Emissions of diffuse sources estimated by immission measurements

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ABSTRACT

Reduction strategies for the emissions of diffuse sources or measures for the stabilization of these inputs into atmosphere require monitoring techniques providing high-quality data of these emissions. Because in many cases these sources are difficult to access, an approach is considered combining optical remote sensing techniques for estimating path-integrated concentrations near sources with dispersion modelling. For this transport modelling to estimate emission rates of diffuse sources different methods are considered and compared. New concepts are shown for the optimization of measurement configurations.

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. By FTIR spectroscopy and a spectrum-analyzing programme developed by IFU concentrations can be measured of CO, CH_4, N_2O, CO_2, H_2O and near sources of NH_3, NO and NO_2. The relation between emission rate and measured concentration near sources can be calculated by dispersion modelling. This approach allows the estimation of emission rates of diffuse sources which cannot be estimated by other methods.

MODIFIED GAUSSIAN THEORY

The relation between emission rate and concentration in the measurement path can be calculated by applying modified Gaussian models [1, 2]. 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 [2]. 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

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

caused by the continuous source $S(x', y', z')$ [3]. 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 by which the concentration calculated for the emission of a point source is optimally matched to the results of tracer experiments [3]. Accordingly, one has

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

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

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 /sec. 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. Other kinds 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 $\langle c(x, y)\rangle$

$$\langle c(x,y)\rangle = \frac{s}{\pi u \sigma_y \sigma_z}\int_0^c dz' \exp\left\{-\frac{(z')^2}{2\sigma_z^{\;2}}\right\}\cdot$$
$$\cdot\int_{-a/2}^{a/2}\int_{-b/2}^{b/2} dx'dy' \exp\left\{-\frac{(y-y_0-\sin(\varphi)\cdot x'-\cos(\varphi)\cdot y')^2}{2\sigma_y^{\;2}}\right\}, \quad (3)$$

taking source extent and orientation into account.

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 it has to be 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 optimizing can be applied to calculate measurement configurations, where the influence of other sources is as small as possible [4, 5].
Considering different sources, the mean concentration $<c_\alpha(x, y)>$ caused by the source α can be written

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

where s_α denotes an emission rate in g / m^3 /sec corresponding with s in the above considered case of a volume ground source and $f_\alpha(x, y)$ 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 $<c(x, y)>$ of all contributions can be written

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

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

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

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

$$s_\alpha = F_\alpha^{-1}(x_m, y_m, \varphi_m, L)\left\{\int_{(c)} dt\;\langle c(x,y)\rangle + \sum_{\substack{\beta=1\\(\beta\neq\alpha)}}^{N} s_\beta F_\beta(x_m, y_m, \varphi_m, L)\right\}. \tag{8}$$

Then, the second term in (8) can be investigated in dependence on (x_m, y_m, φ_m, L) looking for minimal contributions in this area x-y, which is influenced by the considered emission. Denoting this area by E, this configuration (x_m, y_m, φ_m, L) has to be found, for which $(x_m, y_m) \in E$ and

$$F_\alpha^{-1}(x_m, y_m, \varphi_m, L) \sum_{\substack{\beta=1\\(\beta\neq\alpha)}}^{N} s_\beta F_\beta(x_m, y_m, \varphi_m, L) \;\rightarrow\; \text{minimum}. \tag{9}$$

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

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

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

LAGRANGIAN THEORY

More precise calculations of substance transport can be done using Lagrangian dispersion modelling [6-9]. 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 $\mathbf{x}_L(t)$, velocity $\mathbf{U}_L(t)$ (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 $\mathbf{U}_L(t)$ and $\Theta_L(t)$ to the 4-dimensional vector $\mathbf{Z}_L(t) = (\mathbf{U}_L(t), \Theta_L(t))$, these equations read

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

$$\frac{d}{dt} Z_L^i(t) = \langle a^i \rangle + G^{ij}(Z_L^j - \langle Z_E^j \rangle) + b^{ij}\frac{dW^j}{dt}, \tag{11b}$$

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 $<a^i>$ and G^{ij}, where the ensemble average is denoted by $<\cdots>$. The ensemble averages of Eulerian quantities (subscript E) are estimated at fixed positions $\mathbf{x}$ which are replaced by $\mathbf{x} = \mathbf{x}_L(t)$ 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, $<dW^i / dt> = 0$ and $<dW^i / dt\,(t) \cdot dW^j / dt\,(t')> = \delta_{ij}\, \delta(t - t')$, δ_{ij} denotes the Kronecker delta and $\delta(t - t')$ the delta function.
The coefficients appearing in (11b), $<a^i>$, 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 $B^{ij} = 1 / 2\, b^{ik} b^{kj}$,

$$B = \frac{1}{4\tau}\begin{pmatrix} C_0 q^2 & 0 & 0 & 0 \\ 0 & C_0 q^2 & 0 & 0 \\ 0 & 0 & C_0 q^2 & 0 \\ 0 & 0 & 0 & C_1 \langle (Z_E{}^4 - \langle Z_E{}^4 \rangle)^2 \rangle \end{pmatrix}, \tag{12}$$

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 $\langle a^i \rangle$

$$\langle a^i \rangle = \nu \frac{\partial^2 \langle Z_E{}^i \rangle}{\partial x^K \partial x^K} + (\alpha - \nu) \frac{\partial^2 \langle Z_E{}^4 \rangle}{\partial x^K \partial x^K} \delta_{i4} - \langle \rho \rangle^{-1} \frac{\partial \langle p \rangle}{\partial x^K} \delta_{Ki} - g\delta_{i3}. \tag{13}$$

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}

$$G^{ij} = -\frac{k_1}{4\tau}\delta_{ij} + \frac{k_1 - k_3}{2\tau}\delta_{i4}\delta_{j4} + \beta g \delta_{i3}\delta_{j4}. \tag{14}$$

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 [10]. 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.

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.

CONCLUSIONS

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 the measurement configuration and the assurance of data quality.

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