Optimization and management of ambient pollutant concentration measurements by dispersion modeling

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ABSTRACT

Emission rates of difficult to access sources can be calculated by means of modified Gaussian dispersion theory. Here, a concept is developed for the optimization of such measurements. In particular, this method is well matched to the ability of optical remote sensing techniques to provide path-integrated concentration values and can be applied for distributed sources. Measurement configurations can be calculated which provide a maximal partitioning of the measured concentrations into the contributions of different sources. Moreover, measurement configurations can be found which are least sensitive with respect to the variations of model parameters used in Gaussian theory. To overcome application limits of the Gaussian method, Lagrangian modeling of turbulent diffusion can be used and conclusions can be drawn to the quality of results derived by Gaussian theory.

INTRODUCTION

Path integrated ambient pollutant concentrations measured using optical remote sensing methods are very suitable for comparisons with dispersion calculations, because small-scale influences are avoided. The combination of simple turbulent diffusion models and mobile remote sensing techniques allows emission rate estimates to be made for sources which are difficult to access [1].

Based on modified Gaussian dispersion theory this concept is simple to apply for a single source, since there is an analytical relation between the emission rate and the concentration measured in some distance from the source. However, in many cases different sources contribute to a pollutant distribution considered and an estimation of its emission rates by concentration measurements is very difficult.

In the case considered here, N continous emitting ground area sources contribute to a measured path-integrated concentration and the goal is to partition this total concentration into the single contributions from the

corresponding sources. This can be achieved by N different ground concentration measurements optimized to give the best seperation of the different contributions. However, these results are bounded to the application conditions of the Gaussian theory.

On the other hand, a Lagrangian dispersion model can be used for the calculation of ambient concentrations of pollutants. The advantage of this approach is based on the flexibility introduced by considering the motion of fluid parcels themselves. It can be used near sources and includes the influences of inhomogeneities and instationarities of the turbulent flow, where many other techniques are inappropriate or invalid [2]. Buoyancy effects and chemical reactions can be incorporated into this Lagrangian framework [3, 4]. Emission rates provided by Gaussian theory can be used as initial values for rate estimates by Lagrangian modeling. Here, the emission rates are fitted to give the best approximation between calculated and measured values.

GAUSSIAN MODELING

Let us consider in a x-y-coordinate system Σ the ground concentration caused by the continuous emission of N ground area sources. The contribution of a source α to the total ground concentration is described in a x_α-y_α-coordinate system Σ_α with the origin at $(x_{0,\alpha}, y_{0,\alpha})$ in Σ and φ_α as angle between the x_α- and x-axes. Here, the x_α-axis is orientated into the direction of the mean horizontal wind u_α characterizing the transport of a pollutant considered from the source α. The emission is uniform over an area $0 \le x_\alpha \le a_\alpha$ and $0 \le y_\alpha \le b_\alpha$, with the emission rate S_α (in g sec^{-1}). Accordingly, the coordinates (x_α, y_α) are related with (x, y) in Σ by

$$x_\alpha = (x - x_{0,\alpha})\cos(\varphi_\alpha) + (y - y_{0,\alpha})\sin(\varphi_\alpha), \tag{1a}$$

$$y_\alpha = (y - y_{0,\alpha})\cos(\varphi_\alpha) - (x - x_{0,\alpha})\sin(\varphi_\alpha). \tag{1b}$$

The ground concentration $\langle c_\alpha(x_\alpha, y_\alpha)\rangle$ caused by the α-th source can be calculated by modified Gaussian theory [5] as

$$\langle c_\alpha(x_\alpha, y_\alpha)\rangle = \frac{S_\alpha}{\pi u_\alpha \sigma_y(x_\alpha)\sigma_z(x_\alpha)} \frac{1}{b_\alpha} \int_0^{b_\alpha} ds \exp\left(-\frac{(y_\alpha - s)^2}{2\sigma_y^2(x_\alpha)}\right), \tag{2}$$

where we assume total reflection of the pollutant at the ground and the dispersion into mean wind direction is neglected for simplicity. In particular, an empirically determined x_α-dependence is assumed for the parameters σ_y and σ_z characterizing the dispersion into the y_α-direction and the vertical z_α-direction. Hence, writing (2) as

$$\langle c_\alpha(x_\alpha, y_\alpha)\rangle = S_\alpha f_\alpha(x_\alpha, y_\alpha), \tag{3}$$

the total concentration $\langle c(x, y)\rangle$ of all contributions in Σ is

$$\langle c(x,y)\rangle = \sum_{\alpha=1}^{N} S_\alpha f_\alpha(x_\alpha, y_\alpha), \tag{4}$$

where (x_α, y_α) transform according to (1a-b).
Furthermore, the concentration integrated over a path with lenght L, the origin (x_0, y_0) and an angle φ_0 to the x-axis of Σ is given by the path integral

$$\int_{(c)} dt\, \langle c(x,y)\rangle = \sum_{\alpha=1}^{N} S_\alpha F_\alpha(x_0^{\alpha}, y_0^{\alpha}, \varphi_0 - \varphi_\alpha, L), \tag{5}$$

where

$$F_\alpha = \int_0^{L/\sqrt{2}} dt\, f_\alpha(x_0^{\alpha} + t\cos(\varphi_0 - \varphi_\alpha) - t\sin(\varphi_0 - \varphi_\alpha), y_0^{\alpha} + t\cos(\varphi_0 - \varphi_\alpha) + t\sin(\varphi_0 - \varphi_\alpha)), \tag{6}$$

and $(x_0^{\alpha}, y_0^{\alpha})$ is the measure origin (x_0, y_0) considered in Σ_α,

$$x_0^{\alpha} = (x_0 - x_{0,\alpha})\cos(\varphi_\alpha) + (y_0 - y_{0,\alpha})\sin(\varphi_\alpha), \tag{7a}$$

$$y_0^{\alpha} = (y_0 - y_{0,\alpha})\cos(\varphi_\alpha) - (x_0 - x_{0,\alpha})\sin(\varphi_\alpha). \tag{7b}$$

The derived equation (5) can be used as starting point to determine the optimal measurement configuration.

OPTIMIZATION OF MEASUREMENTS

Accordingly, N different measurements are needed to calculate the unknown emission rates S_α ($\alpha = 1, \ldots, N$). Here, the main problem is to partition the total path integrated concentration into contributions from different sources. An optimum solution to this task results, if the single contributions of different sources to the total path integrated concentration achieve maximum seperation. The emission rate S_α is calculated according to (5)

$$S_\alpha = F_\alpha^{-1}(x_0, y_0, \varphi_0, L)\left\{\int_{(c)} dt\, \langle c(x,y)\rangle + \sum_{\substack{\beta=1 \\ (\beta\neq\alpha)}}^{N} S_\beta F_\beta(x_0, y_0, \varphi_0, L)\right\}, \tag{8}$$

where the dependence of S_α on model parameters like $x_{0,\alpha}$, $y_{0,\alpha}$, φ_α, u_α, $\sigma_y(x_{0,\alpha})$, $\sigma_z(x_{0,\alpha})$ entering into the estimation over F_α is not indicated for simplicity, requires the estimation of these (x_0, y_0, φ_0, L) for which:

$$F_\alpha^{-1}(x_0, y_0, \varphi_0, L) \sum_{\substack{\beta=1 \\ (\beta\neq\alpha)}}^{N} S_\beta F_\beta(x_0, y_0, \varphi_0, L) \quad \rightarrow \quad \text{minimum}. \tag{9}$$

The optimal measure configuration can be found to estimate S_α by solving (9). Under this condition the influence of other sources on the measured concentration is as low as possible.

With respect to the above problem, the model parameter like source location ($x_{0,\alpha}$, $y_{0,\alpha}$) and extension (a_α, b_α), wind direction φ_α, horizontal wind u_α and the dispersion parameter $\sigma_y(x_\alpha)$ and $\sigma_z(x_\alpha)$ are assumed to be known. However, this is only approximately the case, so that measurements are needed for which the influence of parameter variations is small. These measurement configurations can be estimated by calculating (x_0, y_0, φ_0, L), for which the different sensitivities of S_α are lowest. As an example, to determine a measurement configuration providing a path-integrated concentration which is relatively independent of wind-direction fluctuations, one has to solve:

$$\frac{\partial S_\alpha}{\partial \varphi_\alpha} = \frac{\partial F_\alpha^{-1}(x_0, y_0, \varphi_0, L)}{\partial \varphi_\alpha} \int_{(c)} dt \, \langle c(x,y) \rangle \quad \rightarrow \quad \text{minimum.} \tag{10}$$

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

LAGRANGIAN MODELING

The main advantage of the Gaussian approach for modeling the distribution of pollutants emitted from sources consists in the analytical relation between emission rates and calculated concentrations. However, the assumed conditions for this relation like a flat terrain and a relatively homogeneous and stationary turbulence are often violated. Instead, the Lagrangian approach was developed [6, 2, 7], which describes the stochastic motion of fluid particles. Here, turbulent dispersion in any complex flow can be calculated including the effects of inhomogeneities, instationarities and non-Gaussianity in the turbulent velocity distribution function and it can be applied near sources. On the other hand, it has not be known now, in which consistent way buoyancy effects and chemical reactions can be handled in this Lagrangian framework [8-10]. Recently Heinz and Schaller [3, 4, 11] have shown that this can be achieved in correspondence with the Eulerian hierarchy of budget equations up to second order. This topic is considered in more detail in a companion paper [12], and our representation is restricted to the main features.

The flow is considered as aggregate of fluid particles. The particles are of a constant mass and the total mass is determined by the single particle mass multiplied by the total number of particles. The equations for the particle position $\mathbf{x}_L(t)$ and velocity $\mathbf{U}_L(t)$ at the time t (the subscript L refers to a Lagrangian quantity) for the i-th component (i = 1, 2, 3, with summation over repeated subscripts) are

$$\frac{d}{dt} x_L^{\,i}(t) = U_L^{\,i}(t), \tag{11a}$$

$$\frac{d}{dt}U_l{}^i(t) = \langle a^i \rangle + G^{ij}\left(U_l{}^j - U^j\right) + G^i(\Theta_l - \Theta) + b^{ij}\frac{dW^j}{dt}, \qquad (11b)$$

where $<a^i>$, G^{ij}, G^i and b^{ij} are coefficients which can be calculated in dependence on gradients of the mean wind and potential temperature field. In equation (11b) a coupling between particle motion and the potential temperature Θ_l of the Lagrangian particle is assumed, and U^i and Θ denote ensemble averages ($U^i = <U_L{}^i>$, $\Theta = <\Theta_L>$) of the corresponding Eulerian quantities (subscript E) depending on $\mathbf{x} = \mathbf{x}_l(t)$ and t. The last term describes the influence of a stochastic force characterized by the white noise dW^i/dt, which is a Gaussian process having a vanishing mean and uncorrelated values at different times,

$$\left\langle \frac{dW^i}{dt} \right\rangle = 0, \qquad (12a)$$

$$\left\langle \frac{dW^i}{dt}(t)\frac{dW^j}{dt}(t') \right\rangle = \delta_{ij}\,\delta(t-t'). \qquad (12b)$$

Here, δ_{ij} represents the Kronecker delta and $\delta(t-t')$ the delta function. In addition, the stochastic equation for the potential temperature of the particle is

$$\frac{d}{dt}\Theta_l(t) = \langle a_\theta \rangle + S_\theta + G_\theta{}^j\left(U_l{}^j - U^j\right) + G_\theta(\Theta_l - \Theta) + b_\theta\frac{dW}{dt}, \qquad (13)$$

where the coefficients $<a_\theta>$, $G_\theta{}^j$, G_θ and b_θ are determined analogously by the gradients of the mean fields, S_θ being a source of loss or gain of potential temperature and dW/dt corresponds to one component of dW^i/dt. Chemical reactions can be embodied into this description by a mixing model similar to a Poisson process [3].

CONCLUSIONS

Through Gaussian modeling an analytical relation between the emission rates and the distribution of pollutant concentration was determined, which can be inverted to estimate the emission rates of sources which are difficult to access. On this basis a concept for the optimization of concentration measurements was developed, which is explicitly suited to take advantage of path-integrated concentrations provided by optical remote sensing methods and which is applicable for cases with distributed sources. In particular, this optimization is aimed to the maximal partitioning of the measured concentration into contributions from different sources. Moreover, by applying a similar concept a configuration can be calculated which has the least sensitivity with respect to model parameter variations such as mean wind direction or dispersion parameters.

Because of the limits of Gaussian modeling these results can be viewed only as a first approximation for measurement planning. By Lagrangian modeling, more precise results can be obtained which are valid for a complex terrain, where inhomogeneities have to be considered and near sources. Starting from emission rates calculated by the Gaussian method these rates will be fitted by Lagrangian modeling to get the best approximation to the measured concentrations. This allows a determination of the limits of the Gaussian method.

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