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Stefan Heinz, Richard Knoche, Klaus Schaefer, "Optimized estimation of emission rates of diffuse sources by dispersion modeling," Proc. SPIE 2506, Air Pollution and Visibility Measurements, (20 September 1995); doi: 10.1117/12.221020

SPIE.

Event: European Symposium on Optics for Environmental and Public Safety, 1995, Munich, Germany

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Optimized estimation of emission rates of diffuse sources by dispersion modelling

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ABSTRACT

Strategies for the reduction of emissions of diffuse sources or measures for the stabilization of these inputs into atmosphere require monitoring techniques providing high-quality data. Because in many cases these sources are difficult to access, an approach is considered combining optical remote sensing techniques for the estimation of path-integrated concentrations near sources with dispersion modelling. Different methods are considered and compared for this transport modelling. By these methods measurement configurations can be found which are optimal in different aspects for estimating emission rates. Hence, measurements can be done mached to specific source configurations and the quality of these estimations can be assessed.

1. EMISSIONS OF DIFFUSE SOURCES

Considerably damages of ecological systems, climate changes or endangerings of human health may be caused by diffuse emissions of pollutants into atmosphere. Such emissions appear in agriculture by crop farming, by livestock production systems or by manure storage. In waste management such emissions can be found related with compost storage and in the industrial field related with leakages, for instance. To develope reduction strategies or measures for the stabilization of emissions of greenhouse gases, monitoring strategies have to be applied which provide quality-assured data to the input of pollutants into atmosphere. Because in many cases it is very complicated to estimate emission rates of these gases, optical remote sensing techniques can be applied for measuring concentrations of pollutants near sources. In this way, concentrations of pollutants can be measured over a path, where the influence of small-scale fluctuations is avoided. For instance, by FTIR spectroscopy and a spectrum-analyzing programme concentrations can be measured of CO, CH_4, N_2O, CO_2, H_2O and near sources of NH_3, NO and NO_2. Using dispersion modelling the relation between emission rates and measured concentrations near sources can be calculated, so that the measured concentrations can be attributed to emisions. For this different optical remote sensing measurements can be used, so that questions arise to well-matched measurement configurations and to the quality of the results.

2. PATH-INTEGRATED CONCENTRATION MEASUREMENTS

In most cases, diffuse gas releases are caused by more or less extended areas (e. g. urban areas, complex industrial facilities, deposits, agriculture). Rapid, mobile, and multi-component measurement techniques as well as new environmental monitoring strategies are needed for a representative source identification and quantification completing conventional point sampling techniques which are unpractical or even not possible to use in many cases.

Open-path Fourier-Transform-Infrared (FTIR)-spectroscopy has achieved growing application during the last years. The application of FTIR-spectroscopy provides a path-integrated determination of gas concentrations along the line from a light source to the receiver. The path can vary from 50 m up to 1 km. Only strong rain and snow influence the measurements. This method has major advantages compared with other optical and, particularly, conventional in situ measurement methods:

- simultaneous analysis of a large number of compounds from one measurement, which is the most cost-effective aspect,
- no sample handling (no sensor contamination) and no requirement of calibration gas supplies,
- surveillance of distant sources,
- polluted area as well as clean air monitoring, and
- line-of-sight-averaged concentrations instead of point results.

0-8194-1864-1/95/$6.00

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The calculation of gas concentrations bases on the description of radiation transfer in the atmosphere. The direct calculation of gaseous absorption coefficients on the basis of a line-by-line procedure from known data on atmospheric absorption line parameters[1, 2] offers the possibility to include current atmospheric and geometrical parameters as well as spectroscopic features of the instrument (spectral resolution, line shape function) and to study interferences of spectral signatures of different compounds in gas mixtures. In open-path FTIR-spectroscopy, a high-temperature blackbody source (glowbar) is used. The thermal self radiation of the observed atmospheric layer can be neglected then. So, the propagation of radiation is described by the Beer-Lambert law. Gas concentrations are determined by a differential absorption method which has the great advantage that calibration gas measurements are not required and all broad band effects (aerosol influence, temperature variation) are eliminated. The concentrations are calculated from the transmittance ratio of the absorbing and non-absorbing part of an isolated spectral line, which is possible due to the high spectral resolution of the instrument used. The source attribution is the difference of two measurements carried out perpendicular to the prevailing wind direction at an upwind and downwind position of the source. Path-averaging of contributions of heterogeneous release points across an area source is an advantage of absorption spectroscopy. The emission rate of the source must be a factor of 10 minimum higher by the source than emissions in the surroundings. To determine the difference of upwind and downwind concentrations with high precision the detection limit of the investigated components by FTIR-spectroscopy must be a factor of 10 minimum smaller than the downward concentration of this component.

3. MODIFIED GAUSSIAN THEORY

The relation between emission rate and concentration in the measurement path can be calculated by applying modified Gaussian models.[3-6] Here, the rarefaction of considered substances by transport from the source to the measurement path may be assessed in a very simply way, where transport by the mean horizontal wind and turbulence modified by temperature stratification are considered analytically. But this approach cannot be used near sources and is related with the assumption of little structured flows. In the Gaussian approach the heigth-dependent mean horizontal wind is characterized by a single value only. But tracer experiments can be applied for estimating required parameters in order to match this concept to real conditions optimally. In this way the functions for the dispersion parameters given below had been found.
To demonstrate the approach let us consider the dispersion of pollutants in a x-y-z coordinate system, where the horizontal transport is described in the x-y area and z denotes the vertical axis. The mean horizontal wind is directed along the x-axis and is characterized in this field by one value u, which is independent of the location. Assuming for simplicity a total reflection of a considered pollutant at the soil, its steady mean ground concentration $\langle c(x, y)\rangle$ is given by

$$\langle c(x,y)\rangle = \frac{1}{\pi u \sigma_y \sigma_z} \int dx'\, dy'\, dz'\; S(x', y', z') \exp\left\{ -\frac{(y-y')^2}{2\sigma_y^2} - \frac{(z')^2}{2\sigma_z^2} \right\}, \qquad (1)$$

caused by the continuous source S(x', y', z').[7] Dispersion into the x-direction is assumed to be small compared with the transport by the mean wind field and the dispersion along the the y- and z-axes is described by σ_y and σ_z, respectively. For these, empirically functions in the distance from the source had been found for which the concentration calculated for the emission of a point source is optimally matched to the results of tracer experiments. Accordingly, one has

$$\sigma_y = R_y (x - x_0)^{r_y}, \qquad (2a)$$

$$\sigma_z = R_z (x - x_0)^{r_z}, \qquad (2b)$$

where the parameters R_y, R_z, r_y, r_z depend on the temperature stratification and x_0 denotes the source position in x-direction. Considering as an example for (1), the steady and homogeneous emission of a source with a strength s in g / m^3 / s. Let us assume that the source has a length a, a width b and a height c. It is centered at x_0, y_0 in the horizontal area and turned by an angle φ against the x-axis (see Figure 1 in section 5). Other kinds

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of sources can be obtained from this one. So, a line source is constructed by the limit $b \to 0$, or a ground point source can be obtained simply by the limits $(b, c) \to 0$. The well known formula of a point-source in a height H can be derived directy from (1). Then one finds for the mean ground concentration $\langle c(x, y) \rangle$

$$\langle c(x,y) \rangle = \frac{s}{\pi u \sigma_y \sigma_z} \int_0^c dz' \exp\left\{ -\frac{(z')^2}{2\sigma_z^2} \right\} \cdot$$

$$\cdot \int_{-a/2}^{a/2} \int_{-b/2}^{b/2} dx'\, dy' \exp\left\{ -\frac{(y - y_0 - \sin\varphi \cdot x' - \cos\varphi \cdot y')^2}{2\sigma_y^2} \right\}, \qquad (3)$$

taking source extent and orientation into account. Using FTIR-spectroscopy for concentration measurements unknown emission sources can be inspected and emission rates of different gas releases can be validated by inverting equation (3). In this way gaseous emission rates from industrial and agricultural areas, highways, garbage deposits, open-cast lignite minings, compost heaps and an ensemble of small-building smoke stacks had been estimated within the SANA programme.[8] The multi-component software MAPS[1, 2], to interpret the measured spectra and the emission rate code AREA[9] have been developed at the IFU. The concentration calculation is performed on-line on board of a mobile laboratory to give the actual values of concentrations every 5 minutes and is possible for any mixture of air pollutants provided their spectroscopic data are known. CO_2, CO, NO, N_2O, H_2O, CH_4, NH_3, NO_2 and HCHO are retrieved from absorption measurements. The detection limits are in the range from 1 to 10 ppb. The deviation between path-integrated concentrations determined by this method and GC-analysis of several air samples along the absorption path during one day differ not more than 5 %. By Piccot et al.[4] an accuracy of the above approach of at least ± 25 to 30 percent is given. A method for the estimation of measurement configurations which are optimal under different aspects is presented in the next section. But these results are bounded to the applicability of Gaussian theory. More precise calculations can be done by Lagrangian dispersion theory which is explained below. This Lagrangian modelling is required in particular, if buoyancy effects have to be taken into account, for the gaseous emission of fires and forest fires.[8]

4. OPTIMIZATION OF MEASUREMENTS

Often it cannot decided easiliy which configuration of measurement is particularly advantageous and matched optimally to the given conditions. Here, for instance one has to find out an optimal distance between measurement path and the source, the height of the measurement path over ground, the length of the measurement path, and eventually the orientation of the measurement path. The modified Gaussian models described above can be used to calculate an optimal measurement configuration to determine the emision rate of a considered source. Moreover, configurations can be found which are only minimal influenced by the parameters used in the calculations. In many cases the superpositions of contributions of different sources will be measured in the path. For instance, the concentration of ambient pollutants caused by starting aircrafts is influenced by the feeder traffic to the airport. Also in this cases the concept of optimization can be applied to calculate measurement configurations, where the influence of other sources is as small as possible.[10, 11]
Considering different sources, the mean concentration $\langle c_\alpha(x, y) \rangle$ caused by the source α can be written

$$\langle c_\alpha(x,y) \rangle = s_\alpha \cdot f_\alpha(x,y), \qquad (4)$$

where s_α denotes an emission rate in g / m^3 / s corresponding with s in the above considered case of a volume ground source and $f_\alpha(x, y)$ is a rarefaction function determining the ratio of emitted concentration at the source to the measured one at the location (x, y). Hence, the total concentration $\langle c(x, y) \rangle$ of all contributions can be written as

$$\langle c(x,y) \rangle = \sum_{\alpha=1}^{N} s_\alpha f_\alpha(x,y). \qquad (5)$$

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Measuring the ground concentration integrated over a path with length L by optical remote sensing technique, one obtains

$$\int_{(c)} dt\,\langle c(x,y)\rangle = \sum_{\alpha=1}^{N} s_\alpha F_\alpha(x_m, y_m, \varphi_m, L), \tag{6}$$

where the center of the measurement path is located in (x_m, y_m) and an angle φ_m is assumed between to the x-axis and the measurement path. For the function F_α one finds

$$F_\alpha = \int_{-L/2}^{L/2} dt\, f_\alpha(x_m + t\cdot\cos\varphi_m,\, y_m + t\cdot\sin\varphi_m), \tag{7}$$

where (x, y) in $f_\alpha(x, y)$ have to be replaced according to (7). The derived equation (6) can be used for determining different optimal measurement configurations. For instance, if various sources contribute to the path-integrated concentration measured by the optical remote sensing technique a configuration can be estimated, where the contribution of one considered source is only minimal influenced by contributions of the other sources. The emission rate s_α of this source is calculated according to (5) by

$$s_\alpha = \{1-\eta\}\cdot F_\alpha^{-1}(x_m, y_m, \varphi_m, L)\cdot \int_{(c)} dt\,\langle c(x,y)\rangle, \tag{8}$$

with

$$\eta = \left[\int_{(c)} dt\,\langle c(x,y)\rangle\right]^{-1} \cdot \sum_{\substack{\beta=1\\(\beta\neq\alpha)}}^{N} s_\beta F_\beta(x_m, y_m, \varphi_m, L). \tag{9}$$

Hence, $\eta(x_m, y_m, \varphi_m, L)$ represents the relative contribution of the other sources to the calculated emission rate s_α. This quantity has to be minimized to optimize the calculation of the emission rates, that means to estimate these rates relative independent from each other. Consequently, these values (x_m, y_m, φ_m, L) are looked for, for which $\eta(x_m, y_m, \varphi_m, L) \rightarrow$ minimum. Of course, the origin (x_m, y_m) of the measurement path has to be located in this area influenced by the considered emission.

Considering the emission of one source, it has to be decided which measurement configuration is optimal to be relative insensitive from parameters used in the Gaussian dispersion modelling. This concerns for instance for the above presented example of a volume source the source location (x_0, y_0), the extent (a, b, c), the mean wind direction along the x-axis, the horizontal wind u and the dispersion parameters $\sigma_y(x - x_0)$ and $\sigma_z(x - x_0)$. Such measurement configurations can be estimated by calculating (x_m, y_m, φ_m, L), for which the different sensitivities of s_α are as small as possible. Accordingly, to determine a measurement configuration providing a path-integrated concentration which is relatively independent of the mean wind direction, one has to solve

$$\left|\frac{\partial s_\alpha}{\partial \varphi}\right| = \frac{\partial F_\alpha^{-1}(x_m, y_m, \varphi_m, L)}{\partial \varphi} \int_{(c)} dt\,\langle c(x,y)\rangle \;\rightarrow\; \text{minimum}. \tag{10}$$

In the same way measurement configurations can be obtained, which are relatively insensitive to variations of the other parameters.

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5. APPLICATIONS

To get more insight into this approach let us consider a source configuration like shown in Figure 1. An area source with length a_1, width b_1 and height c_1 is located in a x-y coordinate system at (x_{01}, y_{01}) with an angle φ_1 against the x-axis, which is directed along the mean wind direction. Line sources are given with corresponding notations (indices 2 and 3), which lead to the area source. This configuration may be a very simple model for an airport with feeder roads. The measurement path is located at (x_m, y_m) with length L and orientation φ_m.

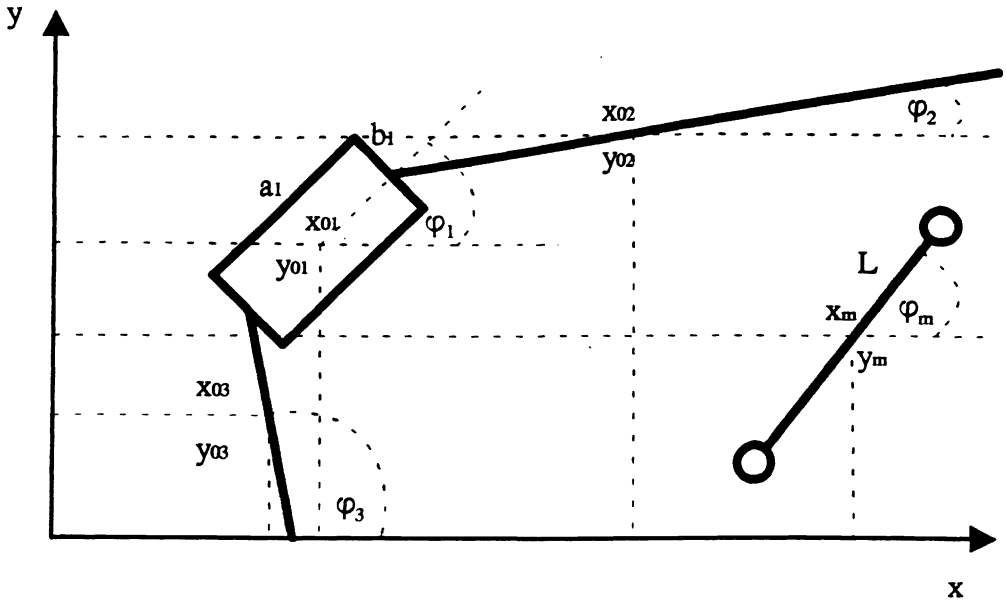

Figure 1. A configuration of area and line sources and the measurement path.

The total concentration caused by these sources and its outlines are depicted in Figure 2 for a ground area of 6 km in wind direction and 2 km in the cross wind direction.

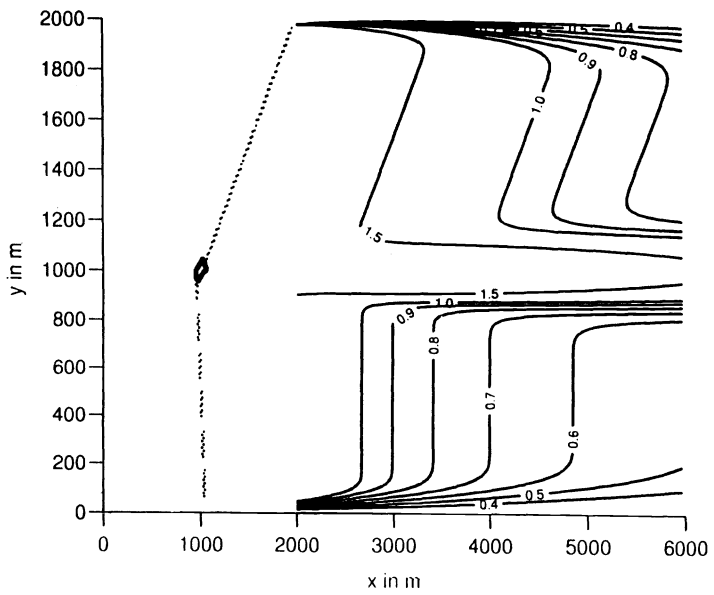

Figure 2. The total concentration field in mg / m^3 arising from three sources. The area source has a length a_1 = 100 m, a width b_1 = 50 m and a height c_1 = 1 m. It is located at (x_{01}, y_{01}) = (1 km, 1 km) and turned with an angle φ_1 = 30°. The line sources have widths $b_2 = b_3$ = 10 m, and heights $c_2 = c_3$ = 1 m and its lengths and orientations are chosen so that (x_{02}, y_{02}) = (1.5 km, 1.5 km) and (x_{03}, y_{03}) = (1 km, 0.5 km). For the emission rates $s_1\ a_1\ b_1\ c_1 = s_2\ a_2\ b_2\ c_2 = s_3\ a_3\ b_3\ c_3$ = 1 g / s are assumed. The mean horizontal wind is 2 m / s and a slightly stable temperature stratification is considered.The outlines of the sources are given additionally (thick line for the area source and dashed lines for the line sources).

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Looking for best measurements of the emission of the area source the contribution of the line sources to the path-integrated concentration can be investigated according to $\eta(x_m, y_m, \varphi_m, L) \rightarrow$ minimum. It is found that only a small influence to the total path-integrated concentration is given by the orientation of the measurement path φ_m. Accordingly, the path-integrated concentration is linear in the path length L and the optimization procedure is reduced to the problem to find values (x_m, y_m), where the contribution of the line sources to the path-integrated concentration becomes minimal. This contribution is shown in Figure 3, where only a y-range near the area source is considered.

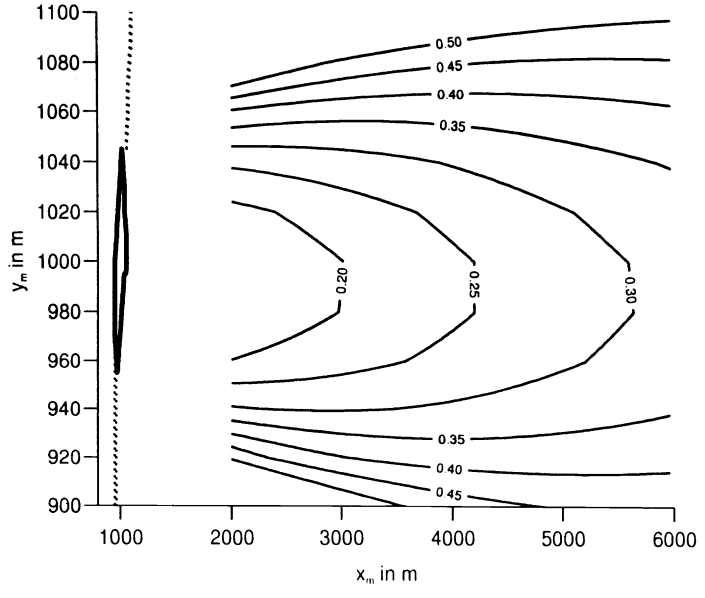

Figure 3. The relative contribution $\eta = [s_2 F_2(x_m, y_m) + s_3 F_3(x_m, y_m)] / \int dt \langle c \rangle$ of the line sources to the emission rate of the area source $s_1 = (1 - \eta) F_1^{-1} \int dt \langle c \rangle$, where $\varphi_m = 90°$ and the path length L = 100 m are assumed. The outlines of the sources are given additionally at x_m = 1000 m.

In cases of separated sources areas can be calculated, where no influence of other sources appears. But for the configuration considered here only areas of lower influence can be found. Of course, the influence of the line sources come into play with the distance of measurement position x_m from the source position x_{01}. Because such a distance to the source is required to ensure the applicability of Gaussian theory, by these calculations the error can be quantified which is related with the other emissions.

As described above the concept presented can be applied for estimating measurement configurations which are relative insensitive with respect to variations of model parameters. This is important in the most of cases, because only limited information is given to determine the latter ones. To illustrate these calculations let us consider one line source and neglect the influence of the orientation φ_m of the measurement path according to the above findings. Setting $\varphi_m = 0$ in (7) and neglecting the variation of the dispersion parameters σ_y and σ_z with φ_m in the measurement path, one finds for the path-integrated concentration field by means of (3)

$$\int_{(c)} dt \langle c(x,y) \rangle = \frac{s \cdot a \cdot b \cdot c \cdot L}{\pi u \sigma_y (x_m - x_0) \sigma_z (x_m - x_0)} \cdot \int_{-1/2}^{1/2} dt \, \exp\left\{ -\frac{1}{2} \cdot (Y - At)^2 \right\}, \qquad (11)$$

where the dimensionless quantities $Y = (y_m - y_0) / \sigma_y$ and $A = \sin\varphi \; a / \sigma_y$ are introduced. Calculating the derivative of the emission rate s by σ_z, σ_y and φ one obtains

$$\frac{\partial s}{\partial \sigma_z} \cdot \left[\frac{s}{\sigma_z} \right]^{-1} = 1, \qquad (12a)$$

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$$\frac{\partial s}{\partial \sigma_y}\cdot\left[\frac{s}{\sigma_y}\right]^{-1} = \lambda_+ - \frac{2Y}{A}\cdot\lambda_-, \tag{12b}$$

as well as

$$\frac{\partial s}{\partial \sin\varphi}\cdot\left[\frac{s}{\sin\varphi}\right]^{-1} = 1-\lambda_+, \tag{12c}$$

where the abbreviations $\lambda_\pm$ are introduced with

$$2\lambda_\pm \cdot \int_{-1/2}^{1/2} dt\ \exp\left\{-\frac{1}{2}\cdot(Y-At)^2\right\} = \exp\left\{-\frac{1}{2}\cdot(Y-A/2)^2\right\} \pm \exp\left\{-\frac{1}{2}\cdot(Y+A/2)^2\right\}. \tag{13}$$

Calculating the right-hand sides of (12b-c) one finds the following Figures 4 and 5.

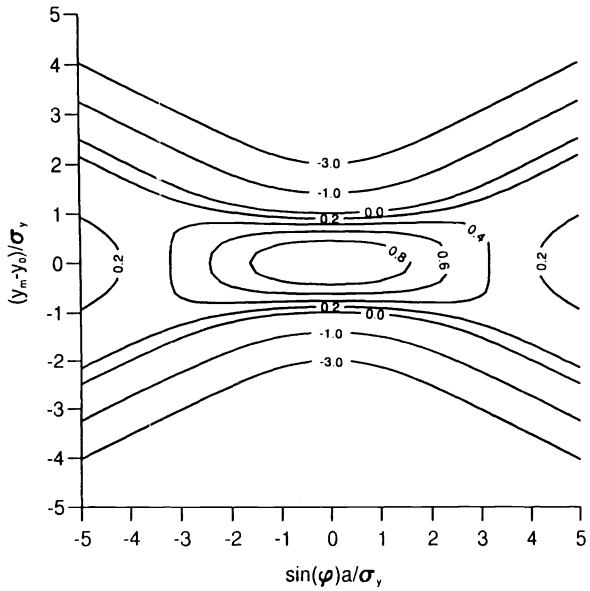

Figure 4. The relative sensitivity $\partial s / \partial \sigma_y \cdot [s / \sigma_y]^{-1}$ of a line source in dependence on $Y = (y_m - y_0) / \sigma_y$ and $A = \sin\varphi\ a / \sigma_y$.

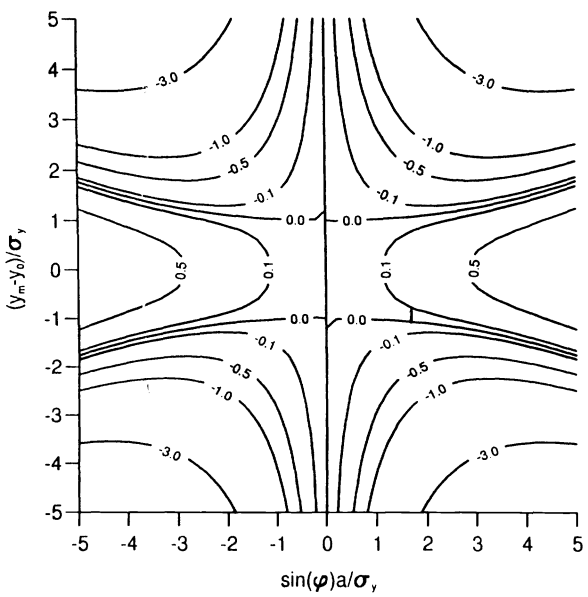

Figure 5. The relative sensitivity $\partial s / \partial \sin\varphi \cdot [s / \sin\varphi\ ]^{-1}$ of a line source in dependence on $Y = (y_m - y_0) / \sigma_y$ and $A = \sin\varphi\ a / \sigma_y$.

Accordingly we find, that the sensitivity becomes minimal if $Y = Y_{min,y}$ with

$$Y_{min,y} = \pm\left[1+(A/4)^2\right], \tag{14a}$$

and if $Y = Y_{min,\varphi}$ with

$$Y_{min,\varphi} = \pm\left[1+(A/4)^2/2\right]. \tag{14b}$$

Hence, requirements to the measurement configuration are given to ensure estimations which are relative insensitive to model parameter variations.

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6. LAGRANGIAN THEORY

More precise calculations of substance transport can be done using Lagrangian dispersion modelling.[12-16] In this approach a turbulent flow is regarded as a whole of fluid particles each having a constant mass. In correspondence with hydrodynamics equations have to be derived for the motion of these particles and their properties. These fluid particles may be characterized by different properties, like a potential temperature or a chemical composition. In this way, the flow can be calculated matched on inhomogeneous terrain and around obstacles, which is related with considerably problems in other approaches. Correspondingly, this Lagrangian approach is very convenient for describing turbulent diffusion in complex flows and near sources. Emission rates can be calculated by simulating dispersion with fitted rates and looking for the best correspondence between measured and calculated concentration in the path.

Neglecting chemical reactions, each particle is characterized at the time t by its position $x_L(t)$, velocity $U_L(t)$ (vectors with components $x_L^I(t)$ and $U_L^I(t)$, where I = 1, 2, 3 and the subscript L denotes a Lagrangian quantity) and potential temperature $\Theta_L(t)$. Combining particle velocity $U_L(t)$ and $\Theta_L(t)$ to the 4-dimensional vector $Z_L(t) = (U_L(t), \Theta_L(t))$, these equations read

$$\frac{d}{dt} x_L^{\,I}(t) = Z_L^{\,I}(t), \tag{15a}$$

$$\frac{d}{dt} Z_L^{\,i}(t) = \langle a^i \rangle + G^{ij}(Z_L^{\,j} - \langle Z_E^{\,j} \rangle) + b^{ij} \frac{dW^j}{dt}, \tag{15b}$$

where the small superscripts run from 1 to 4 in difference to capitals and summation over repeated superscripts is assumed. The first two terms in (11b) give the systematic particle motion with unknown coefficients $\langle a^i \rangle$ and G^{ij}, where the ensemble average is denoted by $\langle \cdots \rangle$. The ensemble averages of Eulerian quantities (subscript E) are estimated at fixed positions x which are replaced by $x = x_L(t)$ in the equations. The last term of (11b) describes the influence of a stochastic force, characterized by the white noise dW_j / dt and an intensity matrix b with elements b^{ij}. Here, dW^J / dt is a Gaussian process having a vanishing mean and uncorrelated values to different times, $\langle dW^1 / dt \rangle = 0$ and $\langle dW^i / dt\,(t) \cdot dW^j / dt\,(t') \rangle = \delta_{ij}\,\delta(t - t')$, δ_{ij} denotes the Kronecker delta and $\delta(t - t')$ the delta function.

The coefficients appearing in (11b), $\langle a^i \rangle$, G^{ij} and b^{ij} have to be derived in correspondence with hydrodynamic theory. This can be guaranteed up to second-order comparing the transport equations for the mean values and variances of the considered quantities. Here, the variance equations are taken in approximations of Kolmogorov and Rotta, that means a locally isotropic dissipation and a return-to isotropy are assumed. The coefficients are calculated in terms of a turbulent timescale τ and closure parameters k_1, k_3 and k_4 arising from the above assumptions. The turbulent timescale can be related with gradients of the mean wind and potential temperature fields by a rescaling procedure, and the closure parameters k_1, k_3 and k_4 can be related with dimensionless flow numbers. These equations are selfconsistent, because the calculation of mean quantities and variances can be included into the solution algorithm. As only remaining parameters three dimensionless flow numbers appear, the Prandtl number under neutral stratification Pr, the critical gradient Richardson number Ri_c, and a new introduced flow number Ri_0.

In this way, according to Kolmogorov theory for the intensity matrix B of stochastic forces with elements $B^{ij} = 1/2\, b^{ik} b^{kj}$,

$$B = \frac{1}{4\tau} \begin{pmatrix} C_0 q^2 & 0 & 0 & 0 \\ 0 & C_0 q^2 & 0 & 0 \\ 0 & 0 & C_0 q^2 & 0 \\ 0 & 0 & 0 & C_1 \langle (Z_E^{\,4} - \langle Z_E^{\,4} \rangle)^2 \rangle \end{pmatrix}, \tag{16}$$

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is found, C_0 and C_1 can be calculated in dependence on the closure parameters k_1, k_3 and k_4, $C_0 = (k_1 - 2) / 3$, $C_1 = 2k_3 - 2k_4 - k_1$, q^2 is twice the turbulent kinetic energy, that means $q^2 = \langle (Z_E^I - \langle Z_E^I \rangle)(Z_E^I - \langle Z_E^I \rangle) \rangle$, and τ is a turbulent timescale. Comparing the transport equations for the means derived from Lagrangian theory (11a-b) with the corresponding of Eulerian theory, one finds for $\langle a^i \rangle$

$$\langle a^i \rangle = \nu \frac{\partial^2 \langle Z_E{}^i \rangle}{\partial x^K \partial x^K} + (\alpha - \nu) \frac{\partial^2 \langle Z_E{}^4 \rangle}{\partial x^K \partial x^K} \delta_{i4} - \langle \rho \rangle^{-1} \frac{\partial \langle p \rangle}{\partial x^K} \delta_{Ki} - g \delta_{i3}. \tag{17}$$

where ν is the kinematic viscosity, α the coefficient of molecular heat transfer, β the thermal expansion coefficient, p the pressure and g the acceleration due to gravity. Comparing in the same way the Lagrangian and Eulerian variance equations it follows for G^{ij}

$$G^{ij} = -\frac{k_1}{4\tau} \delta_{ij} + \frac{k_1 - k_3}{2\tau} \delta_{i4} \delta_{j4} + \beta g \delta_{i3} \delta_{j4}. \tag{18}$$

Hence, $\langle a^i \rangle$, G^{ij} and b^{ij} appearing in the Lagrangian equation (11b) are given by the closure parameters k_1, k_3 and k_4, the turbulent timescale τ, and the Eulerian means $\langle Z_E^i \rangle$. Investigating the local solutions of the variance equations, where all gradients of variances and the third moments are neglected, the turbulent timescale τ normalized to the vertical sheared mean horizontal wind can be calculated in dependence on the gradient Richardson number, and the closure parameters k_1, k_3 and k_4 can be related with three dimensionless flow numbers. These flow numbers can be estimated for homogeneous turbulence.[13] The Eulerian means to be required for the calculation of the coefficients can be estimated by solving the equations (11a-b) for the whole fluid in dependence on the three flow numbers.

7. DATA QUALITY

Comparisons of these emission rate estimations with those obtained by other techniques require the assurance of an equivalent data quality. Moreover, this is the condition to combine the results obtained for single objects for the compilation of emission registers. For this, data quality experiences can be used which had been obtained to asssure the same quality of grenhouse gases. Here, demands appear to the detection limits of measurement techniques, the temporary resolution of measurements, its precision, accuracy and calibration methods. By Piccot et al.[4] an accuracy of at least ± 25 to 30 percent is given for the estimations based on Gaussian theory. By the methods presented here more insight can be provided into the accuracy aspects. Lagrangian models can be used for obtaining data of comparably quality under more complicated conditions.

8. CONCLUSIONS

Open-path FTIR-Spectroscopy are well suited to quantify atmospheric pollutants emitted from industrial and agricultural facilities and traffic. In some applications, optical methods are without a feasible alternative. The estimation of emission rates of diffuse sources is investigated using modified Gaussian and Lagrangian dispersion theory. Different concepts to optimize the measurement configuration of path-integrating optical remote sensing techniques near sources are explained. It is shown, that further investigations are needed to the improvements related with the application of Lagrangian theory. The concept of optimizing can be applied for estimating well-matched measurement configurations and to assess data quality.

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