Chapter 5

Prediction of Higher Moments of Near-Source Concentration by Simulating the Meandering of Pollutant Puffs*

Abstract—A method to estimate the higher moments of near-source concentrations for different averaging times is presented. Puff dispersion models using so-called „absolute dispersion“ predict one-hour-averaged concentrations. In order to be able to predict the higher moments of concentration (the probability density function), the „absolute dispersion“ has to be separated in its puff-growth part and the additional contribution from the meandering of the plume during the averaging time. The Puff-Particle Model (PPM) has been developed as such a puff-meandering model. The PPM aims at combining the advantages of both, 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 (artificial meandering). This trajectory is derived from the low-frequency part of trajectories as simulated by a Lagrangian stochastic particle model. These puff center trajectories are stochastically independent, which allows only for the evaluation of instantaneous concentration variances. A puff-plume meandering scheme is proposed which introduces spatial correlation between puffs which simulate a continuously emitting source. With this approach, the PPM can also be used to compute the probability density function for any other given averaging time. The PPM has been introduced as a sub-model into the CALPUFF Lagrangian puff dispersion model. The combination of CALPUFF and PPM improves the prediction of the highest possibly occurring near-source concentration, as well as the distance down-wind from the source where it occurs, while retaining the advantages of the CALPUFF model.

Key words: concentration pdf, puff models, plume meandering, relative dispersion.

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* this chapter is an enhanced compilation of two publications:

de Haan, P., and Scire, J. S. (1999): Prediction of higher moments of near-source concentration by simulating the meandering of pollutant puffs. Preprints 13th Conference on Boundary Layers and Turbulence, Jan. 10–15, 1999, Dallas TX. American Meteorological Society, Boston, USA

de Haan, P., Scire, J. S., Strimaitis, D. G, and Rotach, M. W. (1999): Introduction of a Puff-Particle approach for near-source dispersion into the CALPUFF model. In: Air Pollution Modeling and Its Application XIII, S.-E. Gryning and E. Batchvarova (eds.), Plenum Press, New York

5.1 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.

Within the field of dispersion modeling, puff models show a variety of advantages compared to Gaussian plume models. They can take into account the spatial variability of meteorological and dispersion conditions, causality effects, wet and dry deposition, low wind speed dispersion, etc. Lagrangian stochastic particle models, on the other hand, are the state-of-science of dispersion modeling, especially for the simulation of inhomogeneous (convective) turbulence. However, their demand of computing time generally limits their application to the simulation of short episodes rather than a full year. Furthermore, physical processes like dry and wet deposition, buoyant plume rise, and chemical transformations within a cluster, are much easier to implement within the framework of a puff model.

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.

Assessment of flammability or toxicity on the basis of ensemble-averaged concentrations can be seriously in error. These effects depend on short temporal- and spatial-scale fluctuations and thus the variance is essential for these predictions (Sykes 1988). Short temporal scale effects call for short (concentration) sampling times. This violates a basic assumption of many common air pollution models, since such averaging times are considerably shorter than the

spectral gap (approximately one hour). Most models assume that the sampling time is a time long enough to include most of the turbulent energy spectrum (Hanna 1982, p. 278).

In practice it is difficult to provide the flow field at a sufficiently high temporal rate to resolve all meandering motions, especially for small puffs. This means that the effect of meandering has to be simulated. For this, in the Puff-Particle Model (PPM) (de Haan and Rotach 1998a), a „cluster dispersion“ puff model using relative diffusion, the meandering of the puffs' centers of mass is generated artificially. These meandering trajectories simulate the effect of all those eddies not resolved by the flow field but still larger than the puff.

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 probability density function of concentration (denoted hereafter as C-pdf), based on Gifford (1959) and Hanna (1986). 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. Sykes *et al.* (1984) present a second-order turbulence closure scheme which accurately describes the near-field meandering and is able to predict the concentration variance along with the ensemble-mean concentration.

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.

Several models have been developed based on Gifford's (1959) meandering plume concept to separate the dispersing effect of instantaneous plume (or puff) growth and of meandering (e.g. Hanna 1984, 1986; Savunen and Rantakrans 1997; Sykes 1988). They all have in common that a certain shape of the probability density function of concentration is assumed and that the variance of concentration fluctuations is derived from the properties of turbulence. SCIPUFF (Sykes 1997) uses a second-order turbulence closure scheme predicting both the

mean concentration and its variance. In contrast, the PPM aims at simulating a realistic three-dimensional meandering of the puffs, which allows for the direct determination of the higher moments of concentration, while enabling the individual puffs to account for inhomogeneous conditions.

In the next section, the concepts of absolute and relative dispersion are used to distinguish between two different so-called puff model families. The basic philosophy and major features of the CALPUFF and PPM puff models are summarized (sections 5.3 and 5.4). The puff-plume meandering scheme, which is proposed to simulate the meandering behavior of a continuously emitting source within the PPM, is introduced in section 5.5. The introduction of the PPM as a sub-model within CALPUFF is described in section 5.6. By interpreting any single meandering puff trajectory as a possible realization, a C-pdf can be computed, which is done for a tracer experiment (section 5.7). Finally. the form of such concentration pdf's for different down-wind distances and averaging times is discussed (section 5.8).

5.2 RELATIVE AND ABSOLUTE DISPERSION

The well-known "absolute" dispersion (so-called 1-particle statistics, or ensemble averaged dispersion), is the ensemble-averaged particle separation with respect to a fixed point in space (Taylor 1921). Richardson (1926) was the first to introduce the concept of relative dispersion, defined as the ensemble averaged separation between pollutant particles released together, i.e. the dispersion around the cluster's center. Gifford (1957a, 1957b) first pointed out that the instantaneous growth of puffs is governed by the concept of relative instead of absolute dispersion. Within the concept of relative dispersion (i.e. 2-particle statistics), turbulent eddies smaller than the actual puff size will contribute to its growth (increasing the ensemble 2-particle separation), while larger eddies move the puff as a whole (without changing the separation of the particles). Thus absolute dispersion is the sum of relative dispersion and meandering during the averaging time (Mikkelsen *et al*. 1987; de Haan and Rotach 1998a).

The missing link between relative and absolute dispersion is the dispersing effect of meandering over time-scales 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.

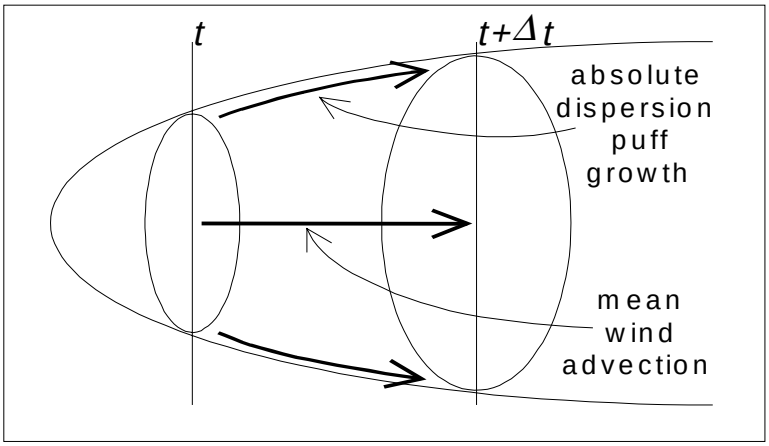

Figure 1 Principle of plume segment models, using puffs as a segment of the ensemble averaged plume.

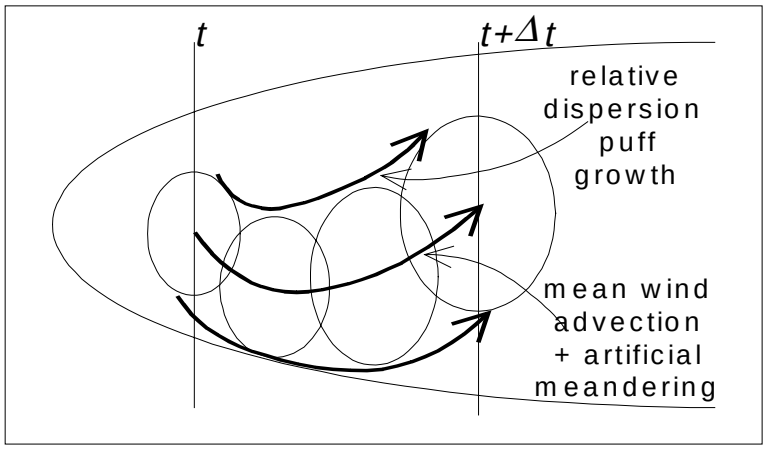

Figure 2 Principle of cluster dispersion models, using puffs as clusters (clouds) of pollutants.

Hence, there are two different kinds of puff models: On the one hand, plume segment models (see **Figure 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, there are cluster dispersion models (see **Figure 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.

Within the framework of these ‘plume segment’ puff models, a puff (consisting of a center of mass, the actual velocity of the puff, and a 3D distribution of the total mass around the center)

stands for the ensemble average of the concentration distribution belonging to a „piece“ (in time) of the pollutant release. These puffs do not correspond to a single cluster in nature, but are an ensemble average over many individual clusters. 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.

‘Cluster dispersion’ puff models identify the puff with an individual, physically realistic cluster of particles. They use the concept of relative dispersion (i. e. 2-particle statistics; Gifford 1957a,b; Borgas and Sawford 1994; Thomson 1990): turbulent eddies smaller than the actual puff size will contribute to its growth (increasing the ensemble 2-particle separation), while larger eddies move the puff as a whole (without changing the separation of the particles). Hence, absolute dispersion is the combined effect of relative diffusion and of the meandering of the puff respective to a fixed point, caused by turbulent eddies larger than the puff.

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.

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. For small (newly released) puffs, absolute dispersion is dominated by meandering rather than by puff-growth. Therefore, in contrast to „cluster dispersion“ models, „plume segment“ puff models are not suited to give correct concentration predictions for a single (instantaneous) release, especially in the intermediate field.

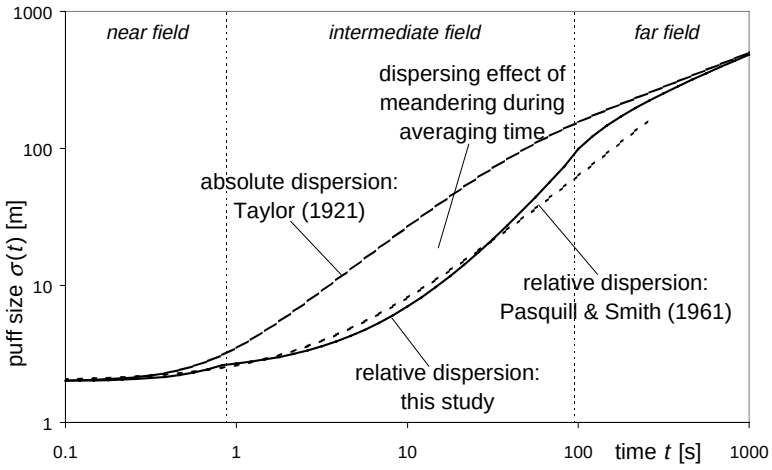

Figure 3 Puff spread as a function of travel time *t*. The upper, long-dashed line is calculated from Taylor's single particle diffusion theory. Figure adapted from de Haan and Rotach (1998a).

5.3 THE CALPUFF MODEL

The CALPUFF model (Scire *et al.* 1995, 1997) is a non-steady-state Lagrangian puff dispersion model for pollutant transport simulations under inhomogeneous and non-stationary conditions for periods of one year or more with a one-hour time step. Among its main fields of application are pollutant transport simulations for inhomogeneous and non-stationary conditions. CALPUFF is a member of the 'plume segment' puff model family. Using parameterizations like the Pasquill-Gifford-Turner scheme, it yields an ensemble averaged dispersion (i.e. 'absolute dispersion', cf. section 5.2). Together with the flow fields of its meteorological model, CALMET, CALPUFF is applicable to complex terrain and coastal situations.

The user-defined grid size allows for high-resolution simulation of episodes as well as for runs of one year or more with a one-hour time step for environmental impact assessments, and studies of air quality and pollutant transport on regional scales. By its puff-based formulation, it can account for a variety of effects such as spatial variability of meteorological and dispersion conditions, causality effects, dry deposition, 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.

The CALPUFF modeling system consists of several parts: The meteorological model CALMET, which is a diagnostic flow field model producing mass consistent and diagnostic

3D flow, temperature and turbulence fields on an hourly basis, based on measurements. Currently, three different dispersion models are designed to use the output of CALMET: 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.

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 SO_2 data from Lovett power plants; Strimaitis *et al.* 1998).

5.4 THE PUFF-PARTICLE MODEL (PPM)

The fact that instantaneous releases require puff models using relative dispersion, but that at the same time, the update frequency of the flow field information in almost all applications is too low to resolve all those turbulent eddies not covered anymore by the relative dispersion concept, gave rise to the development of the Puff-Particle Model (PPM). It represents the group of so-called „cluster dispersion“ puff models. The PPM in its current version is a research model for tracer pollutants, focusing on near-source dispersion, and neglecting deposition and chemical processes. It features a full stochastic Lagrangian particle dispersion model, which fulfills the well-mixed criterion (Thomson 1987). For convective conditions, the vertical component of the pdf is the same as in Luhar and Britter (1989). To provide a perfectly smooth transition between stable/neutral Gaussian turbulence to convective skewed turbulence, the transition function of Rotach *et al.* (1996) has been adopted. Further details on the PPM can be found in de Haan and Rotach (1998a).

Every puff within the PPM follows a turbulent puff trajectory derived from a stochastic particle trajectory. The kinematic turbulent energy belonging to those eddies which are smaller than the puff’s size-already covered within the concept of relative dispersion-is removed from the particle trajectories. For this, a Kalman low-frequency filter is used, where the cut-off frequency depends on the size of the puff (de Haan and Rotach 1998b). Hence, every puff carries along its own position as well as the position and turbulent velocity components of the stochastic particle it ‘belongs to’. The effect of meandering (caused by turbulent eddies larger than the puff but not resolved by the flow field) is simulated by the

puff center trajectories, yielding a complete description of dispersion. It has been shown on the basis of tracer data, that the correct treatment and interpretation of the two contributions to the dispersion process is crucial for reproducing experimental results to a good correspondence (de Haan and Rotach 1998a).

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, 1998b, for details). The principle of this method is depicted in **Figure 4**.

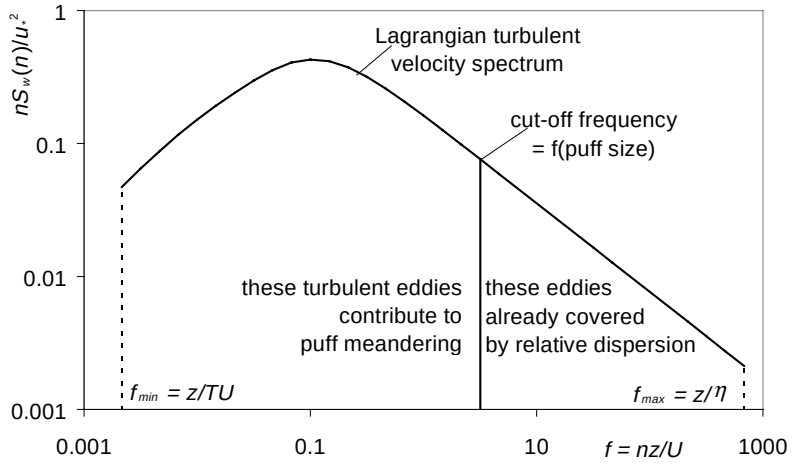

Figure 4 Basic principle to compute stochastic movements of puff centers in the PPM.

5.5 THE PUFF-PLUME MEANDERING SCHEME

Gifford's (1959) meandering plume dispersion model neglects dispersion in the direction of the mean wind, leading to a 'spreading disk' plume dispersion model. The mean concentration distribution as predicted by Gifford's (1959) model is identical with predictions from ensemble-averaged plume models. Additionaly, it predicts statistical properties like the variance of point concentrations. The 'split' between instantaneous plume growth and dispersion due to meandering is a function of down-wind distance, i.e. is different for each 'disk' of the fluctuating plume. These 'disks' do not actually move; it is the statistical property of their movement that is predicted. From this it follows that the statistical properties of concentration as predicted by the Gifford (1959) approach apply to instantaneous (point) concentrations, i.e. with zero averaging time.

For a non-zero, arbitrary concentration averaging time, however, the correlation of the meandering movements between two neighboring 'disks', or puffs, has to be taken into

account. Even though the statistical properties of concentration fluctuations for each point are correct, the statistical properties of concentration *averages* over time differ from non-correlated to correlated meandering. Sykes (1984) and Sykes and Gabruk (1997) present an extension to the Gifford (1959) model, introducing an autocorrelation function for concentration fluctuations. This allows for the computation of the influence of averaging time on the concentration variances.

Within the PPM, the puffs use stochastic paths to artificially produce the correct meandering behavior. This way, the ensemble mean concentration together with its higher moments can be computed for a puff release, for any user-specified averaging (i.e. sampling) time. To obtain correct C-pdf's for continuous plume releases, however, these stochastic puff meandering trajectories are not sufficient. They are derived from Lagrangian particle trajectories which are stochastically independent. But 'neighbor' puffs (i.e. subsequently released puffs) should show 'similar' meandering: the spatial and temporal correlation of turbulence has to be taken into account. If this is neglected, the most extreme concentration events will be underestimated, and the C-pdf will not be correct.

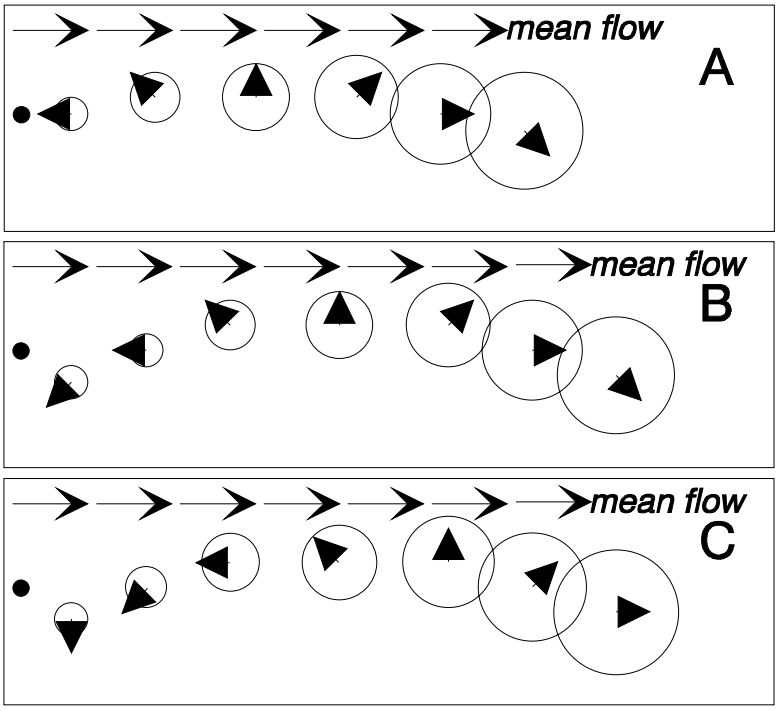

Figure 5 Scheme to produce artificially meandering puff-plumes. The source location is indicated by a solid black dot. The solid triangles depict the stochastic velocity of the puff centers (2D in the figure, but 3D in the model), without the mean wind flow component. For explanations of steps A to C, see text.

The PPM employs a plume meandering scheme based on the puff meandering scheme. Plumes are described as threads of puffs which are correlated in their turbulent movements to their next neighbors in the puff-plume. The PPM puff-plume meandering scheme is illustrated in **Figure 5** and consists of three steps:

- *Step A:* At the beginning of the time-step, each puff has its initial 3D stochastic velocity components.
- *Step B:* After moving the puff-plume with the mean flow and by the stochastic velocities, the newest puff is released. Its stochastic velocities are correlated with those of the second-newest puff (its „mother“ puff) by copying the mother's turbulent velocities and computing a puff trajectory over a time which corresponds 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 (again, by computing a puff trajectory starting with the turbulent velocities from the „mother“ puff). The first (most distant from the source) puff of the puff-plume follows a normal (non-correlated) PPM puff-center trajectory.

This meandering scheme can be illustrated by thinking of a spectrum of turbulent eddies „rolling back“ towards the source along the puff-plume with the average mean wind speed (introducing the spatial correlation). Such a spectrum of eddies is „released towards the origin“ from the front of the plume every time step, based on the puff path of the front puff (temporal correlation). **Figure 6** and 7 show examples of such meandering puff-plumes.

The Lagrangian particle model within the PPM yields a Lagrangian spectrum, when the turbulent velocity of a particle is followed over time. It has the correct „–1“ slope of the inertial subrange, in contrast with the „–2/3“ slope required for Eulerian spectra. The current approach will thus give a „–1“ instead of a „–2/3“-inertial subrange slope when considering the turbulence at a fixed point in space. However, since these spectra are filtered and only their low-frequency part is used, this shortcoming has only a minor influence. The main difference between Eulerian and Lagrangian spectra is the location of the maximum of the spectrum, $f_{max,E}$ and $f_{max,L}$, respectively. This is corrected by shifting the Lagrangian spectrum towards higher frequencies by a factor $\beta = f_{max,E} / f_{max,L}$. So the turbulent velocities of a puff with a separation x to its „mother“ are computed as a puff trajectory over a time $\beta \cdot x/U$ rather than x/U.

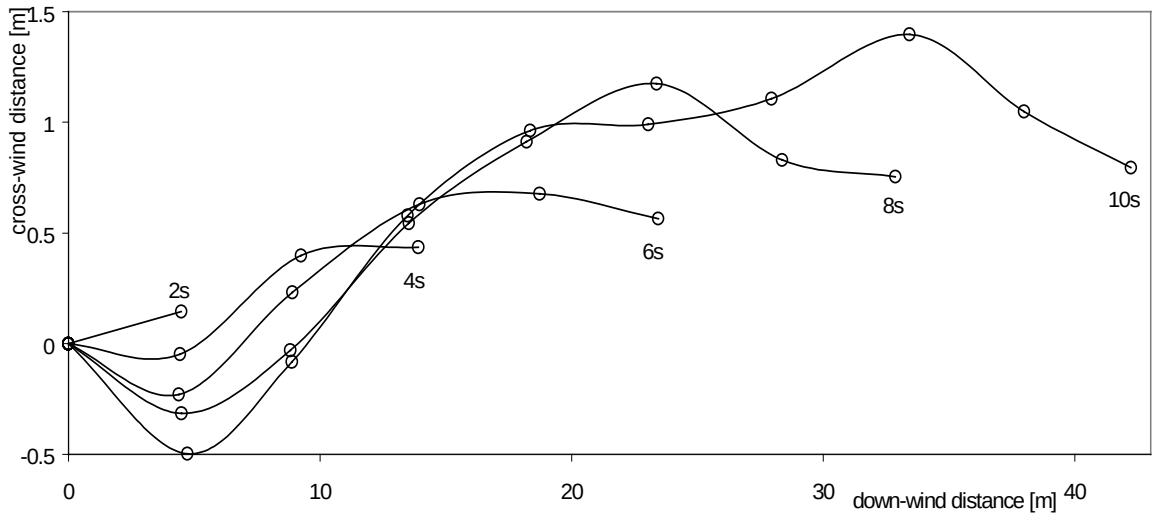

Figure 6 Illustration of the meandering puff-plume scheme. The released puff-plume is depicted 2, 4, 6, 8 and 10 seconds after its release. Puffs belonging to the same puff-plume are connected by a thin line. Circles indicate the position of the center of each puff (not the puff size). View from above.

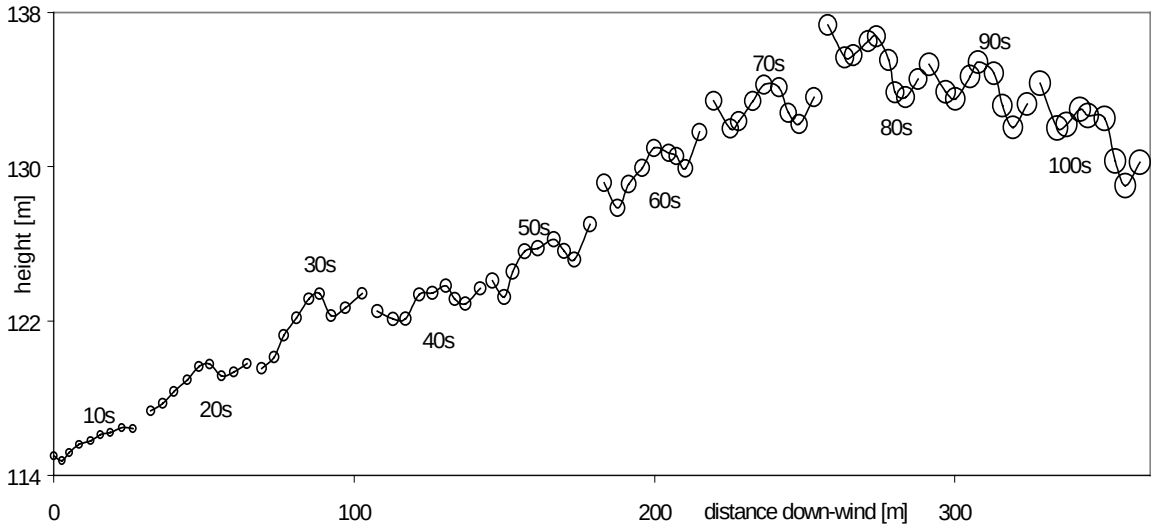

Figure 7 Possible evolution of a plume (consisting of 9 puffs released with a one second interval) over time, when using PPM's meandering puff-plume scheme. The position of the plume's puffs is shown every ten seconds. Example for convective conditions, release height 115 m. View from the side.

5.6 INTRODUCTION OF THE PUFF-PARTICLE MODULE INTO CALPUFF

The CALPUFF model has been developed for a broad range of applications, from micro-scale to meso-scale, from short-term to long-term, with treatments of wet and dry deposition, chemistry, coastal interactions, complex terrain, visibility impacts, etc. The PPM will be one

additional option in this list, providing additional information on the near-source concentration moments. Due to the flexibility of the CALPUFF model on the side of the user, its code has reached a high grade of complexity. Therefore, one of the main objectives for the implementation of the Puff-Particle module into CALPUFF was to introduce as few interactions between the CALPUFF code and the PPM option as possible, in keeping with the module coding concept. Thus, the underlying thought of the concept adopted was that CALPUFF's puff treatment will not change; it is only within the Puff-Particle module that the simulation of artificial meandering of relatively dispersed puffs is computed.

Because CALPUFF and PPM each belong to a different family of puff models (see section 5.2), 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.

Figure 8 illustrates the basic set-up of the PPM within CALPUFF:

- To every newly released puff, a so-called 'mirror ensemble' is attached. Such a mirror ensemble consists of a user-defined number, *N*, of puff-particles.
- In order to take into account the information from all grid cells through which the puff will pass in one hour, CALPUFF divides the one hour model time step into a varying number of sampling steps for each puff. For the duration of the sampling step of the parent puff within CALPUFF's main routine, the mirror-ensemble is advected with an internal user-defined PPM-timestep between, say, 1 and 10 seconds. For every internal PPM-timestep, new particle trajectories are computed, from which puff trajectories are derived. Additionally, the puff-particles are advected by the mean flow.
- At the end of the sampling step, from the mirror ensemble's first and second moments of the mass distribution, the position and size of the parent puff are calculated and handed back to CALPUFF's main routine. CALPUFF may then compute any physical process possibly changing the (parent) puff's mass or chemical composition, but not its size or location. The mirror ensemble remains in existence for use in the next sampling step.

- After a certain time, the size of the relatively dispersed particle-puffs in the mirror ensemble will be such that the largest part of the energy spectrum of turbulent eddies will be 'within' the puff-particle. This means that the relative and absolute dispersion for that ensemble become similar. The artificial meandering will show only little variation of the particle-puff's paths. Then, the 'parent puff' location and size is recomputed, the mirror ensemble deleted and the parent puff restored, which from then on is treated with the common absolute dispersion within the CALPUFF model.

The initial position of the particle-puffs at the time of creation of a mirror ensemble is taken randomly from a three-dimensional Gaussian distribution with mean at the center of the parent puff, and the standard deviations of the parent puff being the second moments of the Gaussian distribution. Analogously, the initial turbulent velocity components of the puff-particles of the mirror ensemble are taken from a three-dimensional Gaussian distribution with zero mean and with the ambient turbulence $\left(\overline{u_i'^2}, i = 1,2,3\right)$ as the second moments.

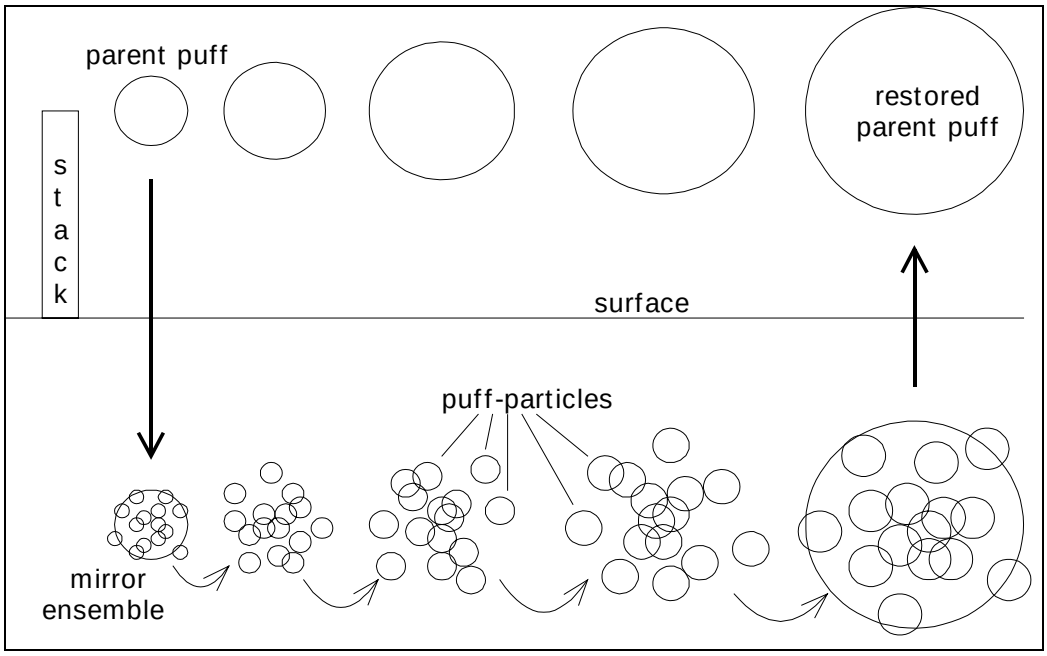

Figure 8 Illustration of the embeddement of the PPM into the CALPUFF model. The procedure for one sampling time step is depicted. After 'restoring' the new location and size of the parent puff, the mirror-ensemble remains stored for use at the beginning of the next sampling step of the same puff, until the puff reaches a mature stage where the mirror-ensemble will be deleted.

5. 7 DETERMINATION OF CONCENTRATION PROBABILITY DENSITY FUNCTIONS

The puff-particle approach gives a realistic picture of the transport (caused by the mean wind, provided by the flow field updates), the amount of meandering (covered by the stochastic puff center trajectories) and the diffusion of the release (caused by eddies smaller than the puff, and taken into account by relative diffusion). For each parent puff, an ensemble of mirror puff-particles is released. Since the meandering part of their motion is reigned by random particle motion, their trajectories will not be identical. Each of them is a possible realization of what might have happened to the parent puff in reality.

To obtain the mean (ensemble averaged) concentration at any receptor, the average over all mirror puff-particles can be taken, where each realization is assumed to represent 1/*N* of the total mass of the parent puff. In theory, the ensemble average concentration thus obtained should be close to the concentration estimation obtained by using the parent puff only, together with absolute dispersion. Of course, in practice, minor differences will appear. To let these differences vanish, it would be necessary to use the same set of parameterized spectra of turbulent energy as a foundation of the absolute as well as the relative dispersion parameterization and at the same time derive the particle pdf within PPM's particle model from this set of spectra.

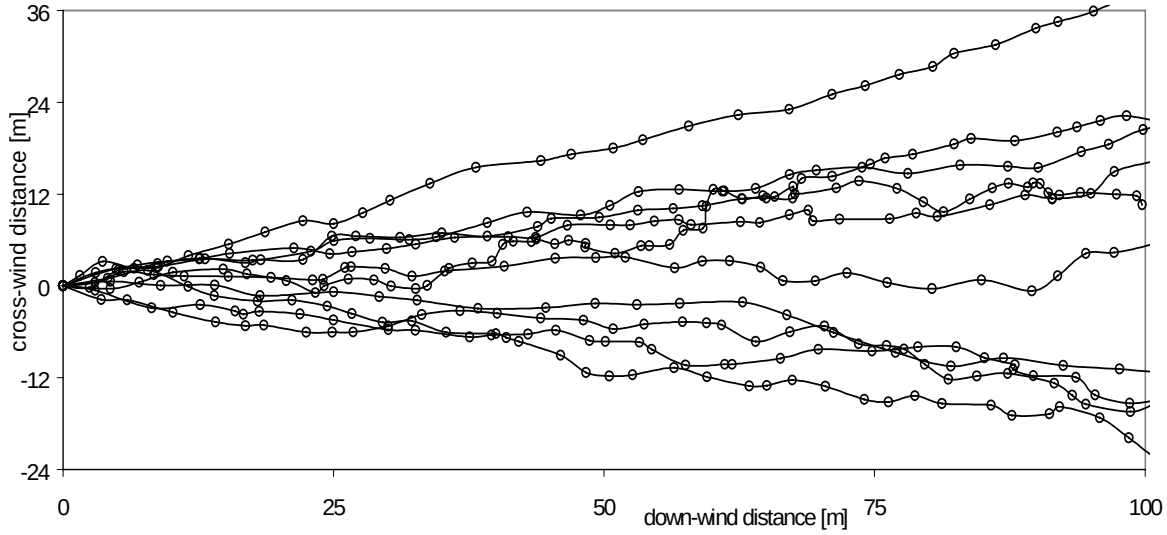

Figure 9 Method to construct a C-pdf. Many individual meandering puff-plumes are simulated. Each is treated as one possible realization that could actually have happened for the given meteorological data. The sorted concentration impacts (for a user-determined averaging time) at specified receptor locations from these many realizations constitute the C-pdf.

To obtain the pdf of concentration at such a receptor, every realization (i. e. mirror puff-particle) carries the whole mass of the parent puff, and the concentration due to this particular realization is computed (**Figure 9**). This leads to *N* possible concentrations, from which the concentration pdf easily can be derived. Of course, to make the concentrations thus computed comparable to the standard CALPUFF results, these *N* possible concentrations have to be computed using all puffs in the modeling domain impacting the receptor, whether they possess a mirror-ensemble or not. In the intermediate to far field, this will lead to a narrowing of the concentration pdf, reflecting the fact that well dispersed pollutant clusters will indeed lead to a higher degree of determination of the corresponding concentration. Therefore, the PPM option within CALPUFF will produce valuable results mainly in the near field, where the differences in puff position due to meandering potentially are of the same order of magnitude as the size of the puff.

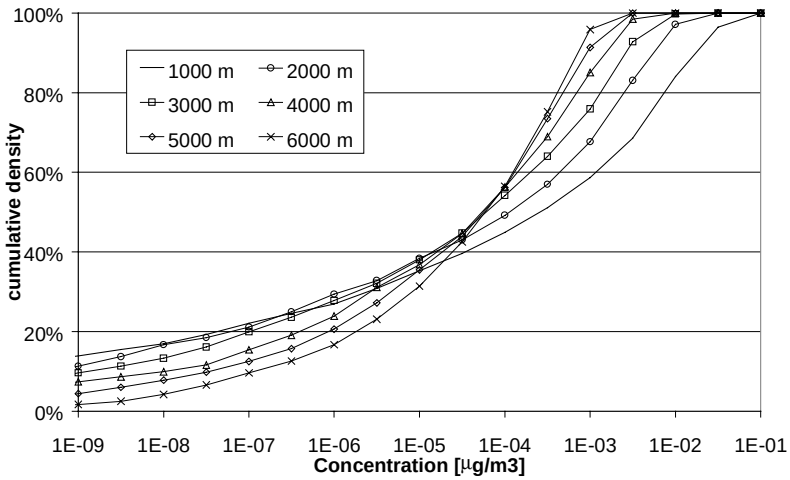

Figure 10 Cumulative concentration pdf's for the plume center line position at six distances down wind from the source; averaging time 60 s. Release height 115 m, conditions of forced convection.

As a first example of the application of PPM's puff-plume meandering scheme, Figure 10 depicts cumulative C-pdf's for the Copenhagen tracer experiment (Gryning and Lyck 1984) for $N = 1000$ for an averaging time of 60 s. 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.

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, will call for a drastic increase in *N* and thus in computational resources, in order to avoid that extreme events become ill-defined because only very few puffs will impact on the receptor location during such short averaging times.

5.8 CHARACTERISTICS OF CONCENTRATION PDF'S

Observations (Fackrell and Robins 1982) show that the most pronounced plume concentration fluctuations are produced very near to the source, and the form of the pdf changes from a near-source exponential to a intermediate-field normal distribution. Despite these forms, the ratio of peak to ensemble-averaged concentrations seems to remain constant. The relative amount of meandering (with respect to ensemble-averages dispersion) depends on atmospheric stability. For stable (night-time) conditions, for example, Hanna (1983) observed very high lateral plume meandering.

When looking at the changes in the form of the C-pdf for increasing down-wind distances (**Figure 11**), two effects are occurring simultaneously. First, a shift of the mean (and the maximum) to the left, i. e. to lower ensemble average concentrations. Second, a narrowing of the C-pdf: the slopes to the left and to the right of the maximum are getting steeper. This means that the ensemble average becomes „better defined", due to the fact that the relevant time scales of fluctuation at these distances are much larger than the averaging time.

The form of the C-pdf also is a function of the averaging time *T*. Whereas a short averaging time of 60 s leads to a wide-spread C-pdf, the C-pdf narrows for larger averaging times (without changing its mean, i. e. ensemble concentration). Additionally, C-pdf's for stable conditions are wider than for convective conditions, because more dispersion leads to a more well-defined ensemble mean, hence a narrowing of the C-pdf.

The form of the C-pdf often is assumed to be log-normal. For most models predicting concentration fluctuation probabilities, this is an input rather than a result. The results from the present model, however, suggest that the C-pdf is log-skewed towards lower concentrations.

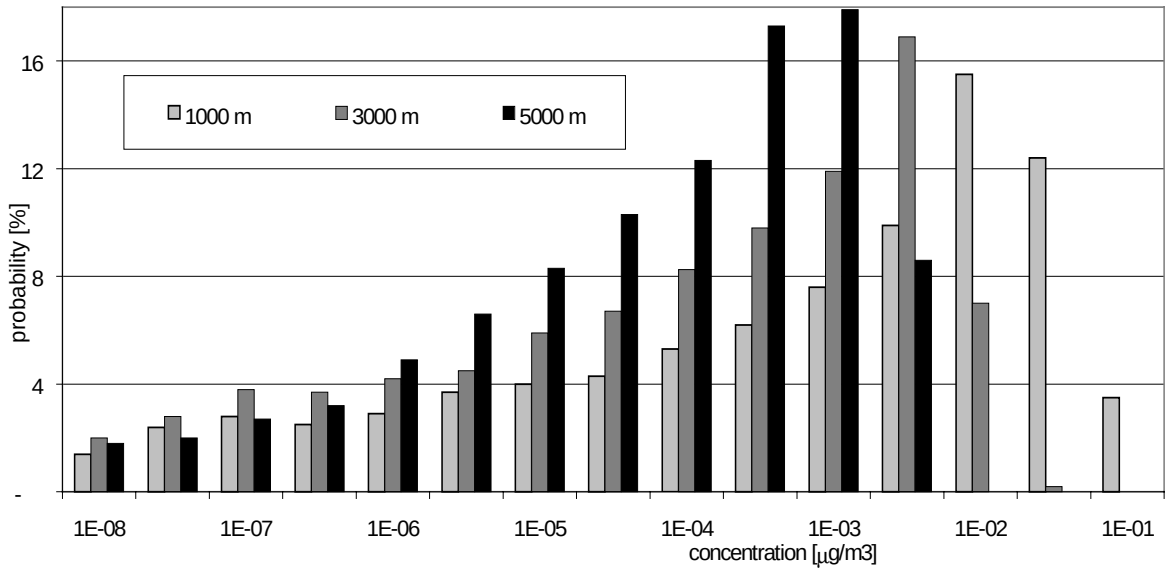

Figure 11 Example of C-pdf's for the plume centerline position at three distances down-wind from the source; averaging time 60 s. Example for the Copenhagen experiment (release height 115 m, conditions of forced convection).

5.9 SUMMARY AND 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. Using the PPM, the present contribution shows a method to simulate the meandering of puffs while dispersing the puffs with relative dispersion. For every emitted pollutant puff, an ensemble of puffs is simulated within the PPM. Every individual meandering puff path is interpreted as one possible realization of the dispersing, meandering and transport of the pollutant puff. The C-pdf then is based on the estimated concentration impacts of the individual realizations from the ensemble.

The particle model incorporated within the PPM, upon which the puff meandering is based, assumes that the particle trajectories be stochastically independent. This allows for the computation of concentration variances for an instantaneous (i.e., puff) release. For any continuous release, however, the correlation between subsequently emitted puffs has to be

taken into account. In order to correctly mimic this spatial and temporal correlation, a puff-plume meandering scheme has been introduced.

The puff- and plume-meandering approach is suited to calculate near-source C-pdf's for any user-specified averaging time. For the estimation of far-field concentration fluctuations, the effect of internal fluctuations has to be taken into account as well. This way, the PPM is especially suited for the simulation of accidental hazardous releases. The artificial meandering scheme allows identifying worst-case scenarios.

The combination of the PPM with an operational, user-friendly Lagrangian puff model, CALPUFF, which uses a different approach to describe the dispersion of pollutants, allows for the combination of the advantages of the CALPUFF model, with additional information on the higher moments of the concentration distribution at near-field receptors from the PPM module. This allows for an additional assessment of concentrations that actually could be observed at a given receptor, together with the corresponding probability. This additional feature is particularly useful for the assessment of hazardous pollutants, where the question of the ensemble-averaged exposure is less relevant than the highest exposure which could actually occur under the given meteorological conditions.

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