Global random walk modelling of transport in complex systems

N. Suciu[*1,2]**, C. Vamoş**[2]**, I. Turcu**[3]**, C.V.L. Pop**[3]**, and L. I. Ciortea**[3,4]

[1] Friedrich-Alexander University of Erlangen-Nuremberg, Institute of Applied Mathematics,
   Martensstrasse 3, 91058 Erlangen, Germany
[2] "Tiberiu Popoviciu" Institute of Numerical Analysis, Cluj Napoca branch of the Romanian Academy,
   P.O. Box 68-1, 400320, Cluj Napoca, Romania
[3] National R&D Institute for Isotopic and Molecular Technologies, P.O. Box 700, 400293 Cluj-Napoca, Romania
[4] School of EEIE, South Bank University 103 Borough Road, London SE1 0AA, UK

Abstract. The Global Random Walk algorithm performs simultaneously the tracking of large collections of particles and permits massive simulations at reasonable costs. Applications were developed for transport in systems with anisotropic, non-homogeneous, and randomly distributed parameters. As a first illustration we present simulations for diffusion in human skin. Further, a case study for contaminant transport in groundwater shows that the realizations of the transport process converge in mean square limit to a Gaussian diffusion. This investigation also indicates that the use of the Kraichnan routine, based on periodic random fields, yields reliable simulations of transport in Gaussian velocity fields.

Key words Global random walk, Lattice gas, Human skin, Groundwater contamination

1 Introduction

The Global Random Walk (GRW) is a recently proposed method for the simulation of transport processes which generalizes the classical Particle Tracking algorithm (PT). The new approach considerably increases the accuracy and reduces the computation time. Nonisotropic and space variable diffusion coefficients as well as various boundary and initial conditions can be easily implemented in GRW [Vamoş et al., 2003].

Unlike PT, GRW acts "globally" since, starting with a given distribution of particles in a computational grid, all the particles lying at a grid site are simultaneously spread, first by an advection displacement, then by diffusion jumps, as shown in Fig. 1. The numbers of particles undergoing diffusion jumps are Bernoulli random variables, describing the number of jumps in a given direction of a discrete random walk process. Thus, the GRW procedure is equivalent to a superposition of many

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[*] Corresponding author: suciu@am.uni-erlangen.de

PT procedures and can manage large numbers of particles. GRW is also equivalent to a finite-difference scheme, for genuine diffusion processes, and it is appropriate to solve more complex problems for parabolic equations. The "overshooting" cannot be avoided in GRW procedure but, for given overshooting, the discretization errors can be controlled and kept under a desirable threshold by an accurate discretization of the velocity [Suciu et al., 2004, 2006a].

The "global change of state" of the whole system of computational particles, as a cellular automaton, is the principle on which are built the "lattice gas" and "lattice Boltzmann" methods, and it allows high performance parallel computing applications [Wilke et al., 2003]. In this respect, GRW is a particular lattice gas model, i.e. it is a stochastic process in the space of configurations, defined at a given time by the number of particles at a grid site. Since the particles do not interact, there is no collision step, as for lattice Boltzmann. The "particle distribution function" along all the possible positions occupied by particles after a "propagation" step consisting of an advection displacement and diffusion jumps is given by the Bernoulli distribution or its approximations. Similarly with gas lattice, the next configuration of the system is obtained by simple summations of the number of particles at each grid site.

Since in GRW algorithm, unlike in lattice gas models, the number of particles per grid site is not limited by an "exclusion principle" it is possible to obtain statistically reliable estimations of the macroscopic quantities, in a single simulation, without averaging over ensembles of simulations. The required number of particles ranges between 10^6, for small size lattices of the order of 100 nodes [Vamoş et al., 2003] to 10^{10} for lattice size of $\sim 10^6$ nodes [Suciu et al., 2004, Fig. 11]. In some simplified implementations of the algorithm there are no limitations as concerning the number of particles. Therefore, GRW can simulate the behavior of the real number of molecules involved in microscopic processes (diffusion, chemical reactions and radioactive decay) as well as in large scale

environmental problems (contaminant transport in water or atmosphere).

The paper is organized as follows. Section 2 presents the principle of the GRW algorithm. A two-dimensional algorithm for space variable and anisotropic diffusion tensor is described in Sect. 3, and in Sect. 4 it is applied for simulations of diffusion in human skin. Section 5 deals with two-dimensional simulation of large scale transport in groundwater and Sect. 6 presents some conclusions.

2 The GRW algorithm

The GRW algorithm simulates the random walk of an ensemble of fictitious particles to solve parabolic partial derivative equations. As an illustration, we first present the GRW algorithm for the case of the one-dimensional diffusion whose probability density (normalized concentration of the diffusing substance) is a solution of the Fokker-Planck equation with space variable drift coefficient $V(x)$ and constant diffusion coefficient D,

$$\partial_t c + V(x)\partial_x c = D\partial_x^2 c. \tag{1}$$

Let us consider a lattice with constant δx, defined by the nodes $x_i = i\delta x$, and the discrete time $t_k = k\delta t$, where i and k, $k \geq 0$, are integers. The configuration of N particles evolving on the lattice is given by the number $n(i,k)$ of particles lying at x_i at the time step t_k. At every time step the particles from every lattice site are grouped into three subsets:

$$n(j,k) = \delta n(j + v_j \mid j,k) + \delta n(j + v_j - d \mid j,k) \tag{2}$$
$$+ \delta n(j + v_j + d \mid j,k).$$

The term $\delta n(j+v_j \mid j,k)$ is the number of particles which are moved from x_j to $x_j + v_j$, where $v_j = V(x_i)\delta t/\delta x$. The terms $\delta n(j + v_j \pm d \mid j,k)$ in (2) count the rest of particles, which undergo diffusive right-left jumps over d lattice nodes and are Bernoulli random variables. The new configuration of the particles after a time step is given by the sum of the particles which at the time $(k+1)\delta t$ are at the same lattice site,

$$n(i,k+1) = \sum_j \delta n(i \mid j,k). \tag{3}$$

By repeating this procedure for different sets of realizations of the Bernoulli variables δn one obtains a statistical ensemble of simulations. The ensemble average of the number of particles undergoing diffusive jumps and of the number of particles remaining at the same site after the displacement v_j are given by the relations

$$\overline{\delta n(j + v_j \pm d \mid j,k)} = \frac{1}{2}r\,\overline{n(j,k)}, \tag{4}$$
$$\overline{\delta n(j + v_j \mid j,k)} = (1-r)\,\overline{n(j,k)},$$

where r, $0 \leq r \leq 1$ is a real parameter. The diffusion coefficient D is related to the amplitude of diffusive jumps d, the parameter r, the lattice constant and the time step by the relation

$$D = r\frac{(d\delta x)^2}{2\delta t}. \tag{5}$$

The concentration simulated by the 1-dimensional GRW algorithm is given by $c(x_i, t_k) = n(i,k)/\delta x$. For vanishing or constant drift coefficients V, the relations (3-5) imply that the average of $c(x_i, t_k)$ over an ensemble of GRW simulations coincides with the solution of the stable finite difference scheme for the diffusion equation [Vamoş et al., 2003]. For variable V, GRW is no longer equivalent to a finite difference scheme. This is due to overshooting errors occurring when particles jump over grid sites with different $V(x_i)$ and is common to most of the random walk methods. Nevertheless, an evaluation by comparisons with a method free of overshooting [Suciu et al., 2005] has shown that, even for highly variable V, the GRW solution remains accurate in a range of 5% for suitable chosen parameters δx, δt, d, and r [Suciu and Vamoş, 2006].

In the "exact" version of the GRW the quantities $\delta n(j + v_j \pm d \mid j,k)$ are picked up from Bernoulli repartition functions, using computer generated random numbers. This procedure requires the expression of the number of jumping particles in a series of powers of 2, the computation and the storage of the Bernoulli repartitions for increasing power of 2, and the generation of some random numbers at every time step and lattice site. Accordingly, the computing time increases for large N. Several approximations of the exact algorithm can be used to avoid this inconvenience. For instance, if the number of particles at a lattice site is larger than 2^{20}, the Bernoulli repartition can be successfully approximated by the Gaussian one (i.e. with the erf-function). Another useful and robust approximation is that leading to the "reduced fluctuations GRW algorithm". In both cases there are no limitations for the maximum number of particles, which can be as large as the maximum double precision value of the computing platform.

The reduced fluctuations GRW algorithm is defined by

$$\delta n(j + v_j - d \mid j,k) = \begin{cases} n/2 & \text{if } n \text{ is even} \\ [n/2] + \theta & \text{if } n \text{ is odd} \end{cases},$$

where $n = n(j,k) - \delta n(j + v_j \mid j,k)$, $[n/2]$ is the integer part of $n/2$ and θ is a variable taking the values 0 and 1 with probability 1/2. This algorithm minimizes the number of calls of the random number generator and makes the computing time independent of N. Therefore the reduced fluctuations GRW algorithm is mainly useful for large scale problems. Moreover, a physical significant shape of the diffusion front is ensured by the threshold concentration defined by one particle at a grid point. This feature also saves memory and computing time by avoiding computations at lattice sites with irrelevant small concentrations, as in case when one gives up the particle indivisibility or in finite differences schemes [Vamoş et al., 2003]. In the following by GRW we shall denote only reduced fluctuations algorithms.

For constant diffusion coefficient D the two and three-dimensional algorithms can be simply built by repeating the same procedure for all space directions. Figs. 1 and 2 illustrate the two-dimensional GRW algorithm for constant D.
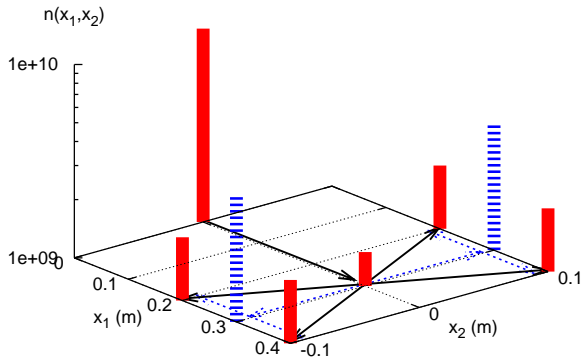
Fig. 1. The advection displacement and diffusive jumps of 10^{10} particles starting at $(0,0)$, for r=0.85; the blue columns and lines represent the succession of the 1-dimensional procedures
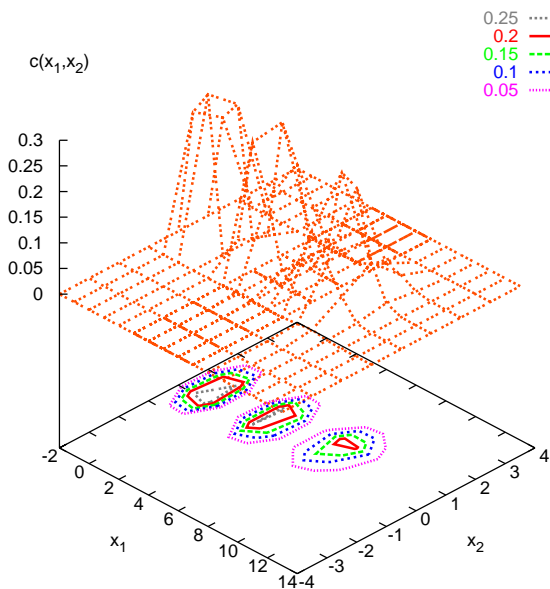

Fig. 2. The resulting concentration field, at $t = 0$, $t = 5\delta t$ and $t = 10\delta t$, for a uniform initial distribution of 10^{10} particles in the square of sides δx_1 and δx_2.

The concentration plotted in Fig. 2 was computed at a given time $t = k\delta t$ and at a lattice site $(x_1, x_2) = (i_1\delta x_1, i_2\delta x_2)$, where δt is the time step, δx_1, and δx_2 are the space steps, with the formula

$$c(x_1, x_2, t) = \frac{1}{N} \frac{1}{\delta x_1 \delta x_2} \sum_{i'_1=i_1-1}^{i_1+1} \sum_{i'_2=i_2-1}^{i_2+1} n(i'_1, i'_2, k\delta t). \quad (6)$$

Let us consider, more generally, an initial condition consisting of N particles uniformly distributed over N_{x_0} grid sites. By $n(i_1, i_2, k; x_{0,1}, x_{0,2})$ we denote the distribution of particles at the time step k given by the GRW

procedure for a diffusion process staring at $(x_{0,1}, x_{0,2})$. Since the distribution of the particles at time k can be written as

$$n(i_1, i_2, k) = \sum_{x_{0,1}, x_{0,2}} n(i_1, i_2, k; x_{0,1}, x_{0,2}),$$

it follows that

$$\frac{1}{N} \sum_{i_1, i_2} n(i_1, i_2, k)$$

$$= \frac{1}{N_{x_0}} \sum_{x_{0,1}, x_{0,2}} \frac{N_{x_0}}{N} \sum_{i_1, i_2} n(i_1, i_2, k; x_{0,1}, x_{0,2}).$$

Thus, the concentration (6), as well as its spatial moments, are averages over the trajectories of the diffusion process starting at given initial positions and over the distribution of the initial positions.

It was shown that for Gaussian diffusion the numerical solution (6) converges to the Gaussian distribution as $\mathcal{O}(\delta x^2) + \mathcal{O}(1/\sqrt{N})$, i.e. for large numbers of particles, N, the convergence order is $\mathcal{O}(\delta x^2)$, the same as for the finite differences scheme [Vamoş et al., 2003]. Therefore, GRW is "self-averaging", in the sense that for large N the solution given by a single simulation obeys the same equation (the finite difference scheme) as the ensemble average of (6). The self-averaging property distinguishes GRW from the gas-lattice approaches, where the "exclusion principle" does not permit the presence large numbers of identical particles at the same site and the solution of the problem is obtained by averaging over ensembles of simulations.

The algorithm is obviously stable since the total number of particles N contained in the lattice is conserved. The condition $r \leq 1$, ensures that there is no numerical diffusion. In [Vamoş et al., 2003] it was also shown that GRW is orders of magnitude faster than PT, in all its possible implementations, and is able to handle large numbers of particles that are prohibitive for the classical PT algorithms.

3 GRW algorithm for two-dimensional diffusion in non-homogeneous and anisotropic media

In the following we consider a two-dimensional diffusion process described by the diagonal diffusion tensor

$$\left\| \begin{matrix} D_x(x,y) & 0 \\ 0 & D_y(x,y) \end{matrix} \right\|$$

and the diffusion equation

$$\partial_t c = \partial_x(D_x \partial_x c) + \partial_y(D_y \partial_y c).$$

The GRW solution of the diffusion equation with spatially dependent diffusion coefficients requires correction terms in (2). These corrections have to account for a drift term equal to $(\partial_x D_x, \partial_y D_y)$ [see e.g. Kinzelbach and Uffink, 1991]. However, for the purpose of illustration, we consider here only the particular case of slowly variable diffusion coefficients, i.e. $\partial_x D_x \approx 0$ and $\partial_y D_y \approx 0$, for which the diffusion equation can be approximated by

$$\partial_t c = D_x \partial_x^2 c + D_y \partial_y^2 c. \quad (7)$$
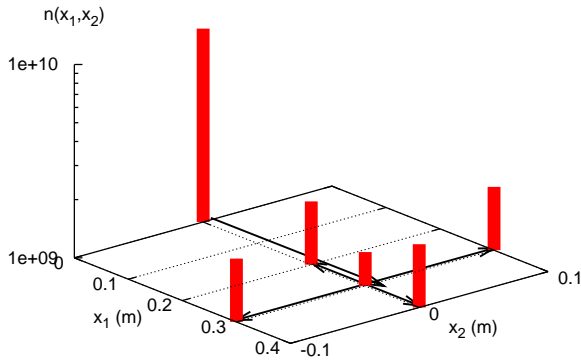
Fig. 3. The two-dimensional GRW algorithm for variable diffusion coefficients.

The equation (7) has no drift terms and the diffusion coefficients vary in space. In this case the GRW relation (2) is replaced by

$$n(i,j,k) = \delta n(i,j \mid i,j,k)$$
$$+\delta n(i - d_x, j \mid i,j,k) + \delta n(i + d_x, j \mid i,j,k)$$
$$+\delta n(i, j - d_y \mid i,j,k) + \delta n(i, j + d_y \mid i,j,k) \tag{8}$$

where $n(i,j,k)$ is the number of particles at the site $(x_i, y_j) = (i\delta x, j\delta y)$ at the time $k\delta t$. Unlike the two-dimensional algorithm presented in Fig. 1, where the one-dimensional procedure is applied successively for longitudinal and transversal directions, the procedure (8) moves the particles simultaneously along the principal directions of the diffusion tensor. Fig. 3 illustrates this two-dimensional algorithm, in the general case of a non vanishing drift.

The anisotropy is taken into account by two different parameters d_x and d_y which describe the diffusive jumps along the coordinates axes. The spatial variation of the diffusion coefficients is described through the variable parameters

$$r_x = \frac{2D_x \delta t}{(d_x \delta x)^2}, \; r_y = \frac{2D_y \delta t}{(d_y \delta y)^2}. \tag{9}$$

After the global change of state of the lattice by the procedure (8) applied at every site containing particles, the new numbers of particles at sites are obtained similarly to (3) by summation over two spatial indices.

The average over an ensemble of simulations of the terms in (8) are related by

$$\overline{\delta n(i \pm d_x \mid j,k)} = \frac{1}{2} r_x(i,j) \; \overline{n(i,j,k)},$$

$$\overline{\delta n(i \mid j \pm d_y, k)} = \frac{1}{2} r_y(i,j) \; \overline{n(i,j,k)},$$

$$\overline{\delta n(i,j \mid i,j,k)} = [1 - r_x(i,j) - r_y(i,j)] \; \overline{n(j,k)}.$$

These relations can be used to show that for slowly variable diffusion coefficients the two-dimensional GRW algorithm approximates the finite difference scheme.

The lattice steps δx and δy are chosen accordingly to the desired resolution of the concentration field. The time step δt is inferred from the condition $r_x + r_y \leq 1$, which states that the numbers of diffusing particles are limited by the numbers of particles at the lattice sites. From (9) it follows

$$\delta t \leq \left(\frac{2D_x^{\max} \delta t}{(d_x \delta x)^2} + \frac{2D_y^{\max} \delta t}{(d_y \delta y)^2} \right), \tag{10}$$

where $D_x^{\max}$ and $D_y^{\max}$ are the upper bounds of the diffusion coefficients $D_x(x,y)$ and $D_y(x,y)$.

4 Diffusion in skin

The human skin varies in texture, structure and thickness and is made up of three main layers: namely epidermis, dermis and hypodermis. The epidermis is the uppermost layer of the skin, measures less than 1 mm in thickness and can be also subdivided into several layers. The Stratum Corneum (SC) is located on the outer surface of the skin and its thickness is in the range $10 - 100$ μm. It is made of keratinized flat, roughly hexagonally shaped, partly overlapping cells embedded in a lipid matrix. Its main constituents are proteins, lipids and water. The dermis is the second layer of the skin and acts as a supportive layer for the epidermis and the hypodermis, the third layer of the skin, acts as a shock absorber and an energy reserve.

The skin is a complex heterogeneous medium that plays an essential part in body protection and regulation: its most outer layer, the SC, acts as a protective barrier between the body and the environment and its main role to prevent fluid loss and to exclude environmental toxins, is fundamental to life. The excellent diffusional resistance of the SC makes the transdermal delivery of drugs pretty difficult. Nevertheless, there remains an important pharmaceutical need to reliably predict the topical and/or transdermal bioavailability of cutaneously applied drugs. In this sense, to measure percutaneous penetration of the externally applied chemicals into human skin is very important for understanding how drugs can be delivered through the skin.

In most cases a constant diffusion coefficient for transported material adequately describes the diffusion process. However, in real biological systems, because of the heterogeneity of the skin, the diffusion coefficient behaves like a random variable. This is why adequate analytical and numerical models able to describe the diffusion process in complex structures must be developed. An example of such a model was developed by [Johnson et al., 1997], who focused on measuring and modeling the molecular diffusion coefficient in order to describe the macroscopic SC permeation via the interkeratinocyte lipid.

The two dimensional algorithm introduced in Sect. 3 can be used as a new approach able to simulate the diffusion through the human skin. A two dimensional geometry with the x axis parallel and the y axis perpendicular to the surface of the skin was used. A thin film consisting of $N = 10^{20}$ water molecules with $\Delta x = 10$ mm was considered to be applied on the surface of the skin. Since the skin structure is stratified, the diffusion
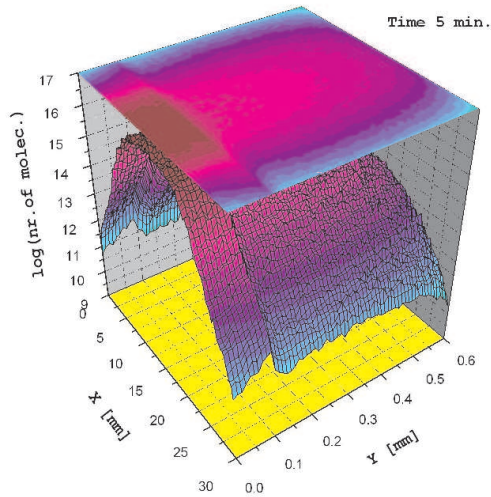
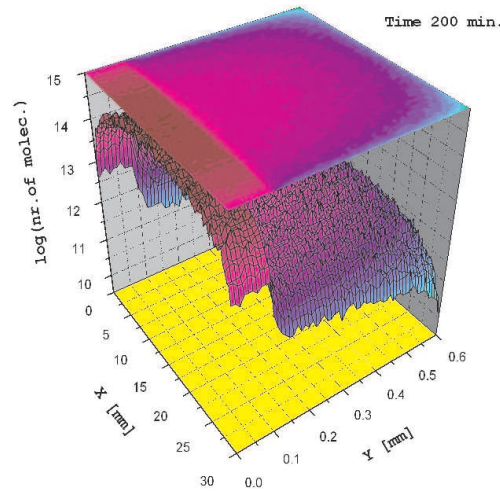**Fig. 4.** The distribution of molecules after $t = 5$ min.

Fig. 5. The distribution of molecules after $t = 200$ min.

Fig. 6. Depth averaged probability density as a function of longitudinal coordinate for fixed times.
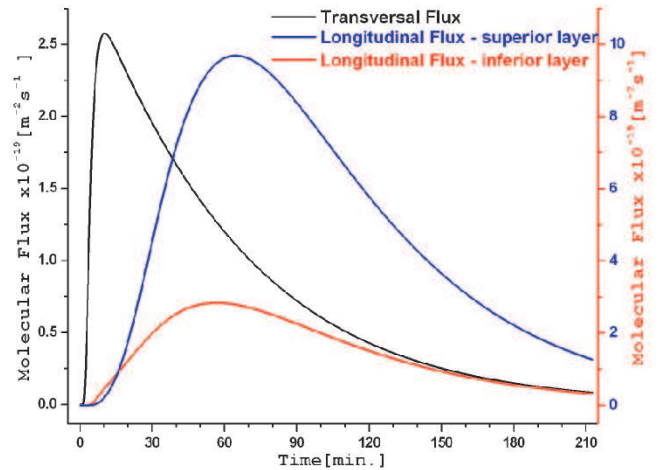
Fig. 7. Time behavior of longitudinal and transversal flux of molecules through the skin.

can be described by the two-dimensional equation (7). To describe the diffusion in the horizontal direction the lattice dimension on the x axis was $3\Delta x$. An acceptable resolution was obtained with $\delta x = 0.1$ mm, (300 nodes per lattice). A simplified two layers structure of the skin was considered, with thickness $y_1 = 0.1$ mm and $y_2 = 0.5$ mm respectively. With a resolution of $\delta y = 0.01$ mm the lattice extended over 10 nodes in the first layer and over 50 nodes in the second.

Because of the nonhomogeneous structure of the skin the diffusion coefficients show spatial fluctuations about the mean value. At every lattice site the coefficients D_x and D_y were generated as normal random variables with the mean value equal to half the maximal values $D_x^{\max}$ and $D_y^{\max}$ and variance equal to a fraction $p < 1$ of the corresponding maximal values. In the simulations presented in the following we chose $p = 0.1$. Since the cells are rather flat, here we considered $D_x^{\max} = 2D_y^{\max} = 5.8810^{-7}$ $m2/s$ in the first layer. In the second layer an isotropic diffusion coefficient ten times larger than in the superior layer was introduced. Between the layers a tran-
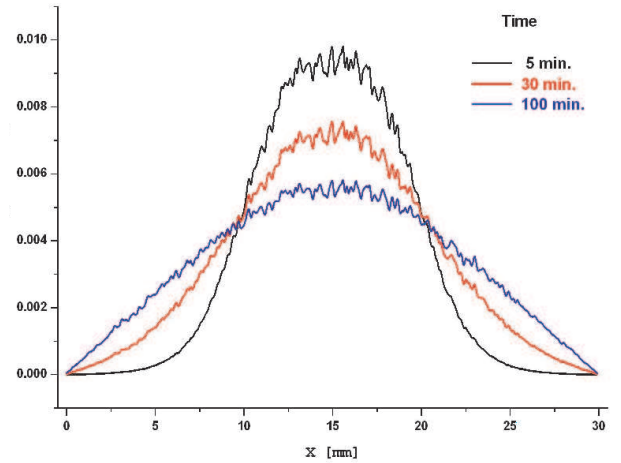
sition zone of thickness $3\delta y$ was placed, where diffusion coefficients vary linearly [Vamoş et al., 2004].

Two different boundary conditions were used. At the surface of the skin, the molecules which have been jumped in the exterior were blocked on the boundary. At the inferior boundary we imposed a "transmission boundary condition" in the first order of approximation Vamoş et al., 2003]. This last condition has the property to not disturb the diffusion front at the boundary. The time step was chosen using the condition (10). Some simulation results are presented in Figs. 4-7.

5 Transport in heterogeneous porous media

Some applications to transport in groundwater show that GRW is suitable to be used in large scale simulations and in Monte Carlo approaches on the predictability of the stochastic model [Suciu et al., 2004, 2006a].

We considered two-dimensional divergence-free velocity fields with constant mean $\langle \mathbf{V} \rangle = \mathbf{U} = (U, 0)$, $U = 1\ m/day$, given by the Darcy law for exponential correlated normal log-hydraulic conductivity with correlation length $\lambda = 1\ m$, and variances $\sigma^2 = 0.1$. The random velocity was numerically generated, as a first order approximation in σ, with the procedure based on the Kraichnan routine, already used in [Jaekel and Vereecken, 1997; Schwarze et al., 2001; Eberhard, 2004; Suciu et al., 2006a,b; Eberhard et al., 2007]. In [Suciu et al., 2006a] it was shown that such velocity fields fulfil the requirements of the limit theorem (on the existence of the Gaussian up-scaling) of Kesten and Papanicolaou [1979].

In every realization of the velocity field the simulation of an isotropic diffusion ($D_{ii} = D = 0.01\ m^2/day$, $D_{ij} = 0$ for $i \neq j$, $i = 1, 2$) was conducted for dimensionless times $t/U\lambda$ corresponding to 3700 correlation lengths, using the reduced fluctuations GRW algorithm illustrated by Figs. 1 and 2. In each realization of the velocity field, $N = 10^{10}$ particles were initially located at the origin of the grid. It was shown that increasing the number of particles between 10^{10} and 10^{24}, the values of the effective coefficients, computed at successive moments over 5000 time steps, are identical in the limit of the double precision [Suciu et al., 2004]. To assess the reliability of the procedure, the simulations were repeated for increasing numbers of periods used in the Kraichnan routine of $Np = 64, 640,$ and 6400. Ensembles of 1024 realizations of the transport simulations were computed for each of the three Np values considered.

The lattice size of $\simeq 10^7$ nodes was larger than the maximum extension of the plume in all simulations. We used a uniform lattice with constant $\delta x = 0.1\ m$, a time step $\delta t = 0.5\ day$ and diffusive jumps in (2) $d = 2$. These parameters are related with $r = 0.25$ accordingly to (5). To reduce the "overshooting" errors in particles methods one impose that the mean displacement in a time step does not overtake a given threshold [Roth and Hammel, 1995]. For the same overshooting, the error of GRW simulations is mainly influenced by the discretization of the velocity, as described by the parameter $U\delta t/\delta x$. It was found that increasing this parameter from 5 (as in the present simulations) to 10 results in differences smaller than 2% [Suciu et al., 2004]. For $U\delta t/\delta x = 5$ and the parameters above, the large scale GRW simulations estimate the observables of the transport in groundwater with errors which are one or two orders of magnitude smaller than the corresponding first-order approximations in σ^2 [Suciu et al., 2005; Suciu and Vamoş, 2006].

The parallelization was trivial since the computation of the Kraichnan velocity field and the simulation of the transport in a given realization were performed on a single processor of an IBM Regatta p690+ computer (the JUMP system at Research Center Jülich, Germany). The costs were considerable, for instance of about 4500 cpu hours for the ensemble of 1024 realizations in the case $Np = 64$. Nevertheless, a comparison with other simulations [see e.g. Schwarze et al., 2001] shows that the costs are 4.4 millions times smaller than they could have been for the PT method.

The effective coefficients of transport in groundwater were estimated through the rate of increase of the second central moment of the plume $D_{ii}^{eff} = \sigma_{ii}^2/(2t)$, which in the large time limit should coincide with the coefficients of the up-scaled Gaussian process. For $Np = 6400$ periodic modes in Kraichnan routine and for an initial solute plume with transverse dimension of 100λ, the root-mean-square deviation of the effective coefficients from the up-scaled coefficients, computed at distances larger than 200λ, was indeed found to be one order of magnitude smaller than the local coefficient D [Suciu et al., 2006a, Fig. 9], as predicted by theory [Kesten and Papanicolaou, 1979; Suciu et al., 2006a].

In the following we present some results concerning the predictive value of the stochastic approach for the contamination in actual aquifer systems, in the case of point instantaneous sources. Following the approach of previous works [Eberhard, 2004; Suciu et al., 2006a] we computed the sample to sample fluctuations of the effective coefficients

$$\left(\frac{\sigma_{D_{ii}^{eff}}}{\left\langle D_{ii}^{eff} \right\rangle} \right)^2 , \text{ where } \sigma_{D_{ii}^{eff}}^2 = \left\langle D_{ii}^{eff\,2} \right\rangle - \left\langle D_{ii}^{eff} \right\rangle^2 .$$

The results presented in Fig. 8 indicate that at large times the fluctuations of the longitudinal effective coefficient tend to zero, i.e. the coefficient is "self-averaging", only when the number of periods in the Kraichnan routine, Np, is of the order of thousands or larger. The self-averaging of the longitudinal coefficients has been also investigated by approximations of the transport equations up to the first order in velocity fluctuations, which were previously evaluated by comparisons with GRW simulations [Suciu at al., 2006b]. It was found that, in order to account for the self-averaging behavior of transport in Gaussian velocity fields, the number of periodic modes in the Kraichnan procedure must be at least as large as the total dimensionless time Ut/λ of the simulation [Eberhard at al., 2007].

The transverse coefficient, shown in Fig. 9, has much smaller fluctuations than the longitudinal coefficient and is self-averaging even for small Np. One can see that $Np = 640$ periodic modes yields practically the same result as $Np = 6400$.

The normalized concentration in given realizations $c(x_1, x_2, t)$ was computed, similarly with (6), as the number of particles in a domain $1\lambda \times 1\lambda$ centered at (x_1, x_2) divided by the total number of particles N. Further, the point concentrations were averaged over a domain of dimensions $1\lambda \times 160\lambda$, oriented across the mean flow. This domain is larger than the transverse dimension of the plume and, consequently the concentration field can be described by an one-dimensional problem for the transverse averaged concentration $C(x_1, t)$. This quantity corresponds to the ideal sampling across reference planes aimed at in field experiments [Vereecken et al., 2000]. Further, We stored the mean concentration at the plume center of mass, $C(\langle x_1 \rangle, t)$, and we computed the mean $\langle C(\langle x_1 \rangle, t) \rangle$ over the ensemble of 1024 realizations of the
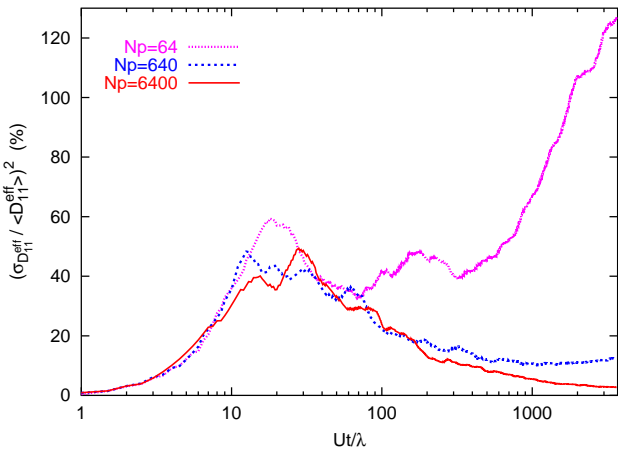
Fig. 8. Sample to sample fluctuations of the longitudinal effective coefficient as function of dimensionless time, number of realizations and number of periods Np, for 1024 realizations of the transport simulation.
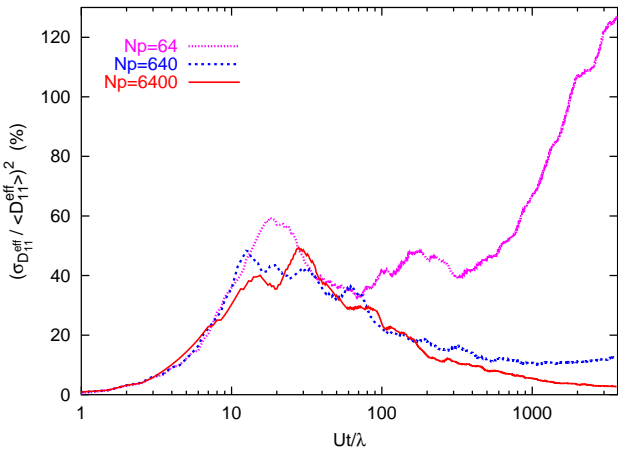

Fig. 9. Sample to sample fluctuations of the transverse effective coefficient as function of dimensionless time, number of realizations and number of periods Np, for 1024 realizations of the transport simulation.

transport. Finally we computed the variance at the center of mass

$$\sigma_c^2(\langle x_1 \rangle, t) = \left\langle C^2 - \langle C \rangle^2 \right\rangle (\langle x_1 \rangle, t)$$

and the coefficient of variation $CV = \sigma_c / \langle C \rangle$. In Fig. 10 one remarks that for $Np = 6400$ and after more than 2000 dimensionless times the coefficient of variation becomes smaller than 10% and shows a monotonous decrease.

The self-averaging behavior of the effective coefficients and cross-section concentration, shown in Figs. 8-10, indicates that single realizations of the transport process behave in the large time limit as the ensemble averaged process. Since the latter tends to an up-scaled Gaussian process, as shown by both theoretical and numerical results, one concludes that the process is asymptotically "ergodic" in a large sense [Suciu et al., 2006a]. Concerning the predictive power of the stochastic up-
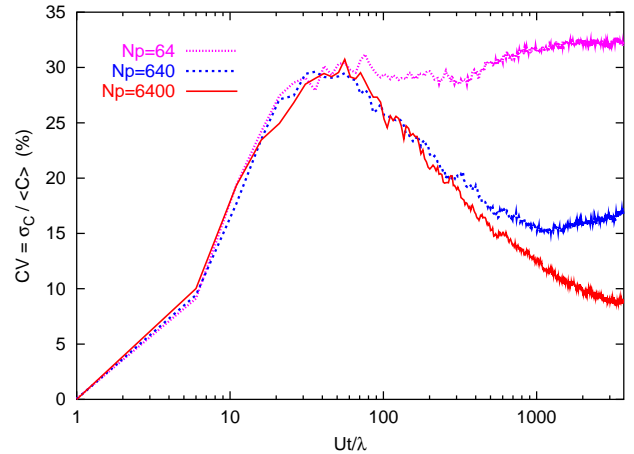

Fig. 10. Coefficient of variation of the cross-section averaged concentration at the plume center of mass.

scaled model, the present results complete and reinforce the conclusions drown by previous works [Schwarze et al., 2001; Eberhard, 2004]: Even under rather ideal conditions ($\sigma^2 = 0.1$), in the case of small initial contaminant plumes the predictions of the up-scaled model are reliable for real groundwater systems only after travelled distances of thousands of heterogeneity scales.

6 Conclusions

The simulated concentration field capture the effects of non-homogeneity and anisotropy of the skin structure. These first results indicate that the simulations can be used in investigations on diffusion and other microscopic processes in biological structures. Comparisons between measured quantities (for instance fluxes of molecules) and GRW simulations could be helpful for the elaboration of more complex codes, able to account for less idealized skin structures.

The stochastic simulations of the advection dominated transport in groundwater, based on GRW procedure, are in good agreement with the exact mathematical result on the existence of the up-scaled Gaussian process. Therefore, we expect that our method could be successfully used to analyze more realistic situations, with larger variance of the log-hydraulic conductivity, for which there are no exact mathematical results. The accuracy of the simulated concentration field and the possibility to describe large scale behavior of the transport process make the GRW method appropriate for simulations of field experiments, such as those presented by Vereecken at al. [2000]. The relatively small computational costs allow the implementation of GRW algorithm in inverse modeling procedures for the assessment of the aquifer's properties.

Though it can be considered as a particular cellular automaton, the GRW procedure differs from most of lattice gas models because no exclusion principle is necessary. As a consequence, the number of particles used in simulations can be as large as the number of molecules

involved in a specific transport problem. This can be a great advantage in simulations for reactive and radioactive decay processes.

The method presented in this paper does not completely remove the overshooting errors associated with the spatial variation of the velocity field. The overshooting can be completely removed with a biased GRW code. The principle of the biased GRW consists in the simulation of the advective displacement by a bias in the random walk jumps [Suciu et al., 2005]. This procedure remains "global", unlike similar approaches [Karapiperis and Blankleider, 1991] where the updated lattice configurations are obtained by sequential random decisions for the jumps of the particles at a lattice site. Since it introduces new constraints, the biased procedure requires larger computational domains. It is yet possible to use the biased GRW algorithm in large scale problems owing to its cellular automaton property which permits domain-decomposition on parallel machines. When domain-decomposition does not increase the computational efficiency, as in case of advection-diffusion simulations for groundwater, biased GRW test simulations can be useful for the calibration and the evaluation of the less expensive un-biased GRW algorithm [Suciu et al., 2006b; Suciu and Vamos, 2006].

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