9B.6 PREDICTION OF HIGHER MOMENTS OF NEAR-SOURCE CONCENTRATION BY SIMULATING THE MEANDERING OF POLLUTANT PUFFS

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1. INTRODUCTION

When modeling pollutant transport and dispersion using gridded meteorological flow fields on an hourly basis, significant parts of the turbulence spectra are not resolved in space and time. Parameterizations of puff or plume dispersion commonly account for this by estimating one-hour averaged dispersion (so-called "absolute dispersion").
For risk assessments and odor impact analyses, the highest possibly occurring concentration during a time considerably shorter than one hour is more decisive than one-hour average values. For this, the probability density function of concentration for a given location and a specific averaging time is required.
To estimate concentration probability density functions, the absolute dispersion has to be split up into its two components, i. e. the instantaneous puff/plume growth ("relative dispersion") and the dispersion caused by meandering of the puff during the time averaging period. The first of these two components is driven by the turbulent eddies being smaller than the size of the puff, which are thus able to increase the mean distance between particles within the puff. The second component accounts for the effect of eddies larger than the puff; they displace the puff without enlarging it.
This contribution presents a method to estimate the higher moments of near-source concentrations for different averaging times, using the combination of the CALPUFF Lagrangian puff dispersion model with the Puff-Particle model (de Haan *et al.*, 1999). This combination improves the prediction of the highest occurring near-source concentration, as well as the distance down-wind from the source where it occurs, while retaining the advantages of puff models for the intermediate to far field range.

2. RELATIVE AND ABSOLUTE DISPERSION

Relative dispersion can be defined as the ensemble averaged separation between pollutant particles, i. e., the dispersion around the cluster's center. The well-known "absolute" dispersion, on the other hand, is the separation with respect to a fixed point in space, and is thus the sum of relative dispersion and meandering (Mikkelsen *et al.*, 1987; de Haan and Rotach, 1999).
The missing link between relative and absolute dispersion is the dispersing effect of meandering over timescales shorter than the averaging time for which the absolute dispersion parameterization used applies. Most formulations of absolute dispersion correspond to an ensemble averaging time of roughly one hour, corresponding to the spectral gap between synoptic and turbulent frequencies.

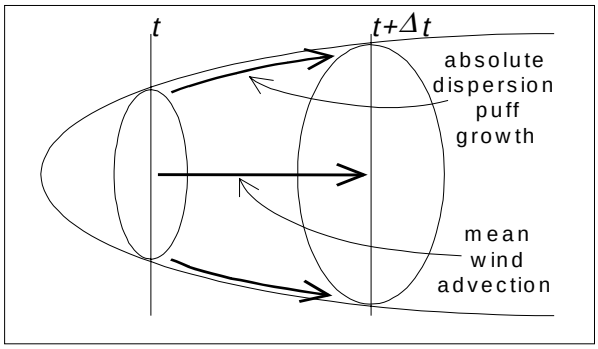

Fig. 1 Principle of plume segment models, using puffs as a segment of the ensemble averaged plume.

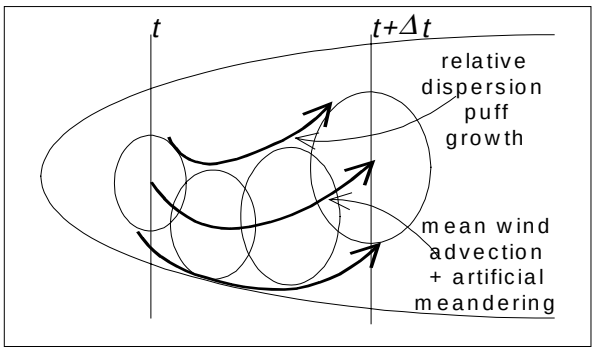

Fig. 2 Principle of cluster dispersion models, using puffs as clusters (clouds) of pollutants.

Hence, there are two different kinds of puff models. On the one hand, plume segment models (see Fig. 1), which describe a segment of an ensemble averaged plume by using a puff; their absolute dispersion scheme accounts for the ensemble averaged dispersion (from cluster growth and cluster meandering) during the averaging time *T* between two consecutive updates of the mean flow field. On the other hand, cluster dispersion models (see Fig. 2), which interpret a puff as being an instantaneous cluster of pollutants. The spectral gap between turbulent eddies covered by the flow field and those covered by the relative dispersion scheme is simulated with artificial meandering.
The CALPUFF model is a member of the first group, a plume segment model. Using parameterizations like

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the Pasquill-Gifford-Turner scheme, it yields an ensemble averaged dispersion. Such models generally assume that there is no gap between the averaging time of the measurements from which the dispersion scheme has been derived, and the time interval between two consecutive updates of the flow field within the model.

The PPM represents the second group of cluster dispersion models. It uses relative dispersion to describe the instantaneous growth of the cluster (puff); the range of turbulent eddies covered by the relative dispersion depends on the size of the cluster. Eddies larger than the cluster will displace the cluster as a whole without dispersing it. This meandering has to be simulated separately. In general, puff models from this second group will emit an higher amount of puffs to simulate the same release.

When computing ensemble averaged concentrations, both model approaches will yield similar results, though not identical: the meandering of puff centers allows for the advection of different parts of the ensemble average plume by the local flow. Especially near the ground, this will lead to differences in ground concentrations. For elevated releases, the meandering concept will cause some puffs to come close to the ground, where wind velocities are lower, and will thus lead to an somewhat earlier and steeper increase of the concentration profile in the plume center line.

Figure 3 compares the two concepts of puff dispersion. The most pronounced difference occurs for travel times of several minutes.

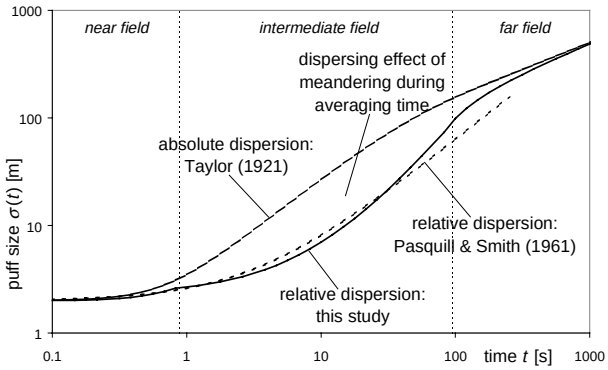

Fig. 3 Puff spread as a function of travel time *t*. The upper solid line is calculated from Taylor's single particle diffusion theory. Figure adapted from de Haan and Rotach (1999).

3. THE CALPUFF MODEL

The CALPUFF modeling system consists of several parts: The meteorological model CALMET, which is a diagnostic flow field model producing 3D flow, temperature and turbulence fields on an hourly basis, based on measurements. Currently, three different dispersion models are designed to use CALMET's output: CALPUFF, CALGRID, and KSP. CALGRID is a photochemical grid model (Yamartino *et al*., 1992). The Kinematic Simulation Particle model (KSP) (Yamartino *et al*., 1996) is using artificial turbulent eddies of different wave lengths to simulate turbulence.

The CALPUFF model (Scire *et al*., 1997) is a non-steady-state Lagrangian puff dispersion model for pollutant transport simulations under inhomogeneous and nonstationary conditions for periods of one year or more with a one-hour time step. Together with CALMET's flow fields, CALPUFF is applicable to complex terrain and coastal situations. The model uses so-called "absolute dispersion".

By its puff-based formulation, CALPUFF can account for a variety of effects such as spatial variability of meteorological conditions, causality effects, dry deposition and dispersion over a variety of spatially-varying land surfaces, plume fumigation, low wind speed dispersion, pollutant transformation, wet removal, and complex terrain effects. It has various algorithms for parameterizing dispersion processes, including the use of turbulence-based dispersion coefficients derived from similarity theory or observations.

CALPUFF has been developed with a broad range of applications in mind. One of the more important applications is estimating air quality impacts from electric generating and other industrial facilities. Such studies are used in determining compliance with ambient air quality standards or guidelines, or to quantify impacts associated with air quality, such as visibility effects and acid deposition.

Evaluation studies of CALPUFF have been done for long range transport distances (for the CAPTEX experiment; U.S. EPA, 1995), intermediate distances (Inel data; Irwin, 1998) and short to intermediate distances (SF6 data from Kincaid and SO2 data from Lovett power plants; Strimaitis *et al*., 1998).

4. THE PUFF-PARTICLE MODEL (PPM)

The PPM is a research model for near-source dispersion with separate treatments of puff meandering and real puff growth. It combines puff and particle dispersion models by moving the center of mass of each puff along a trajectory which mimics the quickly changing turbulent flow field. A full description of the PPM and the relative dispersion scheme used (depicted in Fig. 3) can be found in de Haan and Rotach (1999).

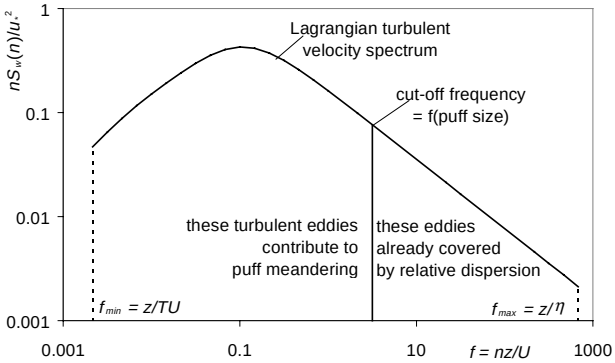

Fig. 4 Basic principle to compute stochastic movements of puff centers in the PPM.

The puff trajectories are derived from particle trajectories as simulated by a Lagrangian stochastic particle model. The particle model used within PPM is a full 3D Lagrangian stochastic particle model fulfilling Thomson's (1987) well-mixed condition. In order to avoid the double-counting of dispersion, only the low-frequency part of the trajectory is used by applying a low-pass Kalman filter (see de Haan and Rotach, 1998, for details). The principle of this method is depicted in Fig. 4.

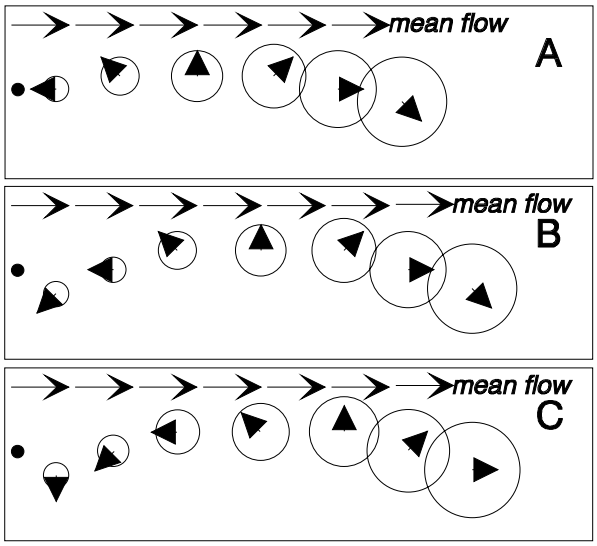

Fig. 5 Scheme to produce artificially meandering puff-plumes. The source indicated by solid black dot. For explanations of steps A to C, see text.

The meandering scheme used within the PPM is illustrated in Fig. 5 and consists of the following steps:
Step A shows the puff-plume at the beginning of the time-step. The solid triangles depict the stochastic velocity of the puff centers (2D in the figure, but 3D in the model), without the mean wind flow component.
Step B: after moving the puff-plume (i) with the mean flow and (ii) by the stochastic velocities, the newest puff of the puff-plume is released from the source. Its stochastic velocities are correlated with those of the second-newest puff. This is done by computing a puff trajectory starting with the turbulent velocities from the second-newest puff for a distance equal to the spatial separation from this second-last puff to the source.
Step C: The size of the puffs is enlarged using relative dispersion, and new stochastic velocity components are computed. The change of these components is correlated with the turbulent velocities of the next-newest puff (see Step B).

5. THE PPM AS CALPUFF-MODULE

The PPM is implemented as an option in CALPUFF to simulate plume meandering and to compute probability density functions of concentration. Because CALPUFF and PPM each belong to a different family of puff models (see section 2), no interference should occur between CALPUFF's puffs and those of PPM.

This is done by attaching so-called "mirror ensembles" of PPM puffs to every newly released CALPUFF "parent puff", which are advected in a "parallel universe" by the PPM-module. Whereas CALPUFF uses sampling steps ensuring that the puffs are advected within each grid cell on their path, the PPM has an internal time step between 1 and 10 s only. At the end of the CALPUFF basic time step, position and size of the "parent puff" are recomputed based on its mirror ensemble's moments. A full description of the combination of CALPUFF and PPM can be found in de Haan *et al.* (1999).

6. DETERMINATION OF CONCENTRATION PROBABILITY DENSITY FUNCTIONS

When assuming that the N puffs of the "mirror ensemble" of each "parent puff" carry one-N^{th} of the total mass each, the mean concentration for a specified averaging time should in principle be the same as CALPUFF's result. Interpreting the N mirror ensemble puffs as N possible realizations of the parent puff allows for the computation of the C-pdf (concentration probability density function).
Fig. 6 depicts cumulative C-pdf's for the Copenhagen tracer experiment (Gryning and Lyck, 1984) for N=1000 for an averaging time of 60 s. It illustrates the "narrowing" of the C-pdf as the release becomes better mixed further down wind, reducing the influence of meandering. Longer averaging times, too, would lead to a C-pdf with less pronounced tails. 60 s averages are considered representative for odor problems here. For the example of 1000 m down wind plume centerline concentrations, the chance that the "true" concentration will not exceed the average is 74%. In 5% of all cases, the actual concentration will be higher than 5 times the ensemble average.

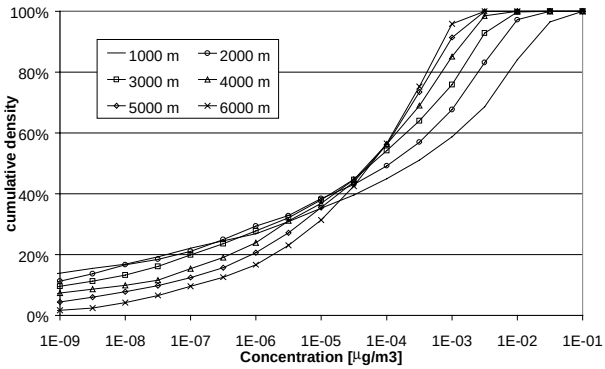

Fig. 6 Cumulative concentration pdf's for the plume center line position at six distances down wind from the source; averaging time 60 s. Release height 115 m, conditions of forced convection.

The precision of the form of the C-pdf at its lower and upper tails is related directly to N, and to the averaging time T. However, it should be kept in mind that the "highest occurring concentration" can only be specified for a given averaging time T. Choosing T shorter than, say, 30 s, tends to produce useless results (what's the concentration of a molecule?).

7. OTHER APPROACHES

The present contribution calculates concentration fluctuations with a plume meandering model. This can be applied to near-source cases, since the internal (i. e., within the puff) fluctuations are neglected; internal fluctuations become important when the release becomes approximately well-mixed in the vertical direction (Hanna 1984, 1986).

Savunen and Rantakrans (1999) present an odor model based on the assumption of a log-normal C-pdf. Its second moment is estimated as the standard deviation of the meandering motion or, when the plume becomes well-mixed, as the standard deviation of turbulent velocity.

Borgas (1998) and Wilson and Hildermann (1999) focus on the prediction of the highest occurring concentration caused by internal fluctuations. Borgas (1998) uses theoretical relative dispersion considerations to derive the higher moments of the C-pdf, whereas Wilson and Hildermann (1999) use a stochastic Markov process to emulate the time series of internal concentration fluctuations, when the moments of the C-pdf are supplied by any plume meandering model.

8. CONCLUSIONS

Hazardous gases and odor complaints show a highly non-linear relationship between their impact and the average concentration. The total dose or the peak concentration are more relevant. To assess such releases, short averaging times are needed as well as the higher moments of concentration, i. e., a C-pdf.

For this, the sub-averaging time and sub-grid meandering of the pollutant release has to be modeled separately from the dispersion itself. The present contribution shows a method to simulate the meandering, disperse the puffs with relative dispersion, and compute the C-pdf based on a large number of realizations from an ensemble. The approach is suited to calculate near-source C-pdf's for user-specified averaging times. For the estimation of far-field concentration fluctuations, the effect of internal fluctuations has to be taken into account as well.

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