}\left(\frac{\sin\theta(p-y)-x\cos\theta}{\sqrt{2}\sigma_y}\right)+\operatorname{erf}\left(\frac{\sin\theta(p+y)+x\cos\theta}{\sqrt{2}\sigma_y}\right)\right], \quad (11)$$

where u (m s^{-1}) is the average wind speed, u_0 (m s^{-1}) is a wind speed correction due to the turbulence induced by the traffic, x, y and z (m) are the coordinates where the origin is at the centre of the line source, p (m) is the half length of the line source, erf is the error function and the other variables are as explained earlier. The numerical value of u_0 = 0.2 m s^{-1} effectively removes the singularity when the wind direction is parallel to the road.

Parameterisation of turbulence parameters

In MPP-FMI, the turbulence parameters are modelled as a function of the Monin-Obukhov length, friction velocity and mixing height. With regard to the atmospheric turbulence, the parameters are written in terms of turbulent intensities, $i_{y,z}$ (Gryning *et al*. 1987):

$$\sigma^2_{ya,za} = i_{y,z} f_{y,z} x, \quad i_{y,z} = \frac{\sigma_{v,w}}{u(z)}, \quad f_{y,z} = \left[1+\sqrt{\frac{t}{2T_{\mathrm{L}y,z}}}\right]^{-1}, \quad (12)$$

where $f_{y,z}$ are functions of x, $\sigma_{v,w}$ (m s^{-1}) are the standard deviations of the turbulence velocity fluctuations in the lateral and vertical directions, $T_{\mathrm{L}y,z}$ (s) are the Lagrangian time scales and the other variables are as explained earlier. σ_v and σ_w are parameterised according to Gryning *et al*. (1987) and can be found in Härkönen *et al*. (1996).

With regard to TPT, the parameterisation is based on the same formulation as in the HIWAY-2 model (Eqs. 5–7).

The WORM model

The WORM (Weak Wind Open Road Model) is currently under development at NILU (S. E. Walker unpubl. data). The version of the model applied in this study is quite similar to the CAR-FMI model, with some modifications regarding the integration technique and parameterisations used. The meteorological pre-processor is based on Monin-Obukhov similarity theory and equations for h_{mix} are applied as recommended by COST-710 (Fisher *et al*. 1998). The horizontal and vertical Lagrangian time scales ($T_{\mathrm{L}y,z}$) were set to 300 s in this study.

Integration method

WORM integrates the plume dispersion function over the line source using a highly accurate and efficient Gaussian quadrature method (Kythe and Schäferkotter 2005), which is accurate also for wind directions parallel to the road.

Parameterisation of turbulence parameters

The horizontal and vertical turbulence profile used in this study are based on Gryning *et al*. (1987) with a minimum setting of σ_v, $\sigma_{v,\min}$ = 0.5 m s^{-1}, to reduce overestimated concentrations at low wind speeds. With regard to σ_y, WORM takes into account horizontal meandering for low wind speeds using an expression given in Oettl *et al*. (2005). The growth of σ_z currently uses the same formulation as in the CAR-FMI model (Eq. 12).

Regarding TPT, WORM currently uses the same formulation as the HIWAY-2 model (Eqs. 5–7). Table 1 summarizes the major features and differences between the models.

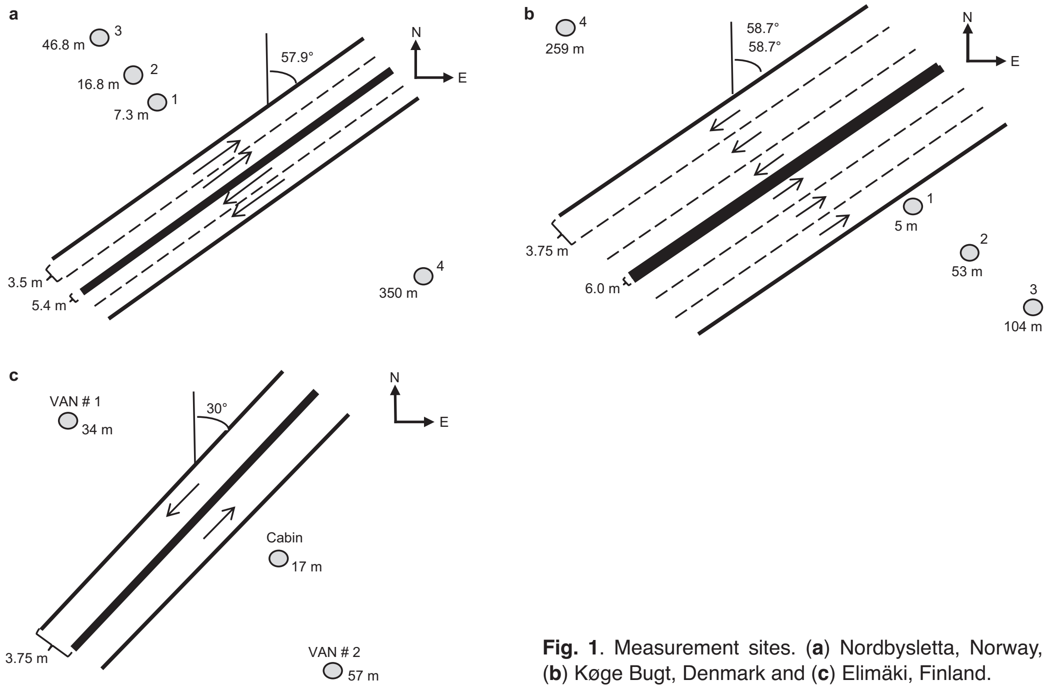

Fig. 1. Measurement sites. (**a**) Nordbysletta, Norway, (**b**) Køge Bugt, Denmark and (**c**) Elimäki, Finland.

Measurement sites, datasets and emissions

The datasets consisted of air quality and meteorological measurements carried out near major roads/highways. Most of the campaigns included a number of stations placed at different distances from the road (Fig. 1, *see* also Table 2). The pollutant NO_x ($NO + NO_2$) was considered, since this compound was measured at all sites, its emissions were best known and it could be treated as a tracer for the short time scales involved.

Emissions

Emission inventories are calculated for each of the sites by the institutes responsible for the monitoring campaigns. However, in one case, Nordbysletta, the emissions were recalculated by the Danish institute, using the emission module WinOSPM.

The Norwegian emission dataset is calculated using the AirQUIS emission module. For line sources, the module needs information such as traffic volume, speed, road characteristics (orientation, road width, etc.) and classification of vehicles. The emission factors are dependent on factors such as fuel, driving speed, slope and ageing factor (Denby *et al*. 2004) and are based on COPERT III (Ekström *et al*. 2004). According to this methodology, the NO_x emissions are set to zero when the traffic speed exceeds 130 km h^{-1}. In AirQUIS, however, this threshold is set to 110 km h^{-1}.

For the Danish data, the emissions are estimated based on traffic data from the Danish Public Roads Administration. These include number of vehicles and vehicle speed, with a classification into light and heavy duty vehicles. Based on these data the emission module in the Danish street pollution model OSPM, WinOSPM, calculates a time series of emissions (Jensen *et al*. 2004).

The emission model in CAR-FMI, which calculates vehicular CO and NO_x, is based on national emission factors of the traffic planning system KEHAR 2.0, developed by the Finnish Road Administration. Speed limit, type of road,

Table 1. Main features and differences between models.

	Norwegian Institute for Air Research	National Environmental Research Institute	Finnish Meteorological Institute	Norwegian Institute for Air Research
Model	HIWAY2-AQ	OML-Highway	CAR-FMI	WORM
Model type	Slender plume Gaussian steady state	Slender plume Gaussian steady state	Slender plume Gaussian steady state	Slender plume Gaussian steady state
Met. data	Single height wind speed, direction, mixing layer. Pasquill-Gifford stability class	Different options, either sonic data or radiosonde	MPP-FMI boundary layer scaling	Single height wind and temperature profiles
Calculation of u_h	Monin-Obukhov similarity theory ($\psi_m = 0$, $h = 2$ m)	Monin-Obukhov similarity theory (different for stable and unstable conditions)	Monin-Obukhov similarity theory ($h = 2$ m)	Monin-Obukhov similarity theory ($h = 2$ m)
u_{min} (m s^{-1})	–	max{0.2, 0.6 w^*}	0.2	0.5
Stability described using	Pasquill-Gifford stability classes	Monin-Obukhov similarity theory	Monin-Obukhov similarity theory	Monin-Obukhov similarity theory
h_{mix}	Boundary layer parameter based on u^*	Pre-processor or from measurements	MPP-FMI	COST-710 eq.
T_L	–	Implicit, dependent on met. conditions	Unstable, $L < 0$: $T_L = 300$ sec, stable, $L > 0$: $T_L = 30$ sec	$T_L = 300$ sec
Integration method	Numerical, Richardson extrapolation	Analytical for crosswind direction, numerical for along wind direction	Analytical (Luhar and Patil 1989)	Numerical, Gaussian quadrature
TPT	Semi-empirical (Petersen 1980)	Empirical, exponential decay of TPT with distance from source	Semi-empirical (Petersen 1980)	Semi-empirical (Petersen 1980)

traffic flow, the share of heavy duty vehicles and the year for the computations is used as input.

Methodology

In this study, the four models were applied to the three datasets and analysed statistically. The overall model performance on all data were assessed using the Pearson coefficient of determination (denoted by R^2) and relative bias (denoted by RB), where

$$R^2 = \frac{\left[n\left(\sum C_{\text{pred}}C_{\text{obs}}\right) - \sum C_{\text{pred}}\sum C_{\text{obs}}\right]^2}{\left\{n\left(\sum C_{\text{pred}}^2\right) - \left(\sum C_{\text{pred}}\right)^2\left[n\left(\sum C_{\text{obs}}^2\right) - \left(\sum C_{\text{obs}}\right)^2\right]\right\}}, \quad (13)$$

where n is the number of data points, C_{pred} is the predicted concentration (g m^{-3}) and C_{obs} is the observed concentrations (g m^{-3}), and

$$\text{RB} = (C_{\text{pred}} - C_{\text{obs}})/C_{\text{obs}}. \quad (14)$$

Handling and selection of the datasets

Since in this study we were interested in the comparison of the models we concentrated largely on the dispersion parts of the models. Several methodologies were at our disposal to do this:

— Model performance were assessed after dividing both observed and model-calculated concentrations by emissions Q (Q-normalisation).
— The ratios of modelled/observed concentrations for different meteorological conditions were compared.
— Model performance as a function of a distance from the road were assessed using Q-normalised concentrations.
— Model performance with regard to atmospheric stability were assessed using the Q-normalised concentrations.

Note that in some cases, we present results only from stations 3 and 2 from the Norwegian and Danish dataset, respectively, as these stations were located at similar distances from the road (Fig. 1). With regard to the Finnish site, the station VAN #1 was the only station where the air quality measurement height was 3.5 m, i.e. a similar measurement height as for the stations at the Norwegian and Danish sites (*see* Table 2); hence, this station was used in the analysis. Furthermore, due to time constraints OML-Highway was not

Table 2. Site and measurement characteristics.

Site	Nordbysletta, Norway	Køge Bugt, Denmark	Elimäki, Finland
Time of campaign	1 Jan.–15 Apr. 2002	17 Sep.–18 Dec. 2003	15 Sep–30 Oct. 1995
Number of datapoints	~900	~900	~75
Number of stations (background excluded)	3	3	1
Length of road segment (m)	~850	1485	~2000
Number of lanes	4	6	2
Orientation of road (due north)	57.9°	58.7°	30°
Traffic flow (veh. day^{-1})	~36000	~100000	~7200
Speed limit (km h^{-1})	90	110	100
Meteorological measurements	Cup anemometer: hourly wind speed, wind direction, temperature, vertical temperature difference between 10 m and 2 m, global solar radiation	Ultrasonic anemometer: hourly wind speed, temperature, etc. Rural background site: hourly global solar radiation data	Hourly wind speed, temperature, wind direction, global solar radiation, relative humidity
Height of meteorological measurements (m)	10	8	3.5, 6 and 10
Height of air quality measurements (m)	3.5	3	3.5
Roughness length (m)	0.25	~0.1	0.2
Average wind speed (m s^{-1})	~2.3	~4.5	~3

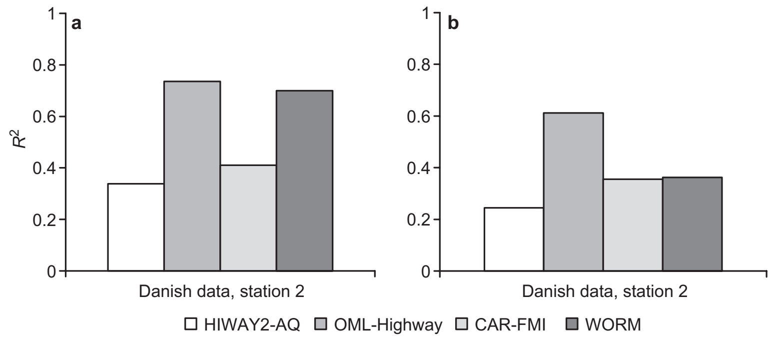

Fig. 2. Coefficient of determination (R^2) for all models applied to the Danish data (station 2): (**a**) non-normalised results, (**b**) Q-normalised results.

applied to the Finnish data. Finally, in all cases we subtracted the background concentration from the measurements and compared these net observed concentrations with the model predictions.

Uncertainties in input data

Regarding emission data, accurate traffic counts were available from all sites so these data should be of relatively good quality. However, when the traffic is low, a large "statistical" uncertainty occurs. Although low emission values do not play a large role in the absolute concentrations, they are quite important when normalising with emissions. Therefore, we used a minimum value of 300 vehicles per hour as a lower limit for the Q-normalised data.

There is a high relative uncertainty in the wind speed measurements when the wind speeds are low (< 1 m s^{-1}). In addition, for low wind speeds, the mean wind direction is not well defined. This is particularly important for wind directions parallel to the road.

Results and discussion

Concentrations normalised with emissions

The main feature for all models is a decrease of R^2 in case of normalized data, due to the natural positive correlation between observations and emissions (Table 3 and Fig. 2). Scatter plots of modelled *versus* observed NO_x for OML-Highway and WORM applied to the Norwegian and Danish data, respectively, are shown in Fig. 3. Generally, when Q-normalising the concentrations, the larger the decrease in R^2, in respect to the non-normalised values, then fewer dispersion parameters are related to the observations. Another feature is a decrease in RB when normalising; this is particularly evident for WORM applied to the Danish data (Fig. 3). As mentioned, the Danish institute recalculated the Norwegian emissions. This was found to result in a difference of ~0.1 in the RB, and did not affect R^2. RB for all models applied to the Norwegian, Danish and Finnish data (Fig. 4) shows that it is evident that the values of RB for the models applied to the Finnish data were similar for both non-normalised and Q-normalised concentrations (Table 4). However, in the Danish dataset, and also to a lesser extent in the Norwegian dataset, RBs of the Q-normalised concentrations were smaller than RBs of the non-normalised concentrations. This was true for the majority of the models. An analysis of the Q-normalised modelled and observed concentrations *versus* emissions (not shown here), showed that the majority of the models underestimated the observed concentrations when the emissions, and hence the traffic volumes, were low, and overestimated when the emissions were high. Therefore, a decrease of RB occurred when normalising the concentrations with the emissions, since the concentrations were underestimated at low emissions (low traffic volumes). Assuming the emissions are valid, though less certain for lesser traffic volumes, this indicated that all the models overestimated the dispersion at lower traffic volumes and this in turn was related to the initial dispersion by TPT. In all models, an

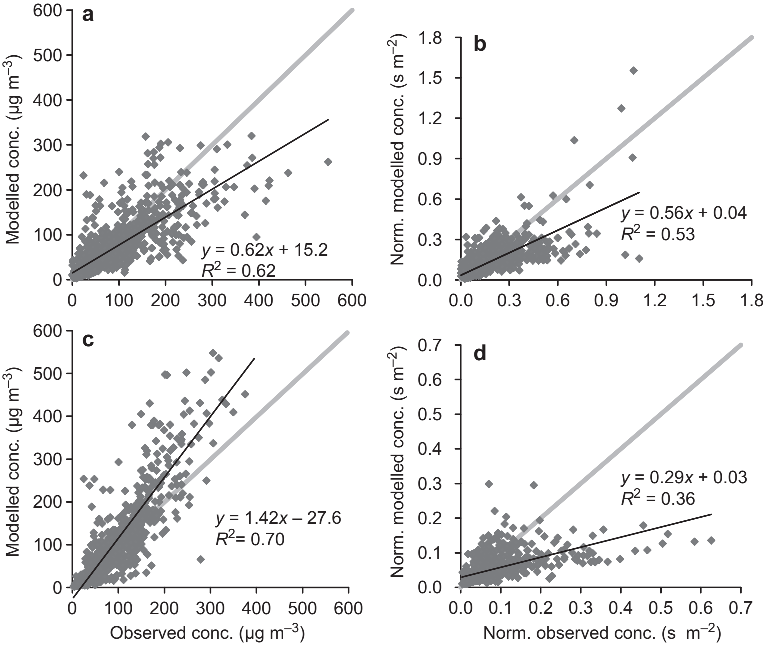

Fig. 3. Scatter plots of modelled *versus* observed concentrations, for (**a**) OML: Norwegian data (station 3) non-normalised, (**b**) OML: Norwegian data (station 3) *Q*-normalised, (**c**) WORM: Danish data (station 2) non-normalised, (**d**) WORM: Danish data (station 2) *Q*-normalised. Included in the plots are the one-to-one line (grey line) and the linear regression fit (black line), with the regression model and coefficient of determination (R^2).

Table 3. Coefficient of determination, R^2 for all models applied to all data, for both non-normalised and *Q*-normalised results.

	HIWAY2-AQ Non-norm.	HIWAY2-AQ *Q*-norm.	OML-Highway Non-norm.	OML-Highway *Q*-norm.	CAR-FMI Non-norm.	CAR-FMI *Q*-norm.	WORM Non-norm.	WORM *Q*-norm.
Norwegian data								
St. 1	0.50	0.18	0.72	0.69	0.50	0.23	0.72	0.42
St. 2	0.52	0.21	0.68	0.60	0.46	0.28	0.68	0.47
St. 3	0.48	0.20	0.62	0.53	0.46	0.37	0.64	0.49
Danish data								
St. 1	0.38	0.18	0.75	0.65	0.49	0.25	0.65	0.28
St. 2	0.34	0.24	0.74	0.61	0.41	0.36	0.70	0.36
St. 3	0.31	0.27	0.71	0.56	0.43	0.50	0.71	0.43
Finnish data								
VAN#1	0.51	0.49	–	–	0.47	0.44	0.51	0.51

Table 4. Relative bias, RB, for all models applied to all data, for both non-normalised and *Q*-normalised results.

	HIWAY2-AQ Non-norm.	HIWAY2-AQ *Q*-norm.	OML-Highway Non-norm.	OML-Highway *Q*-norm.	CAR-FMI Non-norm.	CAR-FMI *Q*-norm.	WORM Non-norm.	WORM *Q*-norm.
Norwegian data								
St. 1	0.02	–0.16	–0.21	–0.22	–0.11	–0.16	–0.31	–0.34
St. 2	0.13	–0.07	–0.19	–0.19	0.03	–0.02	–0.26	–0.29
St. 3	0.12	–0.10	–0.20	–0.22	0.18	0.12	–0.24	–0.28
Danish data								
St. 1	0.16	–0.27	0.04	–0.18	0.42	0.08	0.11	–0.22
St. 2	0.15	–0.35	0.00	–0.30	0.67	0.24	0.13	–0.26
St. 3	0.06	–0.42	0.01	–0.31	0.74	0.29	0.10	–0.28
Finnish data								
VAN#1	–0.13	–0.14	–	–	0.09	0.09	–0.48	–0.49

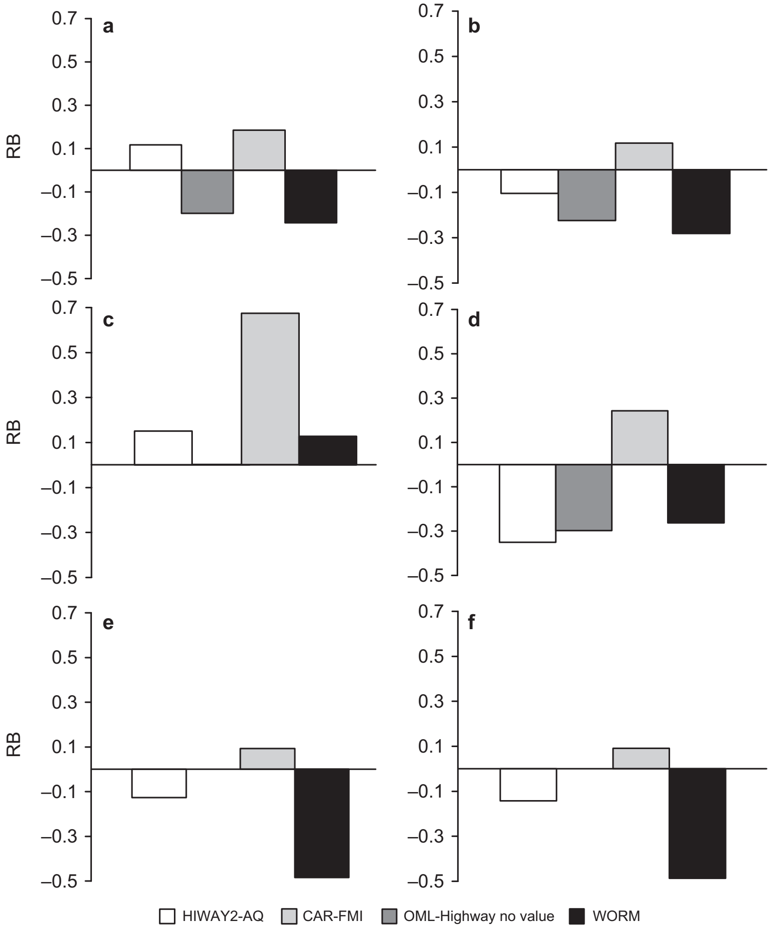

Fig. 4. Relative bias (RB) for all models applied to the Norwegian (station 3), Danish (station 2) and Finnish (VAN station) data. (**a**) Norwegian (station 3) non-normalised, (**b**) Norwegian (station 3) *Q*-normalised, (**c**) Danish (station 2) non-normalised, (**d**) Danish (station 2) *Q*-normalised, (**e**) Finnish (VAN station) non-normalised, (**f**) Finnish (VAN station) *Q*-normalised.

initial dispersion $\sigma_{z,\text{initial}}$ (Eqs. 5 and 10) was used and this appeared to be too large for low traffic volumes, by roughly a factor of three.

With regard to overestimation at higher emission values, analysis (not shown here) also showed that all models except OML-Highway overestimated more for high emission values when applied to the Danish data than when applied to the Norwegian data. This causes the different behaviour with regard to OML-Highway and WORM applied to the Norwegian and Danish data (Fig. 3), respectively. The Danish measurements were carried out on a highway with much higher traffic than the Norwegian ones; ~100 000 vehicles per day as compared with ~36 000 vehicles per day, respectively. The average vehicle speed at the Danish site was also higher, ~109 km h^{-1} as compared with ~90 km h^{-1} at the Norwegian site. As a result, dilution due to TPT should be higher at this site. OML-Highway performed better for higher emissions due to its formulation of TPT, based on parameterisation of the decay of turbulent kinetic energy. Below, we discuss this feature in more detail with regard to wind speed.

The effect of wind speed and wind direction

Scatter plots of the ratio of modelled to observed concentrations *versus* wind speed at 2 m above ground for all models applied to the Norwegian and Danish data at stations 3 and 2, respectively (Fig. 5), show that more scatter and over-predictions were present for low wind-speed conditions, due to more uncertainty in the modelling and the observations; the latter will lead to scatter irrespective of the quality of the modelling. For the Norwegian data at higher wind speeds, underestimations were evident, more so than on the Danish site. This difference occurred as a result of the differences between the datasets with regard to traffic volumes as previously mentioned; higher traffic volumes and emissions at the Danish site imply greater significance of TPT, and the TPT formulation in HIWAY2-AQ, CAR-FMI and WORM was not adequate in this respect.

The ratio of modelled to observed concentrations *versus* wind direction for all models applied to the Norwegian and Danish data (Fig. 6), show that the largest scatter, with significant overestimate, was present under wind directions parallel to the road. Some models performed poorly under such conditions due to inaccuracies in the integration methods, in particular the analytical Luhar and Patil approximation as used by CAR-FMI, but also to a certain extent the trapezoidal method with Richardson extrapolation as used by HIWAY2-AQ. However, observational uncertainties also played a role, as wind directions parallel to the road were poorly defined, especially for low wind speeds. WORM proved to perform best in this regard, although not perfect, as the overestimations are slightly reduced. WORM is also the only model applying Gaussian quadrature as a numerical integration technique, which is highly accurate even for parallel wind directions.

Horizontal profiles

In order to study how the models perform with regard to distance from the road, it was useful to study the *Q*-normalised RB for each station at the Norwegian and Danish sites (Fig. 7). The behaviour of RB is dependent on the initial dispersion, $\sigma_{z,\mathrm{initial}}$, caused by TPT, and the atmospheric dispersion. When applied to both datasets, RB for CAR-FMI increased with increasing distance from the road, indicating that the dispersion did not evolve at the rate indicated by the observations. The Lagrangian time scales, T_L, were probably too small in this model (under stable conditions, T_L = 30 sec, *see* Table 1). For both datasets, stable and unstable conditions amounted to ~40% and 15%–20% of the total amount of hours, respectively. Hence, short time scales dominated, and the overestimate became more pronounced with time and distance from the source. This is shown in Eq. 12 where short time scales imply less dispersion. With regard to all models except CAR-FMI and to a lesser extent WORM, the values of RB decreased as a function of distance from the source, when applied to the Danish data. The average observed wind speed at the Danish site was higher than at the Norwegian site (4.6 m s^{-1} and 2.6 m s^{-1}, respectively). Hence, the atmospheric turbulence played a more significant role as the significance of TPT decreases with distance from the source.

The effect of stability

Differences between the datasets appear clearly when studying RB for all models applied to the Danish and Norwegian datasets for different Pasquill-Gifford stability classes (Turner 1969) for the *Q*-normalised concentrations (Fig. 8). With regard to the Norwegian data, the majority of the models underestimated the concentrations for all stability classes. However, a larger degree of overestimate, especially for unstable conditions, was evident when the Danish dataset is applied, consistent with the analysis in 5.1 and 5.2. HIWAY2-AQ, on the other hand, consistently overestimated the unstable conditions on both datasets because it uses the neutral class for all unstable cases, hence, it did not describe all of the dispersion, which clearly is a weakness with the model. CAR-FMI stood out with regard to its consistent overestimate of stable conditions (classes E–F). For CAR-FMI, this indicated that the positive biases (Figs. 4 and 7) were caused by the stable conditions, which represented most of the data. For both datasets, the lowest observed wind speeds occurred under unstable and stable conditions (classes A–C and E–F, respectively), while the highest observed wind speeds occurred under neutral conditions (class D), where the biases were closest to zero (especially at the Danish site), hence, this was consistent with the distribution of bias discussed earlier

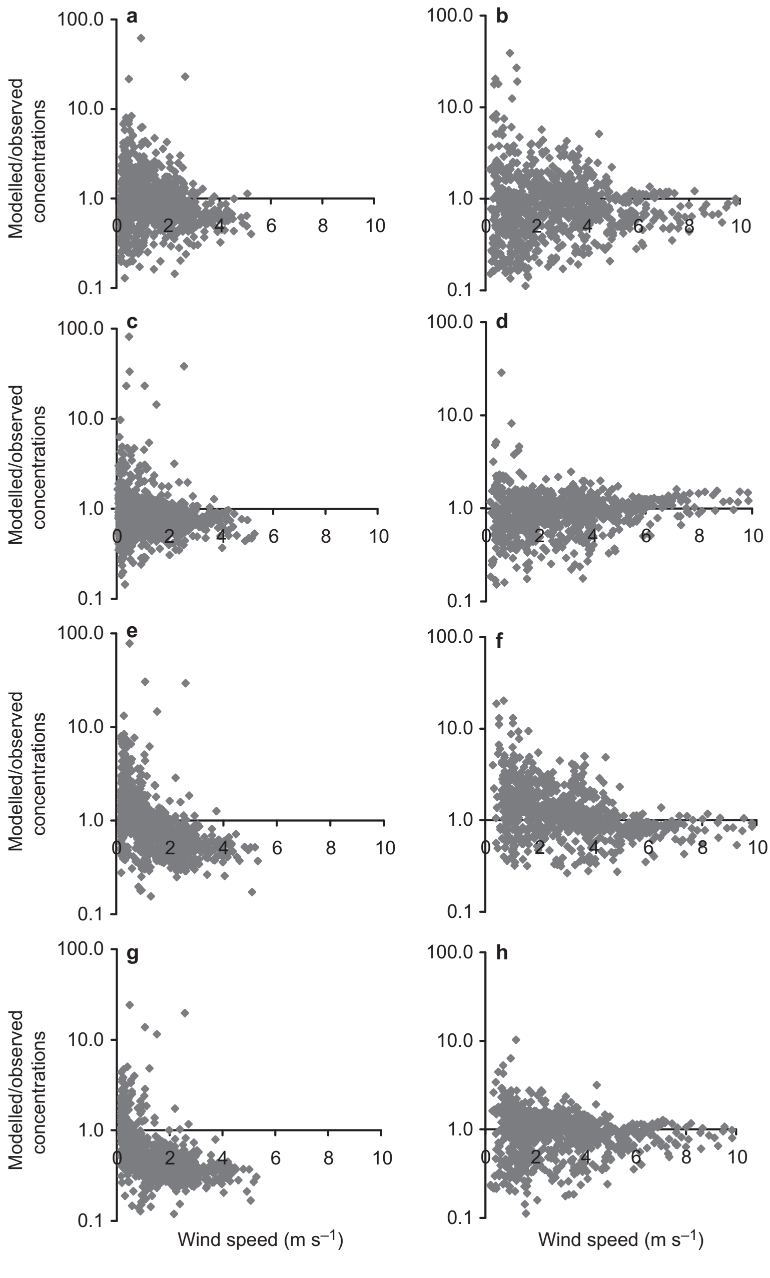

Fig. 5. Scatter plots of the ratio of modelled to observed concentrations *versus* wind speed at 2 m above ground for all models applied to the Norwegian (**a**, **c**, **e**, **g**: station 3) and Danish (**b**, **d**, **f**, **h**: station 2) data.

(Fig. 5). For all stability classes, OML-Highway performed best with regard to bias because of its formulation of TPT. It was therefore likely that OML-Highway's formulation of TPT also played a significant role in model performance with regard to stability; however, as mentioned, the initial $\sigma_{z,\mathrm{initial}}$ must be revised.

Conclusions

Four open road line source models were compared and evaluated based on their application on datasets from measurement campaigns in Norway, Denmark and Finland. The specific aim was to determine under which conditions the

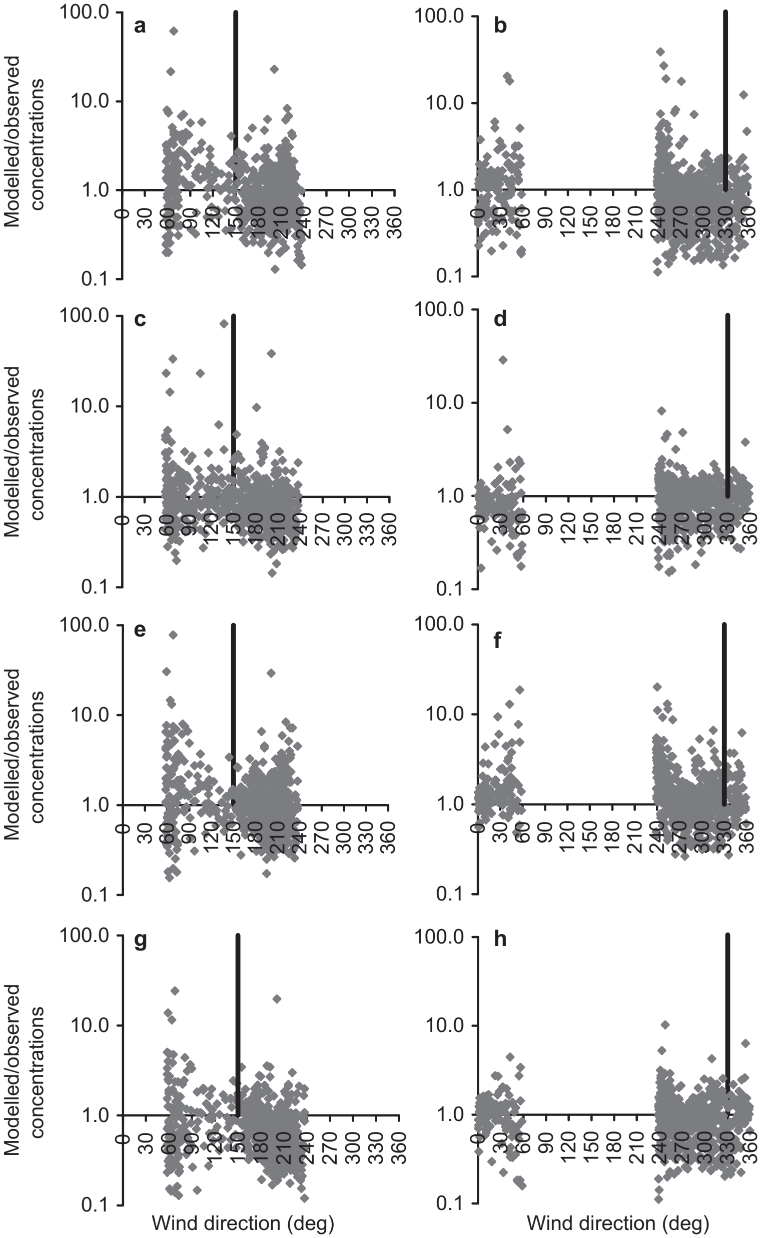

Fig. 6. Same as Fig. 5, but plotted against wind direction with respect to north. The vertical lines indicate the direction perpendicular to the road when the stations are downwind of the road.

models perform well or poorly. The measurement campaigns were conducted near highways in open environments.

When normalising with emissions, R^2 generally decreased as the natural positive correlation between emissions and observations is removed. Analysis of the data indicated that reduction in RB in the Norwegian and Danish data after normalising was caused by overestimation of the dispersion at lower traffic volumes and lower emission values. This occurred because the initial dispersion, $\sigma_{z,\mathrm{initial}}$, was too large in all the models. Also, all models except OML-Highway gave higher RB for high emission values when applied to the Danish data than when applied to the Norwegian data, due to the increased sig-

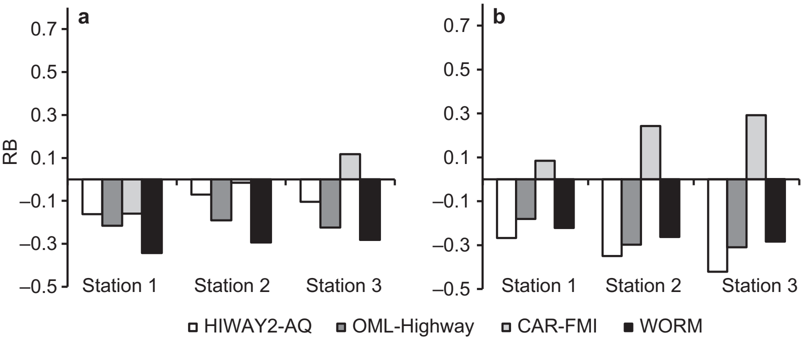

Fig. 7. *Q*-normalised relative bias (RB) for all models applied to (**a**) Norwegian data (all stations) and (**b**) Danish data (all stations).

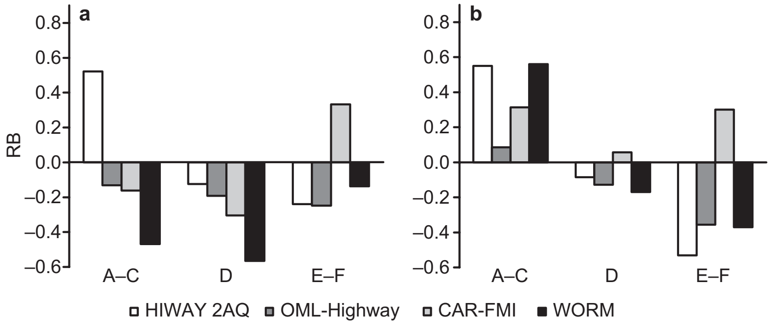

Fig. 8. *Q*-normalised relative bias (RB) for all models applied to (**a**) Norwegian (station 3) and (**b**) Danish (station 2) data for Pasquill-Gifford class A–C (unstable), D (neutral) and E–F (stable).

nificance of TPT at the more heavily trafficked Danish site. The latter feature was also seen in the scatter plots of the ratio of modelled to observed concentrations versus wind speed, at higher wind speeds. OML-Highway performed best in this regard due to its parameterisation of TPT based on decay of turbulent kinetic energy.

OML-Highway's parameterisation of TPT (or similar ones), should be implemented in ORLS models, to describe the turbulence produced by the traffic. However, the initial dispersion must be reduced in order to describe the concentrations when the emissions are low. The OML-Highway formulation is currently being implemented in WORM. Furthermore, in order to reduce uncertainties appearing under near to parallel wind directions, Gaussian quadrature methods, or other highly accurate numerical integration methods, should be implemented in ORLS models.

With regard to horizontal profiles, RB for CAR-FMI increased with increasing distance from the road. This indicates that the Lagrangian time scales, T_L, are too short, and need to be revised. RB for the other models decreased with distance from road at the Danish site, which is an indication of the increased significance of atmospheric turbulence at larger distances from the road.

It is important that the effective transport velocity, u_h, and the height at which it is calculated is well described and documented, as u_h is highly important for the dispersion of pollutants. The stability corrections (e.g. the Businger-Dyer relations (Businger *et al*. 1971)) should be included.

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