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**DETERMINATION OF AVERAGE EMISSION FACTORS
FOR VEHICLES ON A BUSY ROAD**

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Abstract

In this paper, the CALINE4 software package, designed for calculation of concentrations of carbon monoxide near a busy road, is adapted for the analysis of aerosols of fine and ultra-fine particles, generated by vehicles on the road. A scaling procedure of the CALINE4 package is developed and justified. A new efficient method of determination of the average emission factor for fine particle emission from a vehicle on a given road is also developed. This method is based on measurements of the average particle number concentration at just one point near the road.

An example of a specific road in the Brisbane area, Australia, is considered. The average emission factor for the vehicles on this road is calculated to be $\sim 4.5 \times 10^{14}$ particles per vehicle per mile. At the same time, the obtained scaling coefficient (1.12×10^{-12} g/cm³) is shown to be correct, and the procedure is directly applicable for the analysis of an arbitrary road with different types of vehicles and their average speed.

Good agreement between the experimental results and the predicted theoretical dependencies of concentration on distance from the road clearly confirms the applicability of the CALINE4 package for the analysis of propagation of fine particle aerosols from a busy road. Statistical analysis of the experimental and theoretical results demonstrates that the concentration of fine and ultra-fine particles approximately reduces as a power law in distance from the road.

1. Introduction

Accurate prediction of aerosol concentrations and dispersion from different polluting sources is one of the main problems of modern environmental science. Obviously, this is one of the major tasks of the general problem of reliable aerosol forecasting and determination of the actual sources of aerosols in urban environment. Currently, there are two main approaches to this problem. One of them is the statistical analysis of aerosol properties and concentrations on the basis of the knowledge of previous experimental data and meteorological conditions (Morawska, 1998, Cogliani, 2000), and the other is the numerical simulation of aerosol propagation and dispersion from a given source (Csanady, 1980, Pasquill, 1983).

The statistical approach is normally based on a large amount of experimental data, which requires extensive and continuous measurements during long periods of time (usually within several years) (Morawska, 1998, Cogliani, 2000). Therefore, this method may not always be convenient for the fast evaluation of pollution from the existing and proposed roads. From this prospective, the modelling approach is more efficient, since it is based on the direct simulation of turbulent diffusion on the basis of knowledge of physical characteristics of the pollution source and meteorological conditions, such as wind speed and direction, temperature, atmospheric stability, roughness of the surface, etc. (Stull, 1989).

Until recently, the main efforts in environmental science were focused on gaseous air pollution, because this type of pollution was regarded as most dangerous to human health. Several software packages have been designed for prediction of gaseous pollution concentrations from different types of sources such as point source (e.g., factory pollution) (Bowers, 1981), line source (e.g., road emission) (Benson, 1992), and area source (e.g., bush fires) (Hanna, 1984). It is well known that busy roads is one of the main contributing sources to overall air pollution in the urban environment (Zhiqiang, 2000). Therefore, computational simulation of air pollution from a busy road is an essential problem for maintaining and forecasting of air quality in

residential and industrial areas. One of the most well developed packages for the analysis of busy road pollution is CALINE-4 (Benson,1992) that has been designed by California Transport for the analysis of carbon monoxide (and extended for the analysis of NO_x) pollution on the basis of knowledge of gaseous emission factors from stationary and moving vehicles.

However, though carbon monoxide emissions present obvious health risks for humans and the environment, its effect is limited to relatively small distances from a road. Indeed, calculations with CALINE4 demonstrate that usually concentrations of carbon monoxide and nitrogen monoxide are normally reduced to a typical background level within ~ 100 m from a road (Benson, 1992).

At the same time, recent investigation has revealed new and probably more significant health and environmental risks from busy roads. These are related to fine and ultra-fine particle pollution from motor vehicles (Dockery, 1993, Schwartz, 1996). By definition, fine particles have diameters between ~ 0.1 µm and ~ 1 µm, while ultra-fine particles have diameters from ~ 1 nm to ~ 0.1 µm (Willeke, 1993). These particles constitute only insignificant part of the overall particulate mass in the air (due to their small size) and, until recently, were neglected by Environmental Pollution Authorities and researches. At the same time, the latest research (Anderson, 1992) has demonstrated that fine particles significantly and adversely affect human health. Moreover, concentrations of fine particles, that are significantly larger than the background level can be registered at substantially larger (than for CO) distances from a road simply due to typically lower relative background concentrations (Hitchins, 2000).

However, the available software for the analysis of gaseous pollution, for example, the CALINE4 program (Benson, 1992), cannot be used for the simulation of dispersion of fine particle aerosols from a busy road. There are two main problems with using CALINE4 for this simulation. The first problem is related to the replacement of gas parameters in the program (such as the emission factor and concentrations) by those for fine particles. This cannot be done automatically due to different units for these parameters for gasses and aerosols.

The second problem arises from the absence of consistent experimental data on particulate emission factors from motor vehicles, that would be required as an input for the CALINE4 package. Indeed, experimental values of these emission factors presented by different researchers vary by one or even two orders of magnitude depending on type of vehicles and conditions of measurements (Jamriska, 2001, Cadle, 2001, Graskow, 1998, McAughey, 1996, Watson 1998, Gertler, 1999, Grass, 2000). From the same references, typically, the emission factors lie within the intervals between $\sim 10^{12}$ to $\sim 10^{14}$ particles per vehicle per kilometre for gasoline (light-duty) vehicles, and between $\sim 10^{14}$ to $\sim 10^{15}$ for diesel (heavy-duty) vehicles. Note also that very few attempts have been made to determine average emission factors for vehicles on a real road (Gertler, 1999, Grass, 2000, Jamriska, 2001) with rather inconclusive results (i.e., large spread of the obtained data and significant experimental errors).

At the same time, accurate determination of average emission factors for vehicles on a road is, in itself, of a major importance for the evaluation of the impact of road pollution on human health and environment.

Therefore, the aim of this paper is to develop a new effective and accurate method for the determination of average emission factor for a vehicle on a road (based on the knowledge of experimental total number concentration at one point near the road), and simultaneously re-scale the CALINE4 package to make it suitable for the analysis of propagation of fine particle aerosols from a busy road. The predictions obtained by means of the re-scaled CALINE4 package, and the determined emission factors will be compared with two sets of experimental measurements. An excellent agreement between the theory and experiment will be achieved.

2. CALINA4 model

Though the CALINE4 package has been specifically developed for the analysis of carbon monoxide pollution (Benson, 1992), it can readily be adapted for the simulation of fine particle propagation from a busy road. This statement is based on the following physical aspects.

First, unlike particles of larger size ($> 1 \mu\text{m}$), that have sedimentation velocities between $\sim 0.01 \text{ m/min}$ and $\sim 10 \text{ m/min}$, sedimentation velocities for fine particles are $\sim 1 \text{ mm per hour}$ or less (Jacobson, 1999). Velocities of dry deposition due to turbulent diffusion increase with decreasing size of particles. However, these velocities are only: $\sim 0.3 \text{ cms}^{-1}$ for particles with the diameter 30 nm , and $\sim 0.03 \text{ cms}^{-1}$ for 120 nm particles (Jacobson, 1999). As a result, fine particle aerosols can propagate large distances from a source without noticeable sedimentation and/or deposition.

On the other hand, the majority of the emitted particles from a busy road is within the ultra-fine range with the median diameter $\leq 0.05 \mu\text{m}$ []. As a result, one could expect increased speed of coagulation of such particles []. Moreover, a busy road is also a strong source of gaseous reactive pollutants, such as NO_x , which may lead to a possibility of nucleation processes, i.e. formation of new particles in the plume []. These processes of coagulation and nucleation are not taken into account in the CALINE4 software package that has been developed for the analysis of turbulent propagation of a non-reactive gaseous pollutants []. However, there is no clear understanding of these processes, their relationship, and contributions to the total number concentration of particles near a busy road in the current literature. In fact, the analysis and comparison with the experimental results presented below demonstrate that though the processes of coagulation and nucleation do have a profound effect on the distribution of fine and ultra-fine particles [], their combined effect on the total number concentration in an aerosol near a busy road is rather limited.

As a result, the total number concentration of particles near a busy road in the considered range (from $\sim 10 \text{ nm}$ to $\sim 1 \mu\text{m}$), as well as the average emission factors for vehicles on a road, can approximately be described by the theory of Gaussian plume [], and the accuracy of this approximation will be demonstrated by the comparison of the predicted results with the experimental data – see below. Therefore, we will use the CALINE4 package, based on the theory of Gaussian plume, for the approximate analysis of aerosol propagation near a busy road.

As mentioned above, the CALINA4 software package (Benson, 1992) enables calculation of concentrations of carbon monoxide at different distances from a road and presents these concentrations in parts per million (the number of CO-molecules per million of molecules of the air).

The inputs for the package are: roadway geometry, meteorological parameters (wind speed, wind direction and its standard deviation, temperature and humidity), background concentration (concentration of the pollutant in the absence of the traffic on the considered road) in parts per million (ppm), traffic volume (in vehicles per hour), and receptor positions. The program also requires CO-emission factors for vehicles on the road in mg per vehicle per mile.

To adapt the software CALINE4 for fine particle aerosols, we need to find scaling coefficients for emission factors and concentrations, since particle concentrations are measured in particles per cubic centimeter, and emission factors in particles per vehicle per mile. This adaptation will be done simultaneously with the determination of the average emission factor for vehicles on the road, using the experimental measurements of total number concentration at some distance from the road. After this, the scaled package will be tested by comparing its predictions with the experimental results of concentration as a function of distance from the road. Therefore, in the next section we will discuss the experimental measurements of particle concentrations, that will be needed for the developed methods and their verification, and then proceed to the adaptation of CALINE4 in section 4.

3. Experimental measurements

The experimental measurement were taken at Gateway Motorway in the Brisbane area, Australia (Hitchins, 2000). The analysed road and the surrounding area are presented in Figure 1. The total number concentration of fine and ultra-fine particles in the range from 0.015 μm to 0.7 μm was measured at the height $h = 2$ m above the ground and at distances 15 m, 55 m, 135 m,

215 m, 295 m, and 375 m from the curb of Gateway Motorway (of the total width 27 m) by means of a Scanning Mobility Particle Sizer (SMPS). Six sets of measurements (five measurements in each set) were taken during four hours at the 15 m distance from the curb of the road, and one set was taken for each other distance. Typical standard deviation of the mean for these measurements was $\approx 15\%$ for the distance 15 m from the road (this value reduced to $\sim 6\%$ for larger distances). All the concentration measurements were conducted simultaneously with measurements of the traffic flow on the road. A weather station was used to measure temperature, wind speed and wind direction at the time of concentration measurements and at the same height above the ground, i.e. at $h = 2$ m.

The results of the measurements of wind speed and wind direction are presented by points in Fig.2a,b. The solid curves in Fig.2a,b were obtained by means of the “super smoother” method available in the S-Plus statistical package. These dependencies (solid curves in Fig.2a,b) approximately correspond to one hour average values of wind speed and wind direction at any time of observation. This is important for our analysis, because calculations with CALINE4 require one hour averages for the concentration and wind parameters.

Each set of the concentration measurements took 12 minutes. The average of the measured concentrations in each set was assumed to be the one hour average that is substituted into CALINE4. The corresponding one hour average values for the wind direction and speed were taken from the curves in Figs.2a,b at the moments of time corresponding to the middle of each of the 12 minute intervals.

4. Model adaptation

The adaptation of the CALINE4 model to fine particle aerosols can briefly be outlined as follows. Firstly, we take experimentally measured concentrations of particles at some distance (e.g., 15 m) from the curb of the road. Secondly, we substitute all the known meteorological and environmental parameters, and some arbitrary numbers for the emission factors into the model.

Formally changing these emission factors, we adjust them so that the output concentration at the considered distance (15 m) from the curb of the road is equal to the experimental value of concentration divided by 1000. By doing this, we actually assume that the model gives concentration in $\times 10^3$ particles/cm³. Note however that the adjusted emission factors are not measured in particles per vehicle per mile. These are not real emission factors, but rather some input numbers for the CALINE4 model. Therefore, the adjusted emission factors will be called model emission factors and denoted as E_m .

Thirdly, substituting the determined values of E_m and the known meteorological and environmental parameters back into the CALINE4 model, we calculate the concentration of particles (in $\times 10^3$ particles per cm³) at the considered distance from the road (15 m) as a function of height above the ground (vertical concentration profile). Fourthly, taking into account the vertical concentration profile, we determine the total (integral) flux of particles through a plane that is normal to the ground, parallel to the road, and located on the windward side of the road (plane 1 denoted by the dashed line in Fig. 1). On the other hand, the same flux can easily be determined from the average (real and unknown) emission factor from a vehicle on the road, and the number of vehicles. Thus, equalling these two fluxes, we determine the unknown average emission factor, E (in particles per vehicle per mile). Comparing this average emission factor, E , with the previously obtained model emission factor, E_m , we determine the scaling coefficient $\eta = E_m/E$. Thus, the average emission factor E (in particles per vehicle per mile) should be multiplied by η , before substituting it into the CALINE4 model, in order to obtain concentration in $\times 10^3$ particles per cm³.

The following two subsections will present more detailed analysis of the outlined procedure, including the required calculations for the Gateway Motorway (Fig.1).

4.1. Model emission factors

As mentioned above, we calculate model emission factors for vehicles on the road using the concentration measurements at 15 m distance from the curb of the road. The average values of the measured parameters that were used as the inputs of the CALINE4 model in this calculation are presented in Table 1 for each of the six sets of measurements at the distance 15 m from the road.

Wind speed and direction (one hour averages) were determined from the graphs in Figs.2a,b, as described in Section 3. Standard deviations of the wind speed, temperature, and experimental concentrations are not needed for calculations using the CALINE4 model, because their effect on the calculation results is negligible. Therefore, they are not presented in Table 1. Atmospheric stability was of class one.

The background concentration has been estimated from the measurements of concentration on the same (left in Fig.1) side of the road, but when the wind blows in the opposite direction (Hitchins, 2000). These measurements gave the concentrations ~ 3200 particles/cm³. The main reason for using this estimate is related to similar densities of the residential and road areas on both sides of the road (see also the error analysis in end of this section).

Standard deviation of the wind direction is an input parameter for the model. However, the weather station gives only average value for the wind direction during a 6 minute interval (a continuous measurement) with a standard deviation s_j for the same interval. It can be shown that the standard deviation, s , of one hour average wind direction (used as an input for the model) is related to s_j as:

$$s^2 = k^{-1} \sum_{j=1}^k s_j^2. \quad (1)$$

Here, k is the number of continuous measurements undertaken within one hour period, and s_j is the standard deviation for each of these measurements. The standard deviations for one hour average wind directions, calculated by means of Eq.(1), are presented in Table 1.

Using the CALINE4 model and the data in Table 1, we obtain the corresponding model emission factors E_m for each set of measurements at 15 m distance from the curb of the road. The results of these calculations are presented in the last row of Table 1. The units for these emission factors do not matter, because these factors do not have any physical meaning, but are simply some input numbers for the CALINE4 model.

From Table 1, we can also see that the value of E_m for the first set of data is ≈ 1.5 times bigger than for the other sets. This can be explained by the fact that the first set of measurements was taken during a very busy traffic hour – between 8 and 9 am, when vehicle speed was relatively small and changed frequently. This can increase values of E_m , because it takes more time for a vehicle to travel the distance of one mile (see the units of E_m). In addition, changing speed, and in particular acceleration of vehicles obviously results in enhanced emission of particles.

The analysis of propagation of errors demonstrates that the values of E_m are relatively stable with respect to variations of the background concentration. Indeed, 20% variation of the background concentration results in only $\sim 4\%$ variation of the corresponding E_m , and 50% variation of the background results in only $\sim 10\%$ variation of E_m .

4.2. Determination of the emission factor

According to the general outline of the method, described in the beginning of section 4, each of the six values of model emission factors E_m are substituted back into the CALINE4 model, together with the corresponding values of wind speed, wind direction standard deviation, temperature and traffic flow (see Table 1). To simplify further calculations of particle fluxes, we assume that the background concentration is equal to zero, and the wind direction is normal to the road (i.e. 72° to the North – see the dashed arrow in Fig.1) for all six sets of parameters from Table 1. That is, instead of all the values in the boxes of the third row in Table 1 we use 72° . This can be done since in the calculations of the flux we do not use the experimental values of

concentrations (the seventh row in Table 1), but rather the determined values of E_m (the last row in Table 1), which are independent of the background concentration and wind direction.

As a result, concentrations of particles (in $\times 10^3$ particles/cm³) are calculated at the distance 15 m from the curb of the road as a function of height above the ground (vertical concentration profile) on the left of the road – Fig.1. It can also be seen that at the same distance (15 m), but on the lee side of the road (plane 2 in Fig.1), the concentrations are zero within an accuracy of the program (this accuracy is ~ 100 particles/cm³). This is the case for all wind speeds presented in Table 1 (if the wind direction is normal to the road). This means that the flux of particles, caused by turbulent diffusion, into the direction opposite to the wind direction is negligible. Therefore, the overwhelming contribution to particle fluxes is due to transport by wind, and practically all particles that are emitted by vehicles on the road are carried by wind through the vertical plane on the windward side of the road.

In this case, the flux F through the plane on the windward side of the road (plane 1 in Fig.1) per segment of the road of length l is given by the equation (the contribution of the turbulent diffusion to this flux is neglected):

$$F \approx l \int_0^{+\infty} U(h)c(h)dh, \quad (2)$$

where $c(h)$ is the concentration of particles as function of height h , calculated by means of CALINE4 and the procedure discussed above, and $U(h)$ is the average wind speed at the height h .

For all six sets of parameters from Table 1, the vertical concentration decreases to the background level at the height of ~ 15 meters. The average wind speed can be assumed to be constant within this height (the same is assumed in CALINE4 (Benson, 1992)). Indeed, wind starts changing with height if $h \gg 100h_0$, where h_0 is the dynamic roughness coefficient that is approximately equal to 1/30 of the average height of obstacles on the considered surface (Csanady, 1980). In our case, the road is located in a more or less flat region with isolated bushes and scattered buildings, which corresponds to $h_0 \approx 0.5$ m (Stull,1989). Therefore, at the

considered heights (up to 15 m above the ground) variation of the wind speed with height is neglected: $U(h) \approx U_0$, where U_0 is the experimentally measured wind speed at the height of 2 m above the ground level.

On the other hand, in the same approximation, the flux F is simply given by the strength of the line source q (in particles per meter per second) multiplied by the length of the considered segment of the road:

$$F \approx ql = afl/v, \quad (4)$$

where f is the number of particles emitted by one vehicle per second, a is the traffic flow in vehicles per second, v is the average speed of vehicles on the road.

Comparing Eqs. (2) and (4), gives

$$af/v = U_0 \int_0^{+\infty} c(h)dh. \quad (5)$$

This equation determines f – the average number of particles emitted by one vehicle per second. If we multiply f by the average time that takes for a vehicle to travel the distance of one kilometre (or one mile), we obtain the average (real) emission factor, E , in particles per vehicle per kilometre (or particles per vehicle per mile).

The values of E calculated by means of the described procedure are presented in Table 2 for the six sets of measurements from Table 1.

From this table, the mean value of the six presented emission factors $\langle E \rangle = (4.5 \pm 0.4) \times 10^{14}$ particle/vehicle/mile (2.8×10^{14} particle/vehicle/km), and the average scaling coefficient $\langle \eta \rangle = 1.12 \times 10^{-12}$.

Note that the errors of the mean emission factor and scaling coefficient have been calculated as the errors of the mean of the six values of E and η in Table 2. An additional error of $\langle E \rangle$ is associated with uncertainty of the background concentration. However, this additional error cannot be large due to only weak sensitivity of the resultant emission factor E to the uncertainty of the background concentration (see the end of Section 4.1). Therefore, accurate

knowledge of background concentration is not essential for the developed method of determination of average emission factors from busy roads. It is usually sufficient to have just a reasonable estimate of background concentration (with an acceptable error of up to ~ 100%).

Note also that the determination of the scaling coefficient η can be carried out without using any experimental measurements. That is, the described procedure of determining η can be used with arbitrary (hypothetical) emission factors and meteorological parameters. Indeed, when calculating the scaling coefficient in the beginning of this Section, we assumed that the wind is normal to the road, and the background concentration is zero. In the same way, all other parameters in Table 1 can be chosen arbitrarily, including the model emission factors. It can be seen that the subsequent calculation and comparison of the corresponding particle fluxes at some distance from the road (for different input parameters) give similar scaling coefficients as those presented in Table 2, with the same mean value $\langle\eta\rangle \approx 1.12 \times 10^{-12}$. The only error of this result is related to the uncertainty of calculations by means of the CALINE4 model (i.e., by the sensitivity of the model). The formal error of the mean of the obtained value of $\langle\eta\rangle$ is ~ 1%, and it can be made arbitrarily small by taking more than six calculations of η , using different input parameters.

As soon as the scaling coefficient is known, it is very easy to use the CALINE4 model for the determination of average emission factors from vehicles on different roads. To do this, we only need to measure the average total number concentration at just one point near the road, find the model emission factor (using the CALINE4 model) that produces this concentration for given meteorological conditions, and multiply the model emission factor by the scaling coefficient.

5. Comparison of numerical and experimental results

Note again that the procedure for the determination of the emission factors and scaling of the CALINE4 model for the analysis of fine particle aerosols from a busy road has been

developed on the basis of the Gaussian plume theory and experimental measurements of particle concentrations at a particular distance from the road (in our case it was 15 m from the curb of the road). The obtained emission factors are valid only for the particular road under consideration (since they depend on the average speed and type of the vehicles). At the same time, the determined scaling coefficient is correct for any road and is characteristic for the considered software package (CALINE4).

The described procedures were based on the assumption that the CALINE4 model can be used for the analysis of dispersion of fine particle aerosols from a busy road. Therefore, in this section, we verify this original assumption by means of calculating a theoretical dependence of total number concentration on distance from the road, and comparing it with the experimental measurements. The theoretical dependence is obtained by substituting the average model emission factor and the corresponding meteorological parameters, averaged over the four hour interval of measurements, into the CALINE4 model and calculating average particle concentration as a function of distance from the road. The average value of $E_m = 480 \pm 50$ is obtained by averaging the six values in the sixth row of Table 1. The average wind speed for the period of four hours is equal to 1.3 m/sec, with wind speed standard deviation being 0.5 m/sec. The average wind direction for the same period is 17° to the North, and the standard deviation $\approx 46^\circ$. The average traffic flow for the period of measurement is 3218 vehicles per hour (with the error of the mean ≈ 90 vehicles per hour), and the average temperature is 34.9°C (summer period). The background concentration was estimated to be ~ 3200 particles/cm³ – see above.

In the input of CALINE4, coordinates of receptor points can only be integer numbers (in meters) that have to be typed in separately for each of the point. Therefore, it is inconvenient to use this model for plotting an actual theoretical dependence of concentration on distance from the road. Instead, we calculate concentrations only at the distances, corresponding to the experimental measurements (see Section 3). After this, we perform similar curve fitting

procedure (see below) for both experimental and theoretical points, and compare the resultant curves.

The experimental values of concentration at the mentioned distances from the road are presented in Fig.3 by big dots. The theoretical points are not presented in the figure, as they all lie almost exactly on the corresponding theoretical curve (curve 2 in Fig.3a).

The curve-fitting procedure for the experimental and theoretical points was based on the self-similarity theory of concentration distribution (Csanady, 1980). This theory approximates the concentration c (in $\times 10^3$ particles/cm³) as a power law in distance from the road:

$$c = Kd^\mu + c_0, \quad (6)$$

where d is the distance from the road in meters, K and μ are constants to be determined, and c_0 is the background concentration. The constants $K = 289$ and $\mu = 0.73$ (for the experimental curve), and $K = 496$ and $\mu = 0.88$ (for the theoretical curve) were calculated by means of the non-linear regression model in the S-Plus statistical package (Venables, 2000).

The resultant experimental and theoretical dependencies of concentration on distance from the road are presented in Fig.3 by curves 1 and 2, respectively. The significant scatter of experimental points around curve 1 in Fig.3 can be explained by changing average speed and direction of the wind during the period of measurements. Indeed, as can be seen from Figs.2a,b, variations of wind direction within the four hour interval are $\sim 60^\circ$, and variations of wind speed are ~ 1 m/s for the same period.

Curves 1 and 2 in Fig.3 demonstrate an excellent agreement between the theory, based on the approaches developed in this paper, and the experimental results for dispersion of fine and ultra-fine particle aerosols from a busy road. However, using curves 1 and 2 in Fig.3, it is difficult to judge if the theoretical curve lies within the error range for the experimental curve or not. To answer this question, we subtract the background concentration c_0 from all the values of number concentration and re-draw the resultant dependencies $c - c_0$ on distance from the road in the logarithmic scale, where these dependencies must be linear – Fig.4a. The dotted curves in

Fig.4a give the standard errors (Hamilton, 1991) for the experimental (solid) line. It is clear that the theoretical (dashed) line indeed lies well within the standard error of the experimental curve, and CALINE4 with the calculated scaling coefficient can be used for the analysis of aerosol propagation from a busy road.

At the same time, this agreement has been demonstrated so far only for one set of measurements. Therefore, in order to have a better confirmation for the theoretical model, we have taken another set of measurements in the same size range (for particles from 0.015 μm to 0.7 μm), at the same place near the same road, but at the following (different) atmospheric conditions. The average wind speed for the period of four hours is equal to 1.8 m/sec, with wind speed standard deviation being 0.8 m/sec. The average wind direction for the same period is 131° to the North with the standard deviation $\approx 50^\circ$. The average temperature is 20.5°C (winter period). The atmospheric stability was of class one. This time, the background was measured to be ≈ 2400 particles/cm³ with the standard deviation ≈ 240 particles/cm³ (note also that this measured value also confirms the estimate of the background for the previous set of measurements). The measurements of the total number concentration were taken at the following distances from the curb of the road: 15 m, 40 m, 65 m, 90 m, 115 m, 190 m, and 265 m; (the width of the road is 27 m). The corresponding experimental points are presented in Fig.3 by small dots (the distance on the horizontal axis is taken from the middle of the road).

As mentioned above, two inputs for the CALINE4 model are the traffic flow and the model emission factor. These two input are multiplied in the model to produce the strength of the line source (in particles emitted from one mile of the road per unit of time). Actually, the model works with the strength of the line source, rather than with the model emission factor and the traffic flow. Therefore, to plot the theoretical curves, we do not necessarily have to calculate the traffic flow and then adjust the model emission factor so that to obtain the measured total number concentration at some distance from the road. Instead of counting the traffic flow, it is easier to adjust directly the strength of the line source so that to obtain the measured

concentrations. The adjusted strength of the line source has been determined for each experimental point (small dots in Fig.3), and the average adjusted strength has been obtained. Using the adjusted strength of the line source, we obtain the theoretical number concentrations corresponding to the indicated distances (as previously, the corresponding theoretical points are not shown in Fig.3).

Again, using the non-linear regression model in the S-Plus statistical package (Venables, 2000), the corresponding theoretical and experimental curves were plotted (curves 3 and 4 in Fig.3). Both these curves are noticeably lower than curves 1 and 2 (for the previous set of measurements). This is expected, since the wind speed for the second set of measurements was noticeably larger (1.8 m/s compared to 1.3 m/s). If the wind is taken the same for both sets of measurements, all the curves appear to be very close to each other. In particular, this also demonstrates that the effect of temperature (in the considered range) on average emission factors and aerosol dispersion is only weak or even negligible.

Similarly to curves 1 and 2 (for the first set of measurements), curves 3 and 4 in Fig.3 clearly demonstrate good agreement between the theoretical model and experimental results. The dependencies of $(c - c_0)$ on distance from the centre of the road in logarithmic scale (Fig.4b) again demonstrate that the theoretical line lies within the experimental error for the experimental dependence.

It is interesting that the difference in slopes between the experimental and theoretical lines is practically the same for Figs.4a and 4b. This is possibly an indication on some additional processes resulting in deviations of the experimental data from the Gaussian plume theory (e.g., due to particle loss caused by coagulation).

It is important to note that the level of confidence

The average emission factor for vehicles on the road for the second set of measurements was again calculated from the measured average number concentration at the distance 15 m from the curb of the road and the counted traffic flow 4212 vehicles per hour. The calculated value of the emission factor in this case is $E \approx 4.6 \times 10^{14}$ particle/vehicle/mile (with the error $\sim 16\%$

mainly due to uncertainty of concentrations), which is very close to the value of the emission factor, calculated from the summer experiment ($E \approx 4.5 \times 10^{14}$ particle/vehicle/hour - see above).

Both the obtained values of the average emission factor are in reasonable agreement with the previous results obtained by means of a box model [], where the average emission factor was estimated as $\approx 2.8 \times 10^{14}$ particles/vehicle/mile. The discrepancies could be explained by the larger number of heavy duty vehicles in our experiments (14% compared to just 4% in []), and by the significant uncertainty of the results from the box model ($\sim 70\%$ []).

6. Conclusions

In this paper, the CALINE4 software package, that was originally designed for calculation of concentrations of carbon monoxide near a busy road, has been adapted for the analysis of aerosols of fine and ultra-fine particles, generated by vehicles on a busy road. As a result, the scaling procedure for the available CALINE4 model has been developed and justified. A scaling coefficient relating the model and real emission factors has been determined: $\eta \approx 1.12 \times 10^{-12}$. A new method of determination of the average emission factor for fine particle emission from a vehicle on a road has also been developed. This method is based on experimental measurements of particle concentration at just one point at some distance from the road. The average emission factor of $\sim 4.5 \times 10^{14}$ particles per vehicle per mile (with the standard deviation of $\sim 10\% - 15\%$, depending mainly on uncertainty of concentration measurements) has been determined for the road under consideration (Gateway Motorway, Brisbane, Australia). This value must obviously be different for a different roads, since it depends on average speed and type of the vehicles on the road. At the same time, the determined scaling coefficient and the whole procedure of the analysis are correct for an arbitrary road, and arbitrary meteorological and environmental conditions, as long as we use the CALINE4 software package.

It is important that the determined scaling coefficient gives us an easy way to calculate an average emission factor for vehicles on a road, using only measurements of average concentration at one point in the vicinity of the road. When applied to different roads, this method may lead to an estimate of emission factors for different types of vehicles [].

The suggested procedure of the analysis and the calculated values of average emission factors for fine particles have been shown to be relatively stable with respect to noticeable variations of the background concentration. For example, for the considered road ~ 100% variation of the background concentration results in just ~ 20% variation of the calculated emission factors. Thus the exact knowledge of the background concentration is not essential for the developed procedure.

Good agreement between the experimental results for two sets of summer and winter measurements and the predicted theoretical dependencies of concentration on distance from the road has clearly confirmed the applicability of the CALINE4 package for the analysis of propagation of fine particle aerosols from a busy road.

Statistical analysis of the experimental and theoretical results has also demonstrated that the concentration of fine and ultra-fine particles reduces as a power law in distance from the road. The consistent differences in slopes for theoretical and experimental dependencies in the logarithmic scale have been interpreted as a possibility of additional processes resulting in particle losses, e.g., particle coagulation. However, it has been demonstrated that the contribution of such processes to the total number concentration is within the limits of the uncertainty for the experimental results.

The main applicability conditions for the developed model are the same as for the CALINE4 software package using the line source approximation. For example, it is not applicable for roads with traffic lights, roads in canyons and tunnels, roads with very low traffic flow (e.g., one vehicle per several minutes), etc.

7. References

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Table 1.

Set number	1	2	3	4	5	6
Wind speed, (m/s)	0.7	1.0	1.5	1.6	1.5	1.6
Wind direction, (degrees form the North)	17.5	358.2	351.2	1.7	21.9	35
Standard deviation of wind direction, (degrees)	55.5	53.7	48.7	37.5	41.0	38.0
Temperature, (°C)	31.0	34.8	33.4	35.5	36.8	36.4
Traffic flow, (vehicles per hour)	2928	3096	3108	3456	3216	3504
Average experimental concentration, ($\times 10^3$ particle/cm ³)	49.9	26.5	19.7	20.8	23.5	22.0
Model emission factor, E_m	729	462	502	390	422	393

Table 2.

Set of measurements	1	2	3	4	5	6
Real emission factors, E, [10 ¹⁴ particles/vehicle/mile]	6.51	4.64	4.84	3.78	4.07	3.41
Scaling coefficient, $\eta = E_m/E$, [10 ⁻¹² g/cm ³]	1.19	1.06	1.10	1.08	1.09	1.20

Figure captions

Fig.1. Gateway Motorway and the sample road. Dashed arrow indicates the direction of the wind for calculations of emission factor. Dashed lines represent the imaginary vertical planes that parallel to the road (used in the calculations of the particle flux). The scale of the map and the direction to the North are as indicated.

Fig.2. The dependencies of the one hour average wind speed (a) and wind direction (b) on time during the whole period of measurements (four hours).

Fig.3. The experimental (solid curves 1 and 3) and theoretical (dashed curves 2 and 4) dependencies of the total number concentration c on distance from the centre of the road. The two sets of the experimentally measured average total number concentrations are represented by the big dots (for the summer measurements in 1999 (Hitchins, 2000) – experimental curve 1) and small dots (for the winter measurements in 2002 – experimental curve 3).

Fig.4. The experimental (solid curves) and theoretical (dashed curves) dependencies of the average total number concentrations without the background, $c - c_0$, on distance from the middle of the road in the logarithmic scale. The dotted curves give the standard errors for the experimental (solid) lines. **(a)** The summer set of measurements in 1999 (Hitchins, 2000); **(b)** the winter set of measurements in 2002 at the same place near Gateway Motorway, Brisbane area, Australia.