Quality of Groundwater, **Proceedings of an International Symposium, Noordwijkerhout, The Netherlands, 23—27 March 1981, W. van Duijvenbooden, P. Glasbergen and H. van Lelyveld (Eds.),** *Studies in Environmental Science*, **Volume 17** 807

GROUNDWATER POLLUTION MATHEMATICAL MODELLING : IMPROVEMENT OR STAGNATION ?

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Between 1953 and 1972, the mathematical modelling of groundwater pollution has actually started, developed and been recognized as a possible and perhaps efficient tool in the description and prediction of pollution behaviour in aquifers. It took advantage of the interest in the study of the fundamental and applied aspects of miscible displacements in porous media (i.e. the characteristics of the flow of substances miscible to water in porous media) expressed by the petroleum research institutes and aimed at improving the recovery of oil. Particularly, it benefited by the introduction of the dispersion-convection equations, the study of the physical meanings of their various parameters, the derivation of numerical solutions (ref. 1). Simultaneously, as modelling was confronted to real field situations and not only to laboratory work, difficulties and even limitations to its use started to appear related among others to the scale effects and the heterogeneities of natural grounds, to the scarcity of data, and also to the mathematics of the models, which, after the quick developments of the late sixties, resulted in the slowing down of the applications of modelling to real field situations. Meanwhile, the problems of groundwater pollution were developing with industrialization, urbanization, agricultural developments, increased uses of groundwater resources and also the growing awareness in the populations and administrations of the necessity of protecting the environment and especially the water resources, adding more questions to the efficient use of modelling and leading to more thinking on the topics. From an analysis of the difficulties of modelling groundwater pollution and a survey of very recent papers on the subject,we examine the present state of modelling concepts and realizations to determine whether the difficulties have been overcome, which models can be expected to work correctly and, generally, whether new developments have occurred or can occur to transform the models of contaminant transport in groundwater into useful tools. Remark : as announced in the title, we restrict our survey to mathematical models, as physical "models" are not models in the sense of classical fluid mechanics i.e. representations of reality at a reduced scale providing numerical information usable at the scale 1:1, due to the fact that laws of similitude do not exist. It should not be understood as an underestimate of the role and significance of physical models,

Reprinted from ***The Science of the Total Environment*, 21 (1981) 283—298**

which are particular and simplified porous media, where some parameters can be easily identified and isolated ; as such, they represent the indispensable basic stages for groundwater pollution modelling, providing the necessary data to approach basic behaviours and mechanisms, giving ideas and confirming intuitions. Carefully handled and interpreted, they often represent interesting means (sometimes the only one) of predicting the qualitative behaviour of a pollution, when no interpretative mathematical model has yet been derived.

I - THE MODELLING OF GROUNDWATER POLLUTION

The modelling of groundwater pollution consists in describing

. the convection of the contaminant, i.e. its movement with the mean flow,

. the dispersion of the contaminant, i.e. its scattering by mixing or spreading around the mean flow,

. the chemical and physico-chemical reactions of the contaminant with the solid matrix of the porous medium,

. the biochemical reactions of the contaminant with its environment, like, for instance, its biodegradation by bacteries,

. the reactions within the contaminant, like, for instance, the decay of a radioactive contaminant,

by means of mathematical tools, like systems of partial differential equations or probabilistic processes, used in well chosen functional or probabilistic spaces representing the porous medium.

While describing the phenomena is certainly interesting as it allows a better understanding of their fundamentals especially when the mathematical model is coupled with a physical model in the laboratory, the modelling of groundwater pollution is mainly aimed at providing a descriptive and predictive management tool, practical for field problems, whose type and resulting accuracy will depend on the considered management objectives and, of course, the socio-economical constraints of the study. Generally, three types of models have been considered :

. black box models, which represent the aquifer as a system without any assumed structure and relate the inputs and outputs of contaminant in the system by means of a very general mathematical formulation, like a convolution relationship or a general mass conservation equation, based on a few and simple working assumptions. They are usually characterized by a transfer function which, combined in some way to the input, yields the output ; its form and numerical values have to be determined by calibration on existing data. These models provide a very global and partial view of what really happens, as they do not account for most of the physical phenomena but globally ; they generally are more descriptive than predictive.

. grey box models, which represent the aquifer as a system equipped with a few structural properties : for instance a sequence of reservoirs with various behaviours. They are also characterized by a transfer function, the form of which is imposed as a consequence of the physical structural assumptions ; the values of its parameters are determined by calibration on existing data. Although closer to the physics of the phenomena than black box models, grey box models are still lacking accuracy as they account for various physical phenomena globally only.

. complete structural models describing the various aspects of contaminant transport listed above with all possible details and predicting the distribution of pollution in space and time. The most usual models of that category have been derived by considering that the convection and the dispersion of the contaminant are best described by a general diffusion equation, while the other phenomena will be accounted for by adding various functions and their derivatives to this diffusion equation.

While the black and grey box models are by definition not adequate to represent the phenomena and mechanisms of groundwater pollution, the complete structural (or dispersion-convection) models are also limited in various ways, which we discuss in the next paragraph.

Remark : the dispersion-convection models describe contaminant transport through media with permeabilities of either interstices or fissures. Karstic areas are seldom simulated, because of the complexity of the flow domains, which cannot generally be averaged into equivalent continuous media and will usually be approached by a case by case qualitative interpretation of tracer experiments. The size and importance of karstic areas in countries like France, Italy or Jugoslavia, for instance, would certainly justify very systematic studies leading to some quantitative mathematical modelling, at least with black or grey box models.

2 - LIMITS OF GROUNDWATER POLLUTION MATHEMATICAL MODELLING

The mathematical description of groundwater pollution mechanisms has various limits, which can be classified into four categories :

. basic adequacy of the models to represent the phenomena,

. scale effects in the determinantion of the parameters of possible models,

. experimental difficulties in the field,

. mathematical and numerical problems.

It should be stressed that this classification is mostly historical and that these categories may concern the same fundamental problems.

2.1. Basic adequacy of the models to represent the phenomena

The general diffusion equation (dispersion-convection equation) used to model contaminant transport in groundwater has been introduced in two stages :

. by various conceptual approaches, continuous media equivalent to homogeneous porous media at laboratory scale were defined and a diffusion equation was derived to represent the dispersion process at laboratory scale, i.e. the scattering of contaminant particles with respect to a mean flow direction, due to molecular diffusion and velocity variations in the pore space.

. the general diffusion equation was then extended to model field situations, by considering the analogy between the dispersion caused by the velocity variations in the pore space and the dispersion caused by large scale heterogeneities in a continuous medium.

Systematic discrepancies between mathematical and experimental solutions in the laboratory, amazing large values and space-variability of dispersion coefficients in field situations have implied that the validity of these representations be questioned at the local and at the large scale along the following lines :

. the conceptual approach of contaminant transport by a general diffusion equation in an equivalent continuum is not valid because the boundary effects which occur at the frontier between the pore space and the solid matrix are not accounted for correctly when replacing a complex medium of pores and solid by a simpler equivalent continuous medium using classical averaging processes. To take care of the influence of the boundaries, some authors modified the general diffusion equation by introducing either molecular diffusion effects in the medium (dead end pores) or singular functions supported by the boundary between pore and solid and averaged as sinks or sources in the medium (distribution theory) (ref. 2).

. it was shown that the spatial averaging of a diffusion equation generally did not yield a diffusion equation, which was confirmed by probabilistic examples showing that a diffusive behaviour did not generally emerge by averaging unless strict conditions on the medium existed (refs 2, 3, 4). The very analogy between dispersion and diffusion, usually admitted at the laboratory scale, was thus questioned.

. the mathematical processes used to establish the dispersion equation for field situations have been introduced since Taylor and Aris by analogy to the processes used to study turbulent diffusion in fluids, by assuming the validity of a diffusion equation to represent dispersion in a continuum at laboratory or local scale and averaging it into another diffusion equation in a continuum at field scale. In particular, it was shown that turbulent diffusion was represented by a diffusion equation for very large times only (asymptotic time) and that the transient states could be represented locally (in a neighbourhood of a given time) by an equation of the same form as a diffusion equation with a time varying diffusion coefficient (refs 5, 6). Applied to dispersion phenomena, these results imply that the dispersion equation will be valid only after some time or, equivalently, after the contaminant has moved and been correctly mixed over some distance, with a time (or space) varying dispersion tensor. Then the field regimes may not be always asymptotic, which could explain the inadequacy of the dispersion equation in that case. For example, this phenomenon may be a hindrance when modelling the protection zones of a well, on rather short distances.

. established in non reactive media and fluids, the dispersion equation generally does not account for chemical,physicochemical or biochemical reactions. Using the possible linearity of the mathematical models and additivity of the phenomena, authors have added various terms to the general diffusion equation, usually as time partial derivatives of the concentration, to represent adsorption on the solid matrix, ion exchanges, radioactive decay and, more generally, the production or disappearance of the contaminant in the various reactions (ref. 7).

. from a physical point of view, the diffusion equation cannot be completely satisfactory because of its mathematical properties : it instantaneously propagates a concentration front from its source to infinity, while, obviously, the contaminant will actually take some time to travel far from its source. It could lead to an underestimate of the amount of contaminant present at a place.

It appears that the adequacy of the representation of contaminant transport and behaviour in an aquifer by a general diffusion equation is mainly limited by problems of mixing and averaging, usually implying the operation of rather large amounts of contaminants with respect to the dimensions of the studied domains. This consideration leads to study the scale effects on the modelling.

2.2. Scale effects in the determination of the parameters and coefficients of possible models

Although already perceived when deriving the dispersion model by averaging microscopic properties into macroscopic parameters, the scale effects have especially been felt when the dispersion equation was applied to field situations and the dispersion coefficients had to be measured. A rather old problem, as it was already mentioned in early works on turbulent diffusion in estuaries and rivers in the fifties, it came out again in the late sixties from laboratory experiments on either consolidated or heterogeneous porous media giving birth among others to a theory of composition of horizontal layered continua into an equivalent continuum (ref. 8) ; when a little later the theory of dispersion started to be applied to groundwater pollution problems, it was noticed that the dispersion coefficients had much larger values in the field than in the laboratory. These results were explained by the variations of local velocities in the aquifer due to heterogeneities ; furthermore it was considered that, in the same way as these heterogeneities would smooth out into an equivalent homogeneous continuous medium for the hydrogeologic properties of the aquifer, at some scale a porous medium homogeneous for dispersion properties could be reached,especially involving constant dispersion coefficients as expected, for instance, by the theory of composition of horizontal layers. But field experiments showed that generally such a limit medium could not be obtained and that the spatial variations of the dispersion coefficients had to be accounted for, which was done in practice by taking constant dispersion coefficients in a neighbourhood of a given scale. Still convenient for the description of an existing pollution at an easily estimated scale, the dispersion model was almost unusable to forecast the behaviour of a new pollution when no information existed about its scales (ref. 3).

The scale effects do not only include the influence of the structure of the porous medium but also the influence of the sizes of the pollution sources as well as the magnitudes of the pollution volumes involved which must be large compared to the dimensions of the studied domain : otherwise the contaminant may travel through preferential paths, i.e. layers of relatively high permeability, generating fingering without mixing instead of a diffusion regime ; averaging processes are then meaningless and a dispersion equation is not adequate to describe this transport at the scale of the aquifer. This incomplete change of scale from the various layers of the aquifer to the aquifer itself is analogous to the non obtention of the asymptotic regime mentioned above. These effects are of particular significance when describing the transport of a dangerous contaminant in small quantities, which will travel through preferential paths so that it will be useless to define an average movement even if the aquifer has well known mean characteristics : this is the case of toxic contaminants, of microbiological pollutants. The limits of a useful modelling of their behaviour are then obvious as this modelling relies on a good knowledge of the heterogeneities of the aquifer.

Another scale effect, of importance in groundwater management of rather large domains, concerns the continuous injection and transport of large amounts of contaminant on long distances (for instance, non point source pollution in regional problems) : the transition zone between the contaminant and the non polluted water may become negligible with respect to the travelled distance. Then the second order terms of the general diffusion equation may well be neglected and this equation will reduce to a first order partial differential equation or convection equation (ref. 3).

2.3. Experimental difficulties in the field

The experimental difficulties in the field are mostly related of course to the scale problems. As no similitude exists between physical models and the field, experiments have to be performed at the scale 1:1 in situ, usually with some tracers. It implies either the operation of large amounts of tracers if an averaging effect is sought or the setting of refined devices to picture the distribution of flow heterogeneities otherwise.

In the first case, when the behaviour of the contaminant is assumed to obey a diffusion equation, the spatial variabilities of the dispersion coefficients furthermore imply the use of experimental settings allowing the measurements of tracer concentrations at several distances from the injection location. They will mainly consist of either wells or surface geoelectrical devices, all systems presenting various inadequacies : wells are expensive, their drillings to the specifications of the experiments are usually difficult, their equipments for concentration measurements are not always easy to handle (especially for continuous recording), moreover they may considerably disturb the groundwater flow ; geoelectrical devices are not very sensitive and integrate all ground resistivity variations.

In the second case, the distribution of flow heterogeneities in an aquifer is usually sought pointwise with systems of wells and experimental techniques like tracer point dilution involving all the difficulties mentioned above when using wells plus the fact that the description may be quite incomplete because of the local character of the heterogeneities. Sometimes, geoelectrical methods can be introduced, with the help of cumbersome mathematical techniques, like deconvolution, to obtain detailed information from global results.

In general, also, time sequences over large spatial areas will be difficult to obtain and to program and to correlate.

Thus, while compulsory, field experiments are expensive and difficult to realize, especially when the aim is to predict the behaviour of a pollution which has not yet occurred, as in the case of the setting of protection zones for wells.

2.4. Mathematical and numerical problems

A result of the experimental difficulties encountered in the description of contaminant behaviour in groundwater is the scarcity of data. Thus the mathematical modelling will have to take this factor into account, and, particularly, it will appear in many cases that a sophisticated mathematical model is meaningless if the number and quality of data are not sufficient. The choice of a mathematical representation should then depend on its sensitivity to errors on the data and to missing data, which means that the use of a black box, a grey box or a complete structural model will depend on the existing data and the possible new data that can be further collected, as well as on the quality of the interpolations used to obtain continuous (and even twice continuously differentiable) functions from the discrete experimental data.

Besides, all types of models present mathematical and numerical difficulties due to their mathematical structures and, also, to the types of chosen numerical treatments :

. The possible non-linearity of the general diffusion equation has been mentioned for a long time. It occurs when the model has to account for the density variations of the groundwater with contaminant intrusion, the velocity field and hence the dispersion coefficients depending upon the concentration distribution. As this situation results from the migration of large amounts of heavy pollutants, it is not frequently found in the field, with the exception, of course, of salt water intrusions of all kinds. The numerical treatment of the equation is then based on local linearization and the use of iterative procedures, generally more intuitive than rigorous.

Aside from the mathematical difficulties related to partial differential equations in general, either non linear or linear, numerical difficulties specific to the treatment of diffusion and convection equations arise, like numerical diffusion and overshoot. Numerical diffusion results in a smearing of the computed concentration front, while overshoot is characterized by oscillations in the computed concentration profiles. These difficulties have proven to be considerable hindrances to the use of dispersion or convection models as forecasting tools, the alternative usually being either the elimination of numerical dispersion by introducing other methods than finite differences or finite elements, like the method of characteristics, generally cumbersome to operate or the keeping of simpler numerical methods while introducing numerical devices to reduce or at least evaluate the numerical dispersion.

The determination of dispersion coefficients from field experiments is an inverse problem. Although theoretically solved in the case of a Cauchy problem from a theory of pseudo differential operators (refs 2, 3), it has never been introduced as such in practice, like the problem of determining transmissivities from piezometric data. Trial and error methods have been used instead, which can be time consuming and are not rigorous.

. The black box models are usually of the convolution type and their uses as forecasting tools implies the determination of their transfer function from experiments by a deconvolution procedure. All difficulties related to ill-posed problems, like the lack of unicity and the instabilities, then appear.

2.5. Some remarks

The limitations to mathematical modelling listed above have been formulated before 1972, and sometimes as early as the late fifties, by mechanicians of fluids studying turbulent diffusion in pipes and channels, by petroleum engineers and research scientists interested in oil recovery and thermal convection, eventually by hydrogeologists and mechanicians of fluid wishing to represent contaminant behaviour in aquifers, as illustrated by the few following examples :

. For instance, although the spatial variability of the dispersion coefficients and the scale effects have been systematically emphasized in the late sixties when field pollution problems had to be treated, the large field values of the dispersion coefficients already attracted the attention of scientists working on estuary flow in the fifties who found averaging formulas justifying the magnitudes of the numerical values.

. Also, the possible partial inadequacy of the general diffusion equation to represent contaminant transport in groundwater at local scale accurately, related to various inconsistencies in the derivation of an equivalent continuous medium of a porous medium, has been pointed out rather early. Some improvements have been suggested then and also errors have been estimated in order to allow the use of the diffusive models anyway in the laboratory.

. The emergence of a diffusive regime with a constant dispersion coefficient after the dispersion process has taken place for some time, i.e. the asymptotic time concept, was already recognized by Taylor, who listed the necessary assumptions for such an emergence. In the late sixties, it was discussed by various authors, mainly in the context of diffusion in open channels, who studied the early stages of the dispersion process, trying to model them and to estimate minimum asymptotic times (refs 5, 6). Simultaneously, other authors, working on flow through porous media, did show that there existed conditions under which the averaging of equations describing the transport of matter in groundwater at the pore scale did not yield a diffusion equation (refs 2, 3, 4).

3 - RECENT STUDIES AND RESULTS

The limitations listed above have prevented the development of groundwater pollution modelling, the most important limitation certainly being the spatial variability of the dispersion coefficients involving a quasi-impossibility to measure them in the field for most cases. Since 1972, when these limitations were described and tentative explanations proposed, investigations have been carried on, fostered by the appearance of various groundwater pollution problems to be accounted for in water resources management and aimed at improving the modelling of groundwater pollution. These investigations mainly concern

. the changes of scales, involving the possible derivation of a diffusion equation, the spatial variability of the dispersion coefficients, the description of the early stages of the dispersion process ;

. the improvements of the numerical solutions of the general diffusion equation ;

. the derivations of black and grey box models ;

. the experimental research in the laboratory and in the field resulting from new groundwater pollution problems and from improved theoretical approaches to modelling.

3.1. Changes of scales

Theoretical studies have been recently performed to test the representativity of the general diffusion equation as a model of the dispersion process at field scale and, not surprisingly, have concluded that a diffusive regime does not necessarily govern the dispersion. At least three different papers have treated this problem in 1980 :

. Analyzing longitudinal dispersion data in open channel or pipe flows, Chatwin (ref. 9) discusses the assumptions made by Taylor in his approach of dispersion, stressing the consequences on modelling when these conditions are not met i.e. the occurrence of a non diffusive regime, illustrated by non-gaussian curves. The author does not propose a new equation but a quantitative assessment of the deviations of observed concentration profiles (versus space at a fixed time, or versus time at a fixed point) from gaussianity by series of Hermite polynomials depending on various moments of the concentration distribution.

. Studying a bidimensional flow in a horizontally stratified medium, characterized by a constant horizontal velocity per stratum, Matheron (refs 10, 11) uses a stochastic process to describe the movement of tracer particles in that flow. This movement is the sum of a convective movement with the velocity in the stratum and a dispersive movement, assumed to be fickian and isotropic ; the expectation of the concentration at a point and at a given time is the probability density function of a particle at that point and at that time. Taking the x-axis along the velocity direction, Matheron computes the variance of the coordinate $x(t)$ of a particle as a function of the vertical variations of the velocities : a gaussian process being classically characterized by a variance linearly proportional to time, Matheron, defines the mathematical conditions on the variance to obtain a diffusive regime. Justifying the derivation of such a regime by authors like Marle (refs 8, 1) or Gelhar (ref. 12) who make the right assumptions for that, he shows that in many cases there is no reason for a diffusive regime to exist even for very large times : this is particularly the case of stratified flow with horizontal velocities in an infinite bidimensional medium, where no mixing occurs by convection.

. Applying the old concept that dispersion in porous media is due to the variations in the velocity distribution and is therefore related to the spatial heterogeneity in permeability of the aquifer, Smith and Schwartz (ref. 13) develop a stochastic process consisting of the deterministic motion of tracer particles with the flow velocities in a macroscopically heterogeneous permeability field combined by addition to a random motion of these particles accounting for anisotropic microscopic dispersion. The aquifer is taken as a realization of a stochastic process by Monte Carlo techniques, and its permeability field is generated by a random variable with a probability density function at each point in the flow domain. The spatial variance of a set of tracer particles is computed and expressed as a function of time in each realization and the ratio of the number of realizations for which the variance varies linearly with time to the total number of realizations is also given, as a useful indication of a possible diffusive behaviour of the dispersion process. On the basis of 300 realizations, the authors state that less than 20 % were characterized by a diffusive behaviour and attribute this result to the fact that in most cases the particle velocities do not undergo a sufficient number of changes for a correct application of the central limit theorem ; in particular the occurrence of preferential paths will prevent sufficient mixing. A conclusion is that the modelling of field situations with a diffusion equation and large values of the dispersion coefficients is generally uncorrect, except if the dimensions of the domain and of the dispersion process are large enough to ensure sufficient spatial averaging.

It can be noticed that this approach is analogous to Matheron's and was partly proposed, at least with regard to the generation of permeability fields, but not applied by de Marsily (ref. 11).

These studies confirm two main ideas : a dispersion process is not necessarily of the diffusive type ; the emergence of a diffusive regime is related to a sufficient spatial averaging, conditioned by good mixing. Under the assumptions of the emergence of a diffusive regime, studies have been performed to describe the early stages of a dispersion process and to evaluate the asymptotic time i.e. the time necessary for a diffusion regime to take place. The knowledge of this time is necessary for a correct scaling of the field experiments aimed at measuring dispersion coefficients, as the dimensions of the experimental domain should at least be equal to the product of the mean velocity by this time.

. Analyzing unsteady convective diffusion in two dimensional open channel turbulent flows with a given initial concentration distribution of solute, Lee and Gill (ref.14) develop a generalized dispersion theory which models the early stage of the dispersion process by an equation relating the time derivative of the concentration to an infinite series composed of the space derivatives of the concentration at all orders with time dependent coefficients ; in particular, the first two terms of the series are dominant, which provides an approach to a diffusion equation, the first coefficient being an average velocity of the solute cloud and the second coefficient a time varying dispersion coefficient. This model tends towards Taylor's model with constant coefficients for large times, which are estimated by the authors from the depth of the channel flow and the area average of the eddy diffusivity in the vertical direction.

. Studying a bidimensional flow in a horizontally stratified medium with a horizontal velocity constant in each layer, Gelhar et al (ref. 12) use a classical method of turbulence analysis (as did previously Bear or Fried), the Reynolds decomposition and averaging, to obtain an equation to the perturbations in concentrations pointing out a Reynolds term $\overline{u'c'}$ (expectation of the product of velocity and concentration perturbations). The spectral analysis of this equation, particularly of the Reynolds term, results in relating this flux to a mean concentration gradient by a "dispersion coefficient" which is the product of the mean velocity by a time dependent dispersivity, This dispersivity tends towards a limit for large times, which implies the emergence of a diffusive regime and the possible estimate of the asymptotic time.

. Deepening Matheron's approach, mentioned above, for the same stratified medium, Matheron and de Marsily (ref. 10) assume a flow direction no longer parallel to the stratification and show that a diffusive regime will then emerge when time tends to ∞ under rather weak assumptions. It means that to model the dispersion process by a diffusion equation for finite times is not correct ; yet an approaching model could be introduced, as an equation of the diffusion form but with a time varying dispersion coefficient, with the following meaning : if $t \rightarrow D(t)$ is the dispersion coefficient as a function of time, $D(t_1)$ is the value to be used from 0 to t_1 in the diffusion model in order to obtain the best spatial description of the contaminant concentration at t_1 . The authors also compute some dispersion coefficients at various finite times and for $t \rightarrow \infty$ for some usual aquifer characteristics, discussing the significance of tracer tests on finite distances related to the accuracy of the dispersion coefficient estimate.

It appears that most recent theoretical studies concern the change of scale from an already macroscopic continuous medium (as defined by Bear or Fried) to a more macroscopic continuous medium and not the basic problem of the derivation of the dispersion equation at macroscopic level from pore size (microscopic) level (of course, with the exception of some papers in the line of (ref. 15), whose theoretical proposals are not supported by experience). It means that the medium is always considered as continuous in the sense that all variables (like concentrations or velocities) are defined everywhere and that, for some authors, the dispersion process is even described by a diffusion equation at the lower scale. It explains why research on turbulent diffusion in open channels may also lead to results directly applicable to flow through porous media, by an application of good mixing and spatial averaging in the equivalent continuous porous medium analogous to turbulent mixing in fluids.

3.2. Numerical solutions

Under the assumption that the diffusion equation models the dispersion process, some of the numerical problems mentioned above have been studied, mainly overshoot and numerical diffusion.

Although the method of characteristics proved to be free of numerical diffusion, it was seldom used because of the complexity of its programming and authors mostly investigated the possible decreases of the numerical diffusion using dispersion correction terms in the numerical expressions either with finite differences or finite elements with mitigated success (refs 16, 17, 18, 19). A simplified adaptation of the method of characteristics was successfully tested by Migault (ref. 16) and could prove quite useful.

Work on overshoot has been simultaneously performed, mainly on Galerkin finite element schemes, essentially based on possible reductions of space or time increments and then limited by problems related to the solution of large size systems of linear equations (refs 16, 17, 18, 19). In the simple one dimensional case, a combination of finite elements and method of characteristics for the convective part of the diffusion equation has been tested with some success, both for numerical dispersion and overshoot (ref. 20).

It should be mentioned that some work has also been done to produce easily usable numerical solutions in the form of type curves, completing Ogata and Banks as well as Emsellem type curves, and based on analytical solutions of the diffusion equation with simple boundaries and boundary conditions and on numerical solutions for well tests (radial flow) (ref. 21).

These solutions are claimed to be free of numerical dispersion, by a choice of implicit - explicit weighting coefficients which cancel the extra second order derivatives of the limited Taylor expansion of the concentration function.

Already discussed by Fried for a Cauchy problem (refs 2, 3), the dependance of the uncertainty of the concentration function on the uncertainties of the dispersion equation parameters has been again recently examined, again showing a relative stability of the solution of the equation (ref. 22).

Finally some attempts have recently been made to solve the general inverse problem (i.e. the determination of dispersion coefficients from concentration distributions) essentially by an optimization method (ref. 23), while a new deconvolution method was introduced in a black box modelling of contaminant transport in groundwater, as explained below (ref. 16).

3.3. Black or grey box modelling

The limitations in the use of the general diffusion equation for the various reasons mentioned above (inadequacy of the equation related to the dimensions in space and time of the proposed study, unpredictable dispersion coefficients, sparse or unaccurate data) have involved the use of black box models based on the derivation of transfer functions by deconvolution methods and of course applied under the assumption that the modelled system is linear and stationary.

Although most of the work concerning deconvolution has been performed for the determination of the hydraulic parameters of the aquifers, the necessities of modelling transport (to describe new laboratory work, for instance concerning motion with physico-chemical interactions between the contaminant and the solid matrix (ref. 24), or field problems like the transfer of contaminants between a river and its alluvial aquifer (ref. 25)) have implied some theoretical work on deconvolution within the frame of groundwater pollution modelling and the development of an original method by Migault (ref. 16), based on the best approximation of the transfer function by a linear combination of functions generated by translating the input concentration function in a convenient Hilbertspace, the matrix system thus obtained being then solved by successive projections on a sequence of subspaces.

To operate a black box model, a preliminary study of the consistency of the input and output data must be made, especially to analyze the time sequences, when the modeller does not have complete control of the experimental setting and procedures. Statistical methods have been used, especially spectral analysis, with success to take care of possible missing or unadequate data (ref. 25).

Grey box models, either distributed parameter models, or lumped parameter models, have been conceptually developed before 1972. From the extensive review made by Mary P. Anderson (ref. 26), they appear to have been used with some improvements especially concerning the taking into account of biochemical and physico chemical reactions, after 1972.

3.4. Experimental research

Since 1972, numerous groundwater pollution situations have been described with the help of existing models. The results usually emphasize the limitations of these models, involving the development of the theoretical considerations which we have shortly reviewed. Furthermore, specific experiments have been performed mainly to measure the various pollution motion parameters, such as velocities or dispersion coefficients, which have usually displayed the heterogeneous character of even the most homogeneous aquifers and the spatial variability of the dispersion coefficients. Besides, experimental research has been performed in the laboratory to better understand the modelling of pollution movement within a reactive medium, especially the adsorption of the contaminant on the solid matrix (refs 16, 24). These experiments have not generally involved a new mathematical modelling, with respect to the general models introduced by Bear but have helped to provide a better physical basis to the modelling. Worth mentioning in that line, is the extensive experimental work made by Zilliox et al on groundwater pollution by hydrocarbons and aimed at modelling the boundary and source conditions representing the porous medium impregnated by oil above the water table as light or miscible parts of that oil body migrate into the aquifer (ref. 27).

The existing mathematical models do not describe the behaviour of bacterias and laboratory work is now starting on the subject, relating to bacteria properties as a possible biodegradation factor of some pollutions.

4 - CONCLUSIONS

The elaboration of groundwater pollution modelling relies on the solution of two fundamental problems :

i) the derivation of a continuum equivalent to the pore-solid system, i.e. a medium where all functions are defined everywhere and where the complex boundary between void space and solid space vanishes, such that boundary conditions will only appear on the rather simple frontier of the equivalent continuum. The model is then made up of mathematical equations valid in the whole medium to represent the concentration distributions everywhere and accounting for the influence of the solid space on the contaminant motion. It should be noticed that it is a basic problem of the mechanics of continuous media, with a physical meaning.

ii) in the continuum then obtained, the derivation of an equation representing large scale phenomena, especially integrating the influences of the hydraulic heterogeneities of the continuum. It is a conceptual problem, without direct physical meaning.

Problem i) with very few exceptions has not been studied since tentative solutions had been given before 1972, emphasizing two aspects : spatial averaging of a diffusion equation does not necessarily yield a diffusion equation ; the behaviour of the contaminant particles at the boundaries between pore and solid spaces differ from their behaviour in the pore.

The emphasis was put on problem ii) i.e. on the derivation of a diffusion model for field situations (macrodispersion equation) assuming that, at the laboratory scale or local scale, the porous medium is continuous and the corresponding mathematical model of concentrations is a diffusion equation (microdispersion). The problem differs from the above mentioned basic problem i) as it only concerns the averaging, by some regularizing method, of the diffusion equation in a heterogeneous but continuous medium. Progress has been made there, resulting in an improved representation of the so-called spatial variability of the dispersion coefficients. Working with considerations already expressed by people like Taylor, Chatwin or Gill for turbulent diffusion in open channels, authors have made clear the assumptions under which contaminant motion in aquifers could be described or not by a diffusion equation, particularly estimating quantitatively, in the first instance, the asymptotic time i.e. the time necessary for the dispersion process to be sufficiently well described by a diffusion equation. These results stress the limitations of the use of the dispersion equation for field modelling, especially for the determination of dispersion coefficients by tracer experiments. As a corollary, they provide guidelines for the setting and dimensioning of new field tests. Yet all these results have not yet been much tested experimentally.

The dependance of the characteristics of contaminant motion on the hydraulic heterogeneities of the aquifer, a rather old notion, has been stressed again, which justifies the research program of laboratories, like the Institut de Mécanique des Fluides de Strasbourg, partly concentrated on the study of aquifer heterogeneities, by local velocity measurements for instance. It appears that in most pollution problems, when forecasting is a primary objective and no stabilized pollution plume exists for model calibration (as, for instance, when establishing protection zones for wells), the emphasis should be put on the detection and description of flow heterogeneities, like preferential paths. In particular, the description of the motion of contaminants which occur is small amounts, like toxic materials or trace elements, still presents the same difficulties as formerly and is not yet modelled. The improvement of field and mathematical techniques to describe and predict these heterogeneities is a commanding factor of the future improved applicability of complete structural models, like the general diffusion equation for large times or stochastic motion models for the early stages of the dispersion process.

As it does not require a refined knowledge of the medium structure, black box modelling has also been given new attention in field problems, for instance to estimate residence times of a contaminant in the medium. New deconvolution techniques have been introduced and more systematic data analyses by stochastic methods proposed.

Work on the numerical solution of the diffusion equation has been going on, resulting in various analytical solutions and computer programs, with improved accuracy due to a better treatment of numerical diffusion and overshoot. Recent programming has been made in finite differences as well as finite elements and also by introducing the method of characteristics with some variations.

Finally, physical models in the laboratory are now developed again (cylindrical columns for instance) to get a better insight of biochemical or physico-chemical reactions added to the transport of contaminants, especially in what concerns adsorption and desorption of various chemical materials and the general behaviour of microbiological elements.

In brief, to answer the question raised in the title of this paper, it appears that although some progress has been made in the understanding of the modelling of field conditions and the significance of the parameters and their so called variability, which could be of help for setting and scaling field experiments, the practical applicability of the diffusion equation to field problems as a forecasting tool is not insured while other possible models, like stochastic motion models, proposed to simulate the situations when the diffusion equation does not hold, are yet too cumbersome to operate. The classical field methodology correcting the deficiencies of the complete structural models by non numerical considerations on the lithology, geology, morphology of the medium, by the use of black or grey box modelling and by pure kinematic considerations is still very practical.

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