Simulation of pollutant transport in groundwater with the random walk method

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Abstract Standard finite difference and finite element solution methods of the pollutant transport equation require restrictive spatial discretization in order to avoid numerical dispersion. The random walk method offers a robust alternative if for reasons of calculational effort discretization requirements cannot be met. The method is discussed for the case of an ideal tracer starting out from the Itô-Fokker-Planck equation. Features such as first-order reactions, adsorption with a linear isothermal, double porosity and time dependent dispersivities can be incorporated in a natural fashion. A shortcoming of the method results from the general roughness of simulated distributions in space and time due to statistical fluctuations and resolution problems. The discussed method is applied to a field case of groundwater pollution by chlorohydrocarbons.

NOTATION

\begin{align*}
A & \quad \text{cross-sectional area (m}^2) \\
c & \quad \text{concentration (g m}^{-3}) \quad \text{(index } m\text{: mobile phase, index } im\text{: immobile phase) } \\
D & \quad \text{dispersion tensor with components } D_{xx}, D_{xy}, D_{yy} \quad \text{(m}^2 \text{ s}^{-1}) \\
M & \quad \text{pollutant mass (index } P\text{: particle) (kg)} \\
\Delta M & \quad \text{instantaneously injected tracer mass (kg)} \\
\dot{M} & \quad \text{injection rate of pollutant mass (kg day}^{-1}) \\
n & \quad \text{porosity (index } e\text{: effective; index } m\text{: mobile; index } im\text{: immobile)} \\
N & \quad \text{total number of particles} \\
P & \quad \text{time constant for reaching asymptotic dispersion (days)} \\
Q & \quad \text{pumping rate (m}^3 \text{ s}^{-1}) \\
R & \quad \text{retardation factor} \\
t & \quad \text{time (days)} \\
\Delta t & \quad \text{time step (days)} \\
\vec{u} & \quad \text{vector of pore velocity with components } u_x, u_y \quad \text{(m day}^{-1}) \\
u & \quad \text{modulus of } \vec{u} \quad \text{(m day}^{-1}) \\
x, y & \quad \text{horizontal coordinates (m)} \\
\Delta x, \Delta y & \quad \text{grid-distances in } x\text{- and } y\text{-direction (m)} \\
X, Z_1, Z_2 & \quad \text{uniformly distributed random variable from the interval [0,1]} \\
\alpha & \quad \text{exchange coefficient (day}^{-1})
\end{align*}
INTRODUCTION

Transport models have in recent years been adopted as tools for the prediction of pollutant transport in groundwater as well as for the design of aquifer restoration measures. In spite of its well-known shortcomings the convection-diffusion equation in two horizontal dimensions is made use of in a pragmatic attempt to quantify the large scale movement of pollutants in groundwater.

The transport equation for the ideal tracer (e.g. Bear, 1972) is given by:

$$\frac{\partial c}{\partial t} + u_x \frac{\partial c}{\partial x} + u_y \frac{\partial c}{\partial y} = \nabla \cdot (D \nabla c) + \frac{\partial}{\partial y} (D_{yx} \frac{\partial c}{\partial x} + D_{yy} \frac{\partial c}{\partial y})$$

with

$$D_{xx} = \alpha_L \frac{u_x^2}{u} + \alpha_T \frac{u_y^2}{u} \quad D_{yy} = \alpha_T \frac{u_x^2}{u} + \alpha_L \frac{u_y^2}{u}$$

$$D_{xy} = D_{yx} = (\alpha_L - \alpha_T) \frac{u_x u_y}{u}$$

It represents a partial differential equation of second order of the initial-boundary-condition type. The equation can be solved by standard finite difference and finite element methods. But unless strict and in field cases often prohibitive discretization criteria are adhered to the results are unreliable due to numerical dispersion and/or oscillations. The reason for this behaviour lies in the double nature of the transport equation, convection alone making it a hyperbolic equation, while with dispersion-diffusion alone it would be an easy-to-solve parabolic equation (e.g. Courant & Hilbert, 1968). Which aspect will show more prominently depends on the grid Peclet number defined by:

$$P_e_x = \frac{u_x \Delta x}{D_{xx}} \quad P_e_y = \frac{u_y \Delta y}{D_{yy}}$$

which quantifies the relative importance of convection and dispersion on the level of the single mesh of the discretization scheme with large grid Peclet numbers characterizing the situation where convection dominates and the equation is hyperbolic. Therefore methods from the theory of
hyperbolic equations such as the method of characteristics and particle tracking methods in general are applicable to the solution of the transport equation, the most consequent particle tracking method being the random walk method which represents both the convective and the dispersive transport by individual particle stepping.

THE RANDOM WALK METHOD

The random walk method is a method from statistical physics which has been used in the analysis of diffusion and dispersion processes in porous media for a long time (e.g. Scheidegger, 1954; De Josselin de Jong, 1958). An intuitive approach to the method starts out from the momentary injection of a tracer mass $\Delta M$ into a one-dimensional aquifer of cross-sectional area $A$ and uniform pore velocity $u$. The situation can be described by the analytic solution (e.g. Crank, 1956):

$$c(x,t) = \frac{\Delta M}{2An_e\pi \alpha_L u t^{1/2}} \exp \left( -\frac{(x - ut)^2}{4 \alpha_L ut} \right)$$

For a fixed time $t$ this function can be viewed as a normal distribution around an average $x$-value of $x = ut$ with a standard deviation $\sigma = (2\alpha_L ut)^{1/2}$. The same distribution can be generated stochastically by setting $N$ particles of mass $\Delta M/N$ at time $t = 0$ and location $x = 0$ and moving each particle over a distance $x$ given by:

$$x = ut + Z \cdot (2\alpha_L ut)^{1/2}$$

to its position at time $t$, where $Z$ is a normally distributed random variable with average 0 and standard deviation 1. The resulting path lengths $x$ are normally distributed with average $ut$ and standard deviation $(2\alpha_L ut)^{1/2}$. The frequency distribution $f(x,t)$ obtained in the limit $N \to \infty$ is easily identified with the concentration distribution (3) using a normalization factor:

$$c(x,t) = \frac{\Delta M}{An_e} f(x,t)$$

For a finite particle number $N$, concentrations $c(x,t)$ are determined approximately by discretizing the space variable into intervals of length $\Delta x$, counting the number of particles $n(x - 0.5\Delta x, x + 0.5\Delta x, t)$ falling into the interval of length $\Delta x$ around point $x$ and dividing the accumulated mass by the corresponding water volume:

$$c(x,t) = \frac{\Delta M}{NAn_e \Delta x} n(x - 0.5\Delta x, x + 0.5\Delta x, t)$$
In a non-uniform, unsteady flow field and higher spatial dimensions things are more complicated and it pays to start from the basic theory of random walk which says that the frequency distribution obtained from a random walk with steps of the form

\[ x_p(t + \Delta t) = x_p(t) + u_x \Delta t + Z_1(2\alpha_1 \mu \Delta \tau)^{\frac{1}{2}} \frac{u_x}{u} - Z_2(2\alpha_2 \mu \Delta \tau)^{\frac{1}{2}} \frac{u_y}{u} \]

\[ y_p(t + \Delta t) = y_p(t) + u_y \Delta t + Z_1(2\alpha_1 \mu \Delta \tau)^{\frac{1}{2}} \frac{u_y}{u} + Z_2(2\alpha_2 \mu \Delta \tau)^{\frac{1}{2}} \frac{u_x}{u} \]

with

\[ u_x = u_x(x_p(t), y_p(t), t) \]
\[ u_y = u_y(x_p(t), y_p(t), t) \]
\[ u = (u_x^2 + u_y^2)^{\frac{1}{2}} \]

fulfils in the limit of large particle numbers the Ito-Fokker-Planck equation (Ito, 1951, as quoted in Haken, 1983):

\[ \frac{\partial f}{\partial t} + u_x \frac{\partial f}{\partial x} + u_y \frac{\partial f}{\partial y} = \frac{\partial^2}{\partial x^2} (D_{xx} f) + \frac{\partial^2}{\partial y \partial x} (D_{xy} f) + \frac{\partial^2}{\partial y^2} (D_{yy} f) \]

\[ + \frac{\partial^2}{\partial y^2} (D_{yy} f) \]

Now, the Fokker-Planck equation (8) is not identical to the transport equation (1) if the dispersion coefficients are space dependent (usually because of spatial variations of the flow field). By writing the transport equation in the form:

\[ \frac{\partial c}{\partial t} + u_x \frac{\partial c}{\partial x} + u_y \frac{\partial c}{\partial y} = \frac{\partial^2}{\partial x^2} (D_{xx} c) + \frac{\partial^2}{\partial y \partial x} (D_{xy} c) + \frac{\partial^2}{\partial y^2} (D_{yy} c) \]

\[ + \frac{\partial^2}{\partial y^2} (D_{yy} c) - \left( \frac{\partial^2}{\partial x^2} D_{xx} \frac{\partial c}{\partial y} - \frac{\partial^2}{\partial y \partial x} (D_{xy} c) - \left( \frac{\partial}{\partial x} D_{yx} \frac{\partial c}{\partial y} + \frac{\partial}{\partial y} D_{yy} \frac{\partial c}{\partial y} \right) \right) \]

and using

\[ u'_x = u_x + \partial D_{xx}/\partial x + \partial D_{yx}/\partial y \]
\[ u'_y = u_y + \partial D_{xy}/\partial x + \partial D_{yy}/\partial y \]

(10)

it is seen that the transport equation can be interpreted as a Fokker-Planck equation with a corresponding equivalent random walk of
the form (Uffink, 1985; Kinzelbach & Ackerer, 1986):

\[ x_p(t + \Delta t) = x_p(t) + u_x \Delta t + Z_1 (2\alpha_x u \Delta t)^{1/2} \frac{u_x}{u} - Z_2 (2\alpha_x u \Delta t)^{1/2} \frac{u_y}{u} \]

\[ y_p(t + \Delta t) = y_p(t) + u_y \Delta t + Z_1 (2\alpha_y u \Delta t)^{1/2} \frac{u_y}{u} + Z_2 (2\alpha_y u \Delta t)^{1/2} \frac{u_x}{u} \]  

(11)

The flow field must be supplied in every point of space, a task which is achieved by interpolation in numerical flow models.

The conversion of particle clouds to concentration distributions is done by spatial discretization and counting of particles in the cell. A calculated example is shown in Fig. 1 where the concentration distribution is given due to a permanent source in parallel flow with a pumping well downstream of the source.

The concentration distribution is typically ragged due to the stochastic nature of the method. The computation of significant concentrations requires at least 20 particles in a cell. For smoothness of results, the convective particle step should be a fraction of the typical grid distance. Five to ten steps per cell have proven appropriate (Prickett et al., 1981). The limitation of the convective steps corresponds to fulfilment of the Courant-criterion.

As the equation is linear all results can be obtained by superposition as long as there are no prescribed concentration boundaries. This fact allows a reduction of computational effort under steady flow conditions where plumes from permanent sources can be obtained by convolution from the solution for a Dirac-pulse of relatively few particles.

The derivative terms in equation (10) can be neglected if the flow field varies only gradually. Their contribution is, however, important near
stagnation points or at the boundaries of layers. If, in the example of Fig. 1, the derivative terms are neglected, an unphysical build-up of particles at the stagnation point is observed (Fig. 2). This is due to the fact that particles can diffuse into the surroundings of the stagnation point but can hardly leave it as both the convective and the dispersive particle step vanish at the stagnation point. The counter terms generate velocity components which push particles away from the stagnation point. It should be noted, however, that the calculation of the derivatives requires an accurate flow field in order not to introduce hard-to-estimate new errors. In commercially-used programs such as the code by Prickett et al. (1981), counter terms are neglected thus reducing computation time. In layered aquifers with velocity discontinuities across layers, correction terms can no longer be computed by equation (10). Instead, reflection (Uffink, 1985b) or buffer techniques (Ackerer, 1985) have to be used.

![Stagnation Point Diagram](image)

**Fig. 2** Unphysical build-up of particles at stagnation point in simplified random walk.

**ADVANTAGES OF THE RANDOM WALK METHOD**

The random walk method does not show any numerical dispersion in the classical sense. It is therefore predestined for applications where other methods are, at comparable computational effort, plagued by numerical dispersion. These applications involve typically field problems where the arrival of a pollutant at a well or the fraction of a pollutant plume passing by a well must be determined.

In finite difference and finite element methods numerical dispersion usually spoils results if the grid-Peclet number has to be chosen considerably larger than 2. A situation where standard methods show transverse artificial dispersion if the discretization mesh is much larger than the transverse dispersivity is characterized by strongly anisotropic dispersion and flow at an angle to the cell boundaries. An illustrative example for the latter case is shown in Figs 3 and 4. The plume due to a permanent injection into
parallel flow at an angle of 45° to the coordinate axes is calculated by means of a centred finite difference method and the random walk method. The grid is of mesh size $\Delta x = \Delta y = 50$ m while the dispersivities are $\alpha_L = 100$ m and $\alpha_T = 1$ m with a ratio $\alpha_L/\alpha_T = 100$ which is not unrealistic for the field conditions. Figure 3 shows the artificially widened plume obtained from the finite difference method. At comparable computation time, the random walk method yields a slender plume which agrees well with the analytical solution.

A further advantage of the random walk method is the simplicity with which it can be implanted on top of any flow model. Also, switching dispersion off yields pathlines of the averaged flow field, information which is extremely useful in most field applications.

**PROBLEMS OF THE RANDOM WALK METHOD**

The price paid for the suppression of numerical dispersion is of course the random fluctuation of computed concentrations. Its relative size can be diminished by increasing the number of particles used. As, however, the statistical uncertainty is proportional to the inverse square root of the number
Fig. 4 Permanent source in parallel flow with strongly anisotropic dispersion. Results from random walk method.

of particles in a cell, the increase in the total number of particles does not show to the same degree as an improvement of results. Therefore the random walk method remains — at reasonable computational effort — a relatively crude method as far as the estimation of local concentrations is concerned.

Other causes for fluctuations of computed concentrations exist. One of them is demixing and mixing due to low resolution of the plume by the overlaid grid. An example is shown in Fig. 5. Another phenomenon is unphysical demixing due to insufficient resolution of diverging flow by the particles. It is presented schematically in Fig. 6.

Fluctuating concentrations are disturbing in parameter estimation and in a sensitivity analysis where for parameters with low sensitivity the physical variation of concentrations due to a parameter variation may be masked by fluctuations. Generally, a comparison of measurements with computed results should rather be done on the basis of integral measures such as moments, concentrations in a pumping well or averages of concentrations over a collection of cells than on the basis of local concentrations.

Further problems show in the simulation of particle capture by pumping wells. This is usually done by defining a circle around a well such that a particle which enters the circle is considered as swallowed by the well. The
choice of the radius may influence the breakthrough curve at the well. The arbitrariness of fixing the radius can be partially removed by resolving the flow field to a degree that the velocity vectors in all cells adjacent to the well cell point to the well.

While no-flow boundaries and prescribed concentration boundaries are easily modelled by reflection and by adding or subtracting particles respectively, prescribed total flux boundaries cannot be simulated. Particles which have passed a boundary do not have a chance to return dispersively into the modelled domain.
If standard methods can meet discretization requirements the random walk method is – at the same discretization – no alternative due to its larger computational effort. In two-dimensional regional transport problems, as well as in three-dimensional problems, random walk is, however, often the last resort.

**ADDING ON FEATURES**

**First order chemical reaction**

A first order chemical reaction can be incorporated into a random walk model in two alternative fashions. The first method (Prickett *et al.*, 1981) randomly destroys particles at a probability \( p \) which is related to the reaction rate by the equation:

\[
p = \lambda \Delta t
\]

(12)

The time step must be chosen sufficiently small to guarantee that the probability \( p \) is smaller than 1. The second method assigns to every particle a variable pollutant mass \( M_p(t) \) which develops in time according to first-order kinetics:

\[
M_p(t) = M_p(0) \exp(-\lambda t)
\]

(13)

and which is used in the determination of the total pollutant mass per cell. The latter method is preferable to the first as it prevents a dying out of particles and therefore does not lead to insignificant particle numbers in regions of small concentrations. The same method can analogously be used for the simulation of zero-order reactions. Higher order reactions require the knowledge of the cell concentration in the computation of the development of the single particle mass over a time step. Therefore concentration distributions on the grid would have to be computed after each time step, causing a considerable increase in computation time.

**Adsorption**

Equilibrium adsorption according to a linear isothermal is incorporated by replacing the pore velocity \( \vec{u} \) by a retarded velocity \( \vec{u}/R \) everywhere in equation (1). When calculating total mass it must be remembered that for each particle of mass \( \Delta M \) moving at the retarded velocity \( \vec{u}/R \) there is a shadow mass \( \Delta M(R - 1) \) at the same location adsorbed on the matrix.

**Double porosity aquifer**

Slow adsorption or particle exchange between mobile and immobile pore
water in an aquifer can also be simulated by the random walk method provided the exchange kinetics are linear as given by the equations of transport in a double porosity medium (e.g. Coats & Smith, 1964):

\[
\begin{align*}
\frac{\partial c_m}{\partial t} + n_m(u \cdot \nabla c - \nabla \cdot (D \nabla c)) &= -\alpha (c_m - c_{im}) \\
\frac{\partial c_{im}}{\partial t} &= \alpha (c_m - c_{im})
\end{align*}
\]  

(14)

with an index \(m\) indicating the mobile and \(im\) the immobile phase. Every particle is assigned an additional state variable \(s_p(t)\) which indicates whether the particle resides in the mobile \((s_p(t) = 1)\) or in the immobile \((s_p(t) = 2)\) phase. Besides the convective-dispersive step, the transport step involves a transition between mobile and immobile state according to certain transition probabilities. For notational reasons, only the one-dimensional equations in uniform steady flow are given:

**particle in mobile phase at time \(t\):**

\[
x_p(t + \Delta t) = x_p(t) + u(t)\Delta t + Z(2\alpha_L |u(t)|\Delta t)^{\frac{1}{2}} \quad s_p(t + \Delta t) = \begin{cases} 1 & \text{for } X > p_{12} \\ 2 & \text{for } X \leq p_{12} \end{cases}
\]

**particle in immobile phase at time \(t\):**

\[
x_p(t + \Delta t) = x_p(t) \quad s_p(t + \Delta t) = \begin{cases} 1 & \text{for } X > p_{21} \\ 2 & \text{for } X \leq p_{21} \end{cases}
\]

(15)

where, in contrast to \(Z\), \(X\) is a uniformly distributed random variable in the interval \([0,1]\). The transition probabilities leading to the macroscopic law of exchange used in equation (14) are given by:

\[
p_{12} = \alpha \Delta t h_m \quad p_{21} = \alpha \Delta t h_{im}
\]

(16)

The time step \(\Delta t\) must be chosen such that both \(p_{12}\) and \(p_{21}\) remain smaller than 1. Note that \(p_{12}\) and \(p_{21}\) are usually different, guaranteeing an equal flux in both directions if concentrations in the mobile and in the immobile phase are equal. Figure 7 shows a comparison between the breakthrough curves obtained from random walk simulation and from an analytical solution given by van Genuchten & Wierenga (1976).
Variable dispersivities

It is well known (e.g. Dieulin et al., 1981; Gelhar et al., 1979) that Fickian dispersion is inadequate for the description of macrodispersion. A heuristic approach can model the apparent growth of dispersivities with time or distance. The growth law may be taken from a suggestion by (Taylor, 1922):

\[ \alpha_L(t) = \alpha_L(\infty)[1 - \exp(-t/P)] \]  \hspace{1cm} (17)

The incorporation of growing dispersivities into the random walk method is straightforward. A state variable for every single particle memorizes the time (or the convectively covered distance) since injection of the particle. In the actual time step of a particle, the dispersivity corresponding to the intrinsic particle age is used. For the observed slow variation of the dispersivities counter terms due to the Ito-Fokker-Planck theory are negligible. Figure 8 compares the longitudinal dispersivity growing linearly over a time span of 1000 days from 0 to a value \( \alpha_L = 100 \) m and from simulation with a constant longitudinal dispersivity \( \alpha_L = 100 \) m from the beginning. In both cases the transverse dispersivity is kept constant at \( \alpha_T = 10 \) m. It is interesting to note that the plumes do not differ at the tip. Differences show at the source where the variable dispersivity yields higher concentrations, less upstream dispersion and a sharper drop in concentrations.

APPLICATION TO A FIELD CASE

A random walk model including counter terms from the Ito-Fokker-Planck
theory, a first order decay reaction, and double porosity was applied to the field case of groundwater pollution by chlorinated hydrocarbon solvents depicted in Fig. 9 (Kinzelbach, 1985).

The contaminated aquifer is part of the alluvial sand-gravel aquifer of the Upper Rhine valley. Its thickness lies between 40 and 60 m extending
from the water table to a quasi-impervious base formed by a clay layer. Regional flow is directed from east to west as is indicated by the isopotentials in Fig. 9. It is dominated by infiltration from the River Neckar east of the pollution source. The main pollutant in the plume is 1,1,1-trichloroethane. The modelling is restricted to this substance as it allows a distinct separation of the plume from other plumes in the vicinity. The source of pollution consists of an undissolved reservoir of pure solvent that is concentrated at the top of the aquifer and does not extend more than 5 m into the saturated zone. In the vicinity of the Rauschen Water Works, on the other hand, the plume is probably mixed over the whole depth of the aquifer. Only averages of concentrations over the whole depth are compared to computed results. After the discovery of the plume in 1980 a decontamination measure at the pollution source was taken.

![Diagram](image)

**Fig. 10** Simulated extension of plume in August 1980 as defined by the 5 mg m$^{-3}$ contour.

![Graph](image)

**Fig. 11** Simulated and measured development of concentrations at observation well 1101/305.
eliminating further pollutant supply to the plume. It is assumed, that the contamination started when the polluter first handled 1,1,1-trichloroethane 1400 days prior to the start of the decontamination measure.

The calculated plume at the time of discovery is shown in Fig. 10. Further, the computed breakthrough curve at observation well 1101/305 together with the available measurements is given in Fig. 11. An exchange coefficient \( \alpha = 0.00005 \text{ day}^{-1} \) yields the observed tailing. The mass in the immobile phase of the simulated plume at time \( t = 1400 \text{ days} \) amounts to about one third of the total pollutant mass in the aquifer at that time. The calculation uses 4000 moving particles in a time-varying flow field.

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