A DENSITY CORRECTION FOR LAGRANGIAN PARTICLE DISPERSION MODELS

ANDREAS STOHL
Lehrstuhl für Bioklimatologie und Immissionsforschung, Ludwig-Maximilians-Universität München, Am Hochanger 13, 85354 Freising-Weihenstephan, Germany

DAVID J. THOMSON
Meteorological Office, London Road, Bracknell, Berkshire, RG12 2SZ, U.K.

(Received in final form 17 July 1998)

Abstract. Current Lagrangian particle dispersion models, used to simulate the dispersion of passive tracers in the turbulent planetary boundary layer (PBL), assume that the density is constant within the PBL. In deep PBLs, where the density at the boundary-layer top may be lower by more than 20% than at the surface, this assumption leads to errors in the tracer concentrations on the order of 10%. In the presence of a vertical wind shear, this also leads to inaccurate calculations of the horizontal tracer transport. To remove this deficiency, a Langevin equation is presented that contains a density correction term. The effect of the density correction is studied using data from a large-scale tracer experiment. It is found that for this experiment, the main effect of the density correction is an increase in the surface tracer concentrations, whereas the horizontal tracer transport patterns remain largely unaffected.

Keywords: Density correction, Dispersion models, Lagrangian models, Particle models.

1. Introduction

Lagrangian particle dispersion (LPD) models are often used to numerically simulate the dispersion of a passive tracer in the planetary boundary layer (PBL) by calculating the Lagrangian trajectories (Stohl, 1998) of thousands of ‘marked particles’ (Rodean, 1996). With these models, the velocity and position of a particle are assumed to evolve as a Markov process (Wilson and Sawford, 1996) according to the Langevin equation (Thomson, 1987)

$$dv_i = a_i(\mathbf{x}, \mathbf{v}, t)dt + b_{ij}(\mathbf{x}, \mathbf{v}, t)dW_j, \tag{1}$$

where a and b are functions of the position $\mathbf{x}$, the velocity $\mathbf{v}$ and time t. The dW_j are incremental components of a Wiener process with mean zero and variance dt, each component being independent of the other components and uncorrelated in time. The particle positions are calculated from

$$dx_i = v_i dt. \tag{2}$$

Boundary-Layer Meteorology **90:** 155–167, 1999.

The key problem of LPD models is to determine the functions *a* and *b*, known as the drift and diffusion terms respectively, for a particular turbulent flow field for which the Eulerian flow statistics are given (Wilson and Sawford, 1996). Criteria for their determination were given by Thomson (1987), the most general being that particles that are initially well mixed in position and velocity space in a turbulent flow must remain that way (the so-called well-mixed criterion).

In this study, the focus is on the determination of the drift term *a*, which is usually chosen so that well-mixed particles remain well-mixed in a Cartesian coordinate system. Unfortunately, such a model does not account for inhomogeneities in the density of the air. This may be justified in a shallow PBL, but it certainly is inaccurate in a deep convective mixed layer (of order 2000 to 3000 m), where the density may be lower at the top than at the bottom by more than 20%. If an LPD model neglects such density variations, it will systematically under-predict surface concentrations and over-predict concentrations near the top of the PBL. If the air densities at the bottom and at the top differ by 20%, respective over- and under-predictions will be some 10%, a fact recently also noted by Venkatram (1998). In the presence of a vertical wind shear, this will also lead to inaccurate calculations of the horizontal transport.

It seems that a trivial solution to this problem would be to use a normal LPD model and interpret its results in terms of mixing ratio. However, such an approach would not conserve mass, as a particle that moves upward, to an environment with lower density, would lose mass, whereas a particle moving downward would gain mass. Since mass conservation is an important constraint, this is unacceptable and a different solution must be found.

In the next section, we introduce a Langevin equation model that contains a density correction term. By comparing this model against calculations without density correction, we study the importance of vertical density gradients (Section 3). Finally, in Section 4, we draw some conclusions.

2. Model Description

For our investigations, we used the LPD model FLEXPART, version 3.0. FLEXPART was developed by Stohl (1996) and was extensively evaluated by Stohl et al. (1998) against large-scale tracer experiment data. It evolved from the trajectory model FLEXTRA (Stohl et al., 1995; Baumann and Stohl, 1997; Stohl and Seibert, 1997) and is based on data of the numerical weather prediction model of the European Centre for Medium-Range Weather Forecasts (ECMWF, 1995). Since the model was described by Stohl et al. (1998), only the aspects important for this paper are addressed here.

The model solves Langevin equations for the three wind velocity components. The three components evolve independently, apart from linkages caused by the fact that the coefficients in the equations depend on the particle position. This involves neglecting the cross correlations, which has only minor effects on the results of large-scale dispersion models (Uliasz, 1994). Gaussian turbulence is assumed under all meteorological conditions, and the turbulent statistics are obtained using the scheme of Hanna (1982) with some modifications for convective conditions.

Our main interest here is in the equation for the vertical motion. As is usual in models of the type considered here, this is derived under the assumption that changes of the flow properties in the horizontal and in time are sufficiently slow to be neglected. The equation is derived by applying the constraint introduced in Section 1 above, namely that if the particles are well-mixed in the flow (i.e., if the joint distribution of the heights z and vertical velocities w of the particles matches that assumed for all the particles of air) then they should remain so. Let us introduce $g(z, w)$ for the density of the distribution of the z and w values of the tracer particles and $g_a(z, w)$ for the equivalent quantity for all particles of air. With the assumption of Gaussian velocity statistics, g_a is given by

$$g_a \propto \rho \frac{1}{\sqrt{2\pi}\sigma_w} \exp\left(-\frac{w^2}{2\sigma_w^2}\right)$$

where σ_w is the standard deviation of w and ρ is the density of the air, both of which are functions of the height z. We have written $\propto$ here rather than = because the normalisation of this distribution is irrelevant here. Now, for models of the form

$$\begin{cases} \mathrm{d}w = a(z, w)\mathrm{d}t + b(z, w)\mathrm{d}W \\ \mathrm{d}z = w\mathrm{d}t \end{cases}$$

the distribution of tracer g evolves according to

$$\frac{\partial g}{\partial t} = -\frac{\partial}{\partial z}(wg) - \frac{\partial}{\partial w}(ag) + \frac{\partial^2}{\partial w^2}\left(\frac{1}{2}b^2 g\right)$$

(this is a standard result in the theory of stochastic differential equations and is discussed in the LPD context by e.g., Thomson, 1987; and Wilson and Sawford, 1996). The 'well-mixed condition' then means that g_a should also satisfy this equation, leading to

$$a g_a = \frac{\partial}{\partial w}\left(\frac{1}{2}b^2 g_a\right) - \int_{-\infty}^{w} \frac{\partial}{\partial z} g_a(z, w') w' \mathrm{d}w'.$$

With the assumed form of g_a and with $b^2 = 2\sigma_w^2/\tau_{Lw}$, where τ_{L_w} is the Lagrangian timescale for the vertical velocity autocorrelation (this expression for b is chosen to match the form of the Langevin equation in homogeneous conditions), this leads to

$$a = -\frac{w}{\tau_{Lw}} + \sigma_w \frac{\partial \sigma_w}{\partial z} + \frac{w^2}{\sigma_w}\frac{\partial \sigma_w}{\partial z} + \frac{\sigma_w^2}{\rho}\frac{\partial \rho}{\partial z}. \tag{3}$$

To simplify the implementation of this model it is convenient to re-express the model in terms of w/σ_w. This simplification arises because

$$\mathrm{d}\left(\frac{w}{\sigma_w}\right) = \frac{\mathrm{d}w}{\sigma_w} + \mathrm{d}\left(\frac{1}{\sigma_w}\right) w = \frac{\mathrm{d}w}{\sigma_w} - \frac{w^2}{\sigma_w^2}\frac{\partial \sigma_w}{\partial z}\mathrm{d}t$$

and, when $\mathrm{d}w$ is substituted, the last term cancels with that arising from the third term in Equation (3) (see Wilson et al., 1983). This results in the following Langevin equation for the vertical wind component

$$\mathrm{d}\left(\frac{w}{\sigma_w}\right) = -\frac{w}{\sigma_w}\frac{\mathrm{d}t}{\tau_{L_w}} + \frac{\partial \sigma_w}{\partial z}\mathrm{d}t + \frac{\sigma_w}{\rho}\frac{\partial \rho}{\partial z}\mathrm{d}t + \left(\frac{2}{\tau_{L_w}}\right)^{1/2}\mathrm{d}W \ . \tag{4}$$

This equation is valid for inhomogeneous Gaussian turbulence in the PBL with density varying with height. The third term on the right hand side of Equation (4), which was not used in previous studies, corrects for variations in the density of the air. Similar corrections for density variations are also possible in random displacement models where there is no memory of particle velocity (see e.g., Venkatram, 1993; or Thomson, 1995) or in models designed for non-Gaussian turbulence. For non-Gaussian turbulence the corrections are more complex than for the Gaussian case, although only by an amount comparable with the increased complexity of the underlying constant density model.

The above derivation shows that an extra term is required if the model is to reflect the well-mixed condition in flows where ρ varies with height. This extra term (the last term in Equation (3) or the third term on the right hand side of Equation (4)) takes the form of an additional acceleration imposed on the particles and this acceleration arises physically as a consequence of pressure forces. To see this, consider, following Legg and Raupach (1982) and Ley and Thomson (1983), the ensemble average of the Navier-Stokes equations with the assumption of stationarity and horizontal homogeneity:

$$\frac{\partial}{\partial z}\overline{\rho}\sigma_w^2 = -\frac{\partial \overline{p}}{\partial z} - g\overline{\rho}. \tag{5}$$

Here, the ensemble average is denoted by an overbar, p is the pressure and g is the gravitational acceleration. (Strictly speaking σ_w^2 here is a density weighted average of w^2, but the density weighting makes little difference if, as here, we are concerned not so much with density fluctuations at one height as with changes in the mean density with height. In the following we ignore density fluctuations at a single height and write ρ for $\overline{\rho}$.) It follows from Equation (5) that the mean pressure gradient does not exactly balance gravity but differs by an amount $\partial(\rho\sigma_w^2)/\partial z$. This gives rise to an acceleration

$$\frac{1}{\rho}\frac{\partial}{\partial z}\rho\sigma_w^2 \tag{6}$$

and the extra acceleration arising from density variations with height,

$$\frac{\sigma_w^2}{\rho}\frac{\partial \rho}{\partial z},$$

is precisely the extra term derived in Equation (3). Of course, the whole of the acceleration (Equation (6)) matches the average of all the terms on the right hand side of Equation (3) and so gives a partial explanation of the other terms in Equation (3) too (although, at least in the simple form presented here, the argument does not explain the w dependence of the terms).

The time step used for the simulations was the minimum of $c\tau_{L_w}$, $c/|\partial\sigma_w/\partial z|$ and $cz_i/|w|$, where $c = 0.05 \ll 1$. The second criterion was used to prevent σ_w at the particle location changing by a large fraction over a time step, the third one to ensure that particles do change their position only by a small fraction of the PBL height. Overall, minimum values of 10 and 1 sec were also imposed on τ_L and the time step respectively. A further possible limit on the time step would be $c\rho/\left(\sigma_w\,|\partial\rho/\partial z|\right)$, but this criterion is less rigourous than the others. Air density and density gradients were calculated at each model level and half-level, respectively, and were interpolated to the particle's position.

3. Results

The validity of the proposed method was investigated using a simple test case with stationary and horizontally homogeneous meteorological conditions, characterized by the following parameters: friction velocity $u_* = 0.5$ m s^{-1}, convective velocity scale $w_* = 1.5$ m s^{-1}, Obukhov length $L = -100$ m, and PBL height $z_i = 1000$ m. Furthermore, it was assumed that the density in the mixed layer decreases exponentially with height, with density at z_i being 37% of that at the ground. Since this is a sharper density gradient than will ever occur in reality, it will clearly reveal the capability of the model. 8000 particles were released equally distributed between the levels 0 and z_i and were followed for 10 h. The first 5 h were used as spin-up time to perfectly equilibrate particle densities, and during the second 5 h, average particle densities were calculated every 25 m.

Two simulations were made: in the first one, the density correction term was not used, while in the second one it was. The results of the first simulation are shown in Figure 1. The particles are uniformly distributed in z, except for small random deviations. Since no density correction was used, the model failed to predict the correct vertical distribution of the tracer. This shortcoming was removed in the second simulation (Figure 2), where the normalized particle density reflected the assumed air density decrease with height. Thus, density variations can be successfully accounted for with the proposed density correction term.

To assess the importance of the density correction term under realistic conditions, we calculated the tracer dispersion for a few cases for which surface tracer

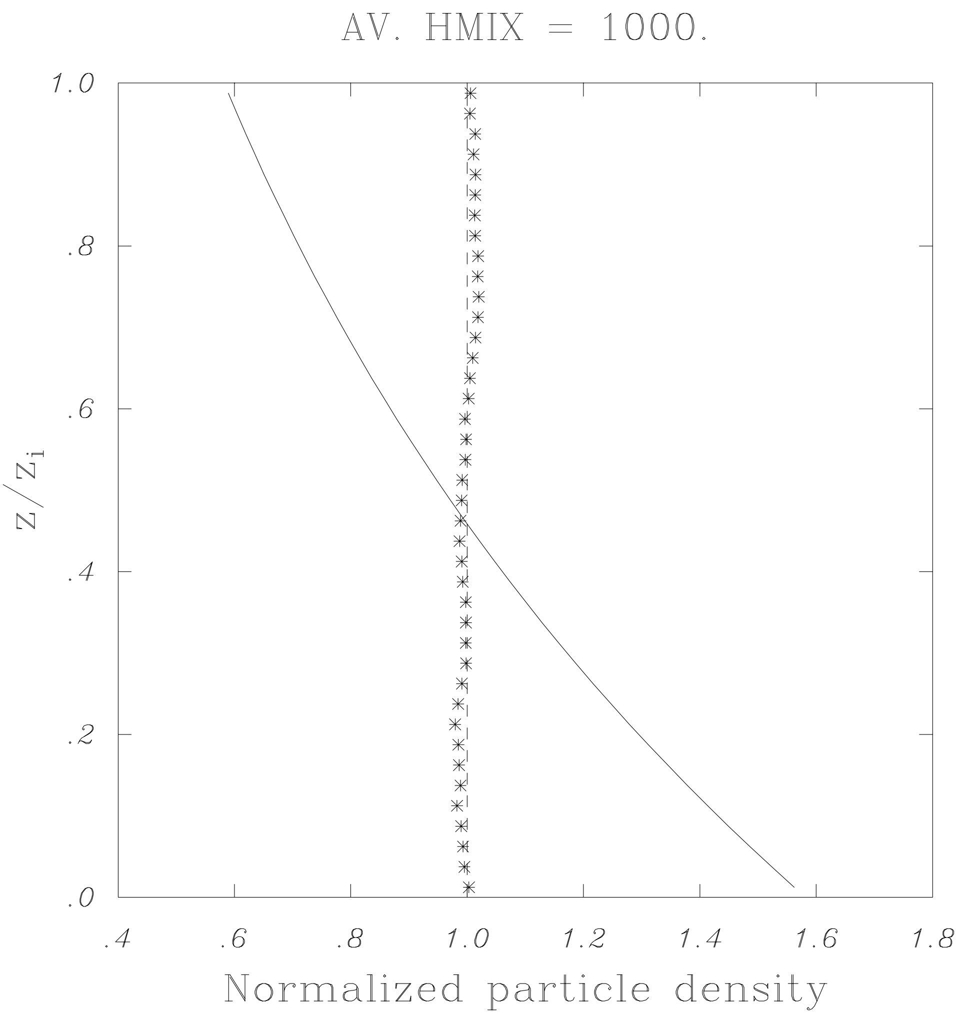

Figure 1. Normalized particle densities as a function of height for a simulation without the density correction term. Data points show the model results, the straight vertical line indicates a perfectly well-mixed state in Cartesian coordinates, and the solid line shows the assumed normalized density of the air.

measurements from a large-scale tracer experiment were available. Stohl et al. (1998) used data from three tracer experiments, the Cross-Appalachian Tracer Experiment (CAPTEX), the Across North America Tracer Experiment (ANATEX) and the European Tracer Experiment (ETEX), to evaluate the performance of FLEXPART. Of these, relatively deep PBLs were diagnosed only for CAPTEX. Hence, only for CAPTEX can a clear effect of the density correction be expected. Therefore, we re-calculated the tracer dispersion for CAPTEX using the density correction.

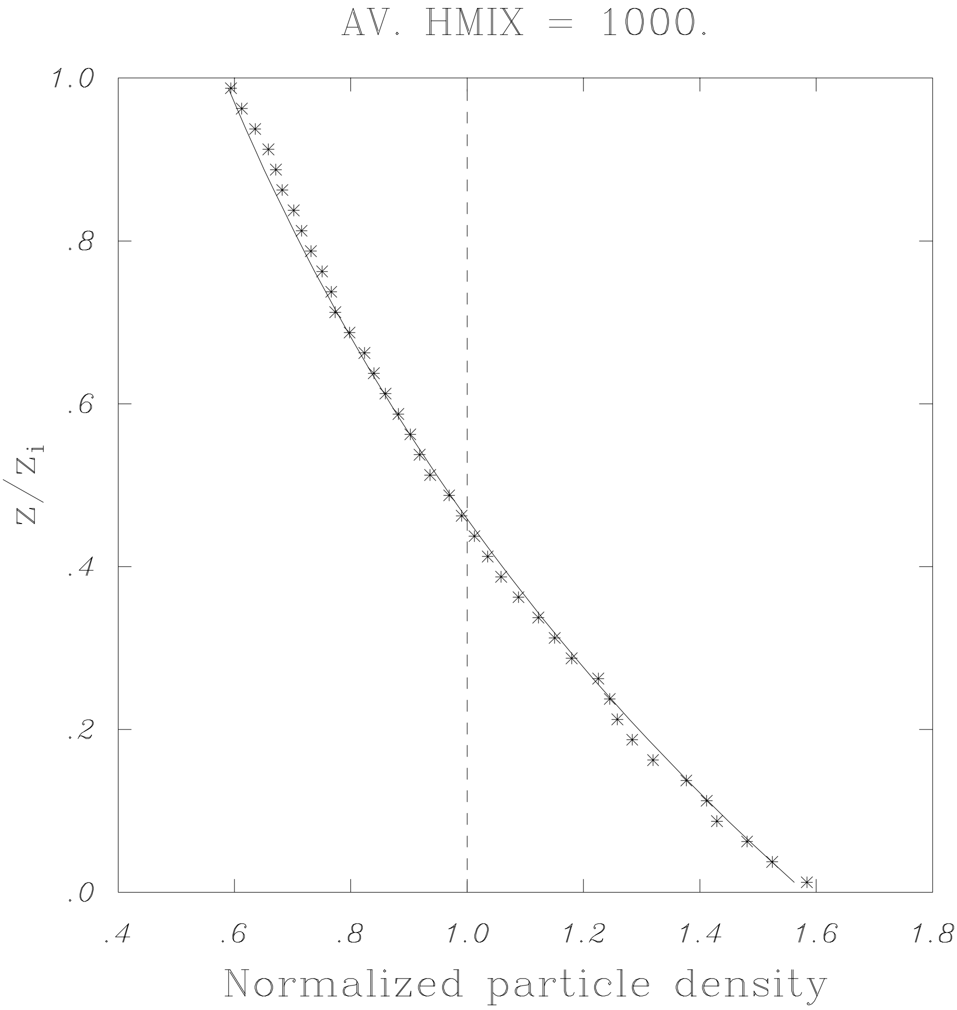

Figure 2. Same as Figure 1, but for a simulation including the density correction term.

CAPTEX was conducted during September and October 1983 in the north-eastern U.S. and south-eastern Canada (Ferber et al., 1986). There were seven 3-h releases, C1–C7, of approximately 200 kg perfluoro- monomethyl-cyclohexane (PMCH) from two different locations. Data from release C6 were not used here because the amount of material released was small and few samples were collected. For the remaining six releases, tracer samples of 3-h and 6-h duration were collected at 84 sites. The 3-h samples were converted to 6-h averages to have a uniform dataset.

Releases C1–C4 were made during daytime under anticyclonic conditions with well-developed convective boundary layers. The south-westerly winds carried the tracer right over the sampling network. C5 and C7 were made from the second location during the nighttime behind cold fronts. Winds were from a north-westerly direction.

The analyzed PBL heights at the release location and time, obtained from the ECMWF data by a combined Richardson number and lifting parcel technique (Vogelezang and Holtslag, 1996), were 1700, 1500, 1700, 1400, 400 and 1000 m for the six releases. During the dispersion, the particle ensembles experienced maximum ensemble-averaged PBL heights of 1700, 1700, 1700, 1500, 1500 and 1400 m. These were not very deep PBLs, but a 6–8% change in the surface concentrations due to the density correction could be expected nevertheless.

100,000 particles were released for each of the six cases and their trajectories were followed for 48–72 h, depending on the length of the period for which measurement data were available. Most of the samples, however, were taken within 36–48 h after the releases. Two calculations were done: the first without, the second with, the density correction term.

The evaluation of the simulations was done in two steps. First, the gridded surface concentration fields (resolution 1° × 1°) obtained with the two methods were compared against each other. Second, the concentrations at the measurement locations, estimated with a parabolic kernel (this method does not produce concentration values by summing particle mass in a fixed volume and dividing by the volume, but weighs the particle masses and respective volumes by the distance to the receptor location; see Uliasz, 1994), were compared to the measured values. For the comparison of the model results and the measurements, global analyses based on four statistical measures were used:

- Fractional bias $FB = \frac{2B}{\overline{P}+\overline{M}}$, where $B = \frac{1}{N}\sum_{i=1}^{N}(P_i - M_i)$ is the bias, N is the number of paired data points, P_i and M_i are the model predictions and the measurements, and $\overline{P}$ and $\overline{M}$ are the average predictions and measurements, respectively.
- Normalized mean square error $NMSE = \frac{1}{N}\sum_{i=1}^{N}\frac{(P_i-M_i)^2}{\overline{P}\,\overline{M}}$
- Pearson correlation coefficient r
- Figure of merit in space $FMS = 100\frac{A_p \cap A_m}{A_p \cup A_m}$, where A_p and A_m are the predicted and measured subsets of concentrations above a significant level (2 fl l^{-1}).

Furthermore, the relative bias $RB = (\overline{C_d}/\overline{C_{nd}} - 1)$, where $\overline{C_d}$ and $\overline{C_{nd}}$ are the average concentrations calculated with and without the density correction, was used to compare the results of the two model versions directly with each other. RB gives the average underestimation of the concentrations when the density correction is not used.

First, we will discuss the results of the model against model comparisons. For the global analyses of the individual releases, RB ranged from 2.9–8.0%, with an average of 5.5% (Table I). The smallest value occurred for C5, where the PBL height at the time of the release was only 400 m and the PBL was shallow during much of the period considered. All other RB were above 5%. If values of RB

TABLE I

Global analyses of the model against model comparisons for the six CAPTEX releases. The last row shows the results for a global analysis performed over all the data.

Release	RB(%)	*NMSE*
C1	8.0	9.4
C2	6.6	5.4
C3	5.4	6.6
C4	6.0	6.3
C5	2.9	1.6
C7	6.2	3.3
C1–C7	5.5	4.3

were calculated from individual fields and not globally for the whole periods of the model calculations, they covered a larger range from 1–15%. Larger values of RB were usually found at the end of a simulation than at its beginning. For instance, if only the concentration fields later than 36 h after the release were considered in the statistics, the average RB was 7.3% (compared to 5.5% globally). This was due to the fact that the maximum PBL height experienced by the particles determined how many of them escaped to the free troposphere when the PBL height decreased subsequently. The loss was systematically too large without the density correction.

The global *NMSE* statistic for the difference between the simulations with and without the density correction ranged from 1.6–9.4 with an average of 4.3. The largest value was in the range of the smallest values reported by Stohl et al. (1998) in comparisons of model results against measurements. Thus, neglecting the density correction can, in principle, significantly impair otherwise good model results. We split the model results into two subsets with 50,000 particles each to make sure that these relatively large values of *NMSE* between the two simulations were statistically significant and not due to an under-sampling of the plume by too few particles. When we compared these two subsets, we found values of the *NMSE* statistics for the difference between the subsets that ranged from 0.3–1.8 for the individual releases. Thus, it was clear that the larger *NMSE* values found in the first comparison were not due to under-sampling, but were actually caused by the difference in the model formulation.

Values of the *FMS* obtained in comparing the simulations with and without the density correction were all above 95% and correlation coefficients r were always very close to 1, indicating that the transport of the tracer cloud was very similar

TABLE II

Global analyses of the model simulations for which the density correction was not used. N is the total number of data pairs used for the analysis (including zero values).

Release	N	*FB*	*NMSE*	*FMS*	*r*
C1	362	+0.46	21.8	50	0.61
C2	366	−0.11	10.4	63	0.76
C3	366	+1.12	32.4	28	0.31
C4	312	+0.10	35.6	49	0.40
C5	316	−0.47	53.3	37	0.25
C7	206	−0.38	15.6	45	0.40
C1–C7	1928	+0.08	22.4	46	0.48

for the two model versions. This was also confirmed by visual comparisons of the tracer footprints, where no large differences were found, except for the magnitude of the values.

Now we will discuss the evaluation of both model versions against measurement data. Tables II and III show the global analyses of the model results when the density correction was not used and when it was used, respectively. It appears that there was little difference in the performance of the two model versions. Most obvious were the higher concentrations obtained with the density correction, leading to higher values of the *FB* in all cases (which actually meant an improvement in three of six cases). *NMSE* values were, with the exception of C3, always smaller when the density correction was used. But the differences are small and could be due to the fact that *NMSE* favours over-predicting models (Poli and Cirillo, 1993), and does not necessarily indicate an actual improvement. Values of *FMS* and *r* were very similar for both model versions.

The reason why there was no demonstrable improvement of the model results due to the density correction obviously lies in the various other sources of error, such as wind field analysis errors or errors in the diagnosed PBL heights. For instance, Wotawa and Stohl (1997) found that during stable conditions long-term averaged PBL heights diagnosed from ECMWF data with different methods can differ by up to 50%. Individual errors can be even larger. Improvements of the calculations due to the density correction can easily be masked by these errors.

TABLE III

Global analyses of the model simulations for which the density correction was used.

Release	N	*FB*	*NMSE*	*FMS*	*r*
C1	362	+0.53	20.8	48	0.62
C2	366	−0.02	9.2	63	0.76
C3	366	+1.16	35.2	28	0.31
C4	312	+0.18	34.3	49	0.41
C5	316	−0.42	51.8	39	0.25
C7	206	−0.32	15.1	43	0.39
C1−−C7	1928	+0.15	21.5	46	0.48

4. Conclusions

Lagrangian particle models which are currently used to describe the dispersion of passive tracers in the turbulent PBL neglect the decrease in air density with height, although, in a deep PBL, density at the PBL top may be lower than at the ground by more than 20%. This leads to a systematic underestimation of surface tracer concentrations and, in the presence of a vertical wind shear, to an inaccurate calculation of the horizontal transport.

In this paper, we introduced a density correction for atmospheric Lagrangian particle dispersion models that is valid for Gaussian turbulence and is simple to apply. We demonstrated that the proposed Langevin equation accurately describes turbulent diffusion in a PBL where the density decreases with height. We calculated the dispersion of a passive tracer under realistic conditions and found that the density correction caused a change in time-averaged surface tracer concentrations of up to 8%. This result is in good agreement with the effect expected for the meteorological conditions considered with maximum PBL heights of approximately 1700 m. Larger effects can be expected for higher PBLs. Even larger differences will occur if LPD models are to be used for simulating deep convection.

Theoretically, in the presence of a vertical shear of the horizontal wind, a change in the vertical distribution of the particles should also affect the horizontal transport patterns. However, we did not detect a significant change in the position and extension of the tracer clouds in the six cases considered. Also, we could not find a clear improvement in the model results when they were evaluated against independent measurements from the CAPTEX tracer experiment. Obviously, the small improvements due to the density correction were masked by larger errors from other sources such as inaccuracies in the determination of the PBL heights.

Unfortunately, we had no experimental dataset available that was especially suitable for an evaluation of the density correction. Such an experiment should be conducted under meteorological conditions with very large PBL heights and should cover a period of three to four days. The large PBL heights would increase the effect of the density inhomogeneities; a long duration of the experiment would facilitate the detection of changes in the horizontal transport patterns, which develop very slowly when the vertical wind shear is not extremely strong. However, detecting such systematic but relatively small effects compared to other sources of error is likely to be difficult even under such idealised conditions.

The computational costs of the density correction are relatively small. All that is required for its application are interpolations of the air density and the vertical air density gradients to the particle's position. As a first approximation, it would be even possible to avoid this interpolation by assuming a typical decrease of density with height. A minor increase in calculation time results from the fact that, due to the correction, more particles reside in the lowest parts of the PBL, where Lagrangian time scales are usually small and short time steps are thus required.

Acknowledgements

We are thankful to ECMWF for giving us data access (special project 'Validation of trajectory calculations') and support (Norbert Kreitz). Discussions with Stefan Heinz and the comments of an anonymous reviewer were very helpful. The people who did the valuable CAPTEX measurements are gratefully acknowledged.

References

Baumann, K. and Stohl, A.: 1997, 'Validation of a Long-Range Trajectory Model Using Gas Balloon Tracks from the Gordon Bennett Cup 95', *J. Appl. Meteorol.* **36**, 711–720.

ECMWF: 1995, *User Guide to ECMWF Products 2. 1*. Meteorological Bulletin M3. 2. ECMWF, Reading, U.K.

Ferber, G. J., Heffter, J. L., Draxler, R. R., Lagomarsino, R. J., Thomas, F. L., Dietz, R. N., and Benkovitz, C. M.: 1986, 'Cross-Appalachian Tracer Experiment (CAPTEX 83)', Final Rep. NOAA Tech. Memo. ERL ARL-142, Air Resources Laboratory, NOAA Environmental Research Laboratories, Silver Spring, MD, 60 pp.

Hanna, S. R.: 1982, 'Applications in Air Pollution Modeling', in F. T. M. Nieuwstadt and H. van Dop (eds.), *Atmospheric Turbulence and Air Pollution Modelling*, D. Reidel Publishing Company, Dordrecht, 358 pp.

Legg, B. J. and Raupach, M. R.: 1982, 'Markov-Chain Simulation of Particle Dispersion in Inhomogeneous Flows: The Mean Drift Velocity Induced by a Gradient in Eulerian Velocity Variance', *Boundary-Layer Meteorol.* **24**, 3–13.

Ley, A. H. and Thomson, D. J.: 1983, 'A Random Walk Model of Dispersion in the Diabatic Surface Layer', *Quart. J. Roy. Meteorol. Soc.* **109**, 867–880.

Poli, A. and Cirillo, M. C.: 1993, 'On the Use of the Normalized Mean Square Error in Evaluating Dispersion Model Performance', *Atmos. Environ.* **27A**, 2427–2434.

Rodean, H.: 1996, 'Stochastic Lagrangian Models of Turbulent Diffusion', *Meteorological Monographs* **26**(48), American Meteorological Society, Boston, U.S.A.

Stohl, A.: 1996, 'The FLEXPART Particle Diffusion Model – Scientific Description and Technical Documentation', Report to the Central Institute of Meteorology and Geodynamics, Hohe Warte 38, 1190 Wien, Austria. Available as an updated user guide via the internet from http://www. forst. uni-muenchen. de/LST/METEOR/stohl/flexpart. html.

Stohl, A.: 1998, 'Computation, Accuracy and Applications of Trajectories – A Review and Bibliography', *Atmos. Environ.* **32**, 947–966.

Stohl, A., Hittenberger, M., and Wotawa, G.: 1998, 'Validation of the Lagrangian Particle Dispersion Model FLEXPART Against Large-Scale Tracer Experiments', *Atmos. Environ.*, in press.

Stohl, A. and Seibert, P.: 1997, 'Accuracy of Trajectories as Determined from the Conservation of Meteorological Tracers', *Quart. J. Roy. Meteorol. Soc.* **125**, 1465–1484.

Stohl, A., Wotawa, G., Seibert, P., and Kromp-Kolb, H.: 1995, 'Interpolation Errors in Wind Fields as a Function of Spatial and Temporal Resolution and their Impact on Different Types of Kinematic Trajectories', *J. Appl. Meteorol.* **34**, 2149–2165.

Thomson, D. J.: 1995, 'Discussion', *Atmos. Environ.* **29**, 1343.

Thomson, D. J.: 1987, 'Criteria for the Selection of Stochastic Models of Particle Trajectories in Turbulent Flows', *J. Fluid Mech.* **180**, 529–556.

Uliasz, M.: 1994, 'Lagrangian Particle Dispersion Modeling in Mesoscale Applications', in P. Zannetti (ed.), *Environmental Modeling*, Vol. II, Computational Mechanics Publications, Southampton, U.K., pp. 71– 101.

Venkatram, A.: 1998, 'Response', *Atmos. Environ.* **32**, 259.

Venkatram, A.: 1993, 'The Parameterization of the Vertical Dispersion of a Scalar in the Atmospheric Boundary Layer', *Atmos. Environ.* **27A**, 1963–1966.

Vogelezang, D. H. P. and Holtslag, A. A. M.: 1996, 'Evaluation and Model Impacts of Alternative Boundary-Layer Height Formulations', *Boundary-Layer Meteorol.* **81**, 245–269.

Wilson, J. D., Legg, B. J., and Thomson, D. J.: 1983, 'Calculation of Particle Trajectories in the Presence of a Gradient in Turbulent-Velocity Scale', *Boundary-Layer Meteorol.* **27**, 163–169.

Wilson, J. D. and Sawford, B. L.: 1996, Review of Lagrangian stochastic models for trajectories in the turbulent atmosphere', *Boundary-Layer Meteorol.* **78**, 191–210.

Wotawa, G. and Stohl, A.: 1997, 'Boundary Layer Heights and Surface Fluxes of Momentum and Heat Derived from ECMWF Data for Use in Pollutant Dispersion Models – Problems with Data Accuracy', in S.-E. Gryning, F. Beyrich, and E. Batchvarova (eds.), *The Determination of the Mixing Height – Current Progress and Problems*, EURASAP Workshop Proceedings, 1–3 October 1997, Risø National Laboratory, Denmark.