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AN ANALYTICAL MODEL FOR MULTIDIMENSIONAL TRANSPORT OF A DECAYING CONTAMINANT SPECIES

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ABSTRACT

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A mathematical model is developed for a finite source that incorporates one-dimensional groundwater velocity, longitudinal and transverse dispersion, and some form of decay for either radionuclides or biodegradable organics. The model is employed in a calibration procedure that permits the determination of up to seven parameters from a known spatial distribution of concentrations. The determined parameters include the velocity of the contaminant, source size and concentration, and up to three dispersivities for a three dimensional problem. For a biodegradable organic, a combined parameter incorporates the ratio of velocity to a degradation half-life and is readily determined from the field data.

INTRODUCTION

The difficulty in obtaining physically-based parameters from data provided by contamination histories lies in the preponderance of unknowns. In the most simple case for common inorganics, up to eight parameters have to be determined to understand and model the transport. These include the source dimensions for a finite source, the source concentration, the velocity of the groundwater, which requires further information on effective porosity, the time since the contaminant first entered the groundwater, and up to three dispersivities for a three-dimensional problem. This simple scenario assumes that no chemical reactions are taking place. In the presence of reactions, the problem is magnified in that both dispersion and reaction tend to decrease the prevailing concentrations and it is difficult to separate the influence of each. When dealing with organics, additional problems arise. Chemical attenuation, biodegradation, and dispersion all act to lower the concentrations. The mapped chemical enclave will reflect these processes but will not reveal the absolute or even relative influence of each. Inverse methods are of some help here but sometimes are misleading in that values must be assigned to some of the parameters in order to determine the others. If errors are involved in the

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assignment of values, then errors will be incorporated in the "calibrated" parameters. The solution, then, is non-unique and often without physical basis.

A transport model recently published by Domenico and Robbins (1985) purportedly offered some techniques by which at least some of these problems may be removed. This model allows for a calibration procedure whereby all of the relevant unknowns are determined directly in an independent fashion from a mapped chemical enclave. The model has since been applied to determine the regional dispersivity of selected basalt flows at the Hanford site, Washington (LaVenue and Domenico, 1986), the Babylon plume in Long Island, N.Y. (Kelley, 1985), and a chloride plume at the Idaho National Engineering Laboratory (Fryar, 1986). Recently, the model was independently checked against a three-dimensional finite element code with rather good results (Fig. 1). The correspondence between these two models is in agreement with comparisons against other models as reported in the original manuscript.

In this paper, the model will be modified to account for a decaying species. As expected, this will require some alterations of the calibration procedure. The decaying species may be a radionuclide or a biodegradable organic, provided the biodegradation follows similar laws as established for radioactive decay.

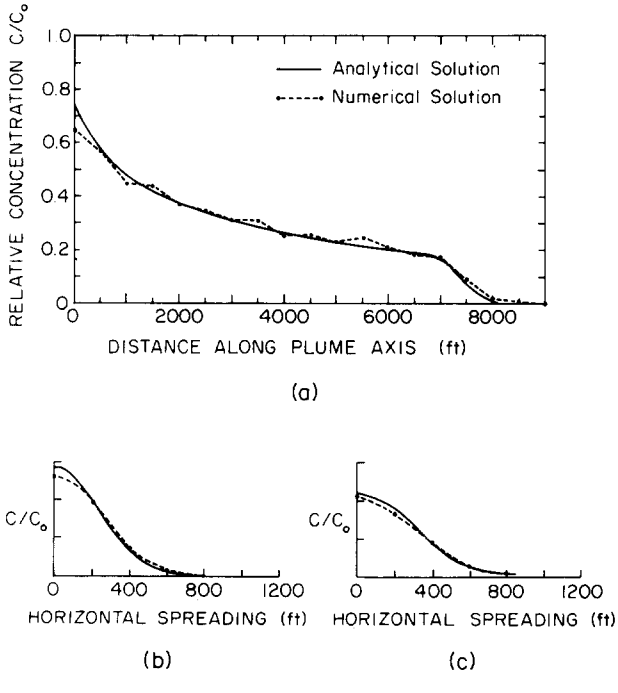

Fig. 1. Comparison of finite element result with analytical model: (a) along plume axis; (b) at 1070 m from source; and (c) at 1680 m from source (after Collette, 1985).

THE EXTENDED PULSE MODEL FOR TRANSPORT WITH DECAY

The original transport model described above was referred to as an "extended pulse" type model in that it was derived by an infinite spatial extension of a well known instantaneous finite source pulse model. Certain modifications, more heuristic than rigorous, were then incorporated to arrive at the final workable form. Any modification of this result must also take on heuristic overtones in that an exact solution to this problem cannot avoid some form of numerical integration, which negates a calibration procedure where several variables are involved (e.g., Domenico and Robbins, 1985). Fortunately, an approximate solution may be arrived at purely on the basis of physical arguments pertaining to the transport process. Figure 2 will help in developing the pertinent physical ideas.

Figure 2a demonstrates the familiar plug flow model void of all longitudinal and lateral mixing. The source material enters the flow tube at a source concentration C_0 and immiscibly displaces the original fluid in the tube. The displacement or advective front has progressed to the position $x = vt$ where v is the velocity of the displacing fluid and t is the time since the process started. The concentrations are at steady state and this steady state is everywhere equal to the source concentration C_0 in that neither lateral nor longitudinal dispersion has been permitted. This, of course, is not a realistic field or laboratory model, but it is the starting place to describe various dispersion models.

Figure 2b illustrates a plug flow model with longitudinal dispersion. Here the material enters the flow tube and, in the absence of lateral dispersion, mixes with and displaces the original fluids in the x direction strictly within the flow tube. It is noted now that the concentration at $x = vt$ is less than the original concentration C_0 in that longitudinal dispersion causes some of the material to move ahead of the advective front (vt) at the expense of material behind the advective front. However, at some distance behind the advective front, the concentration takes on a steady-state value equal to the source concentration C_0 and thereafter possesses all the characteristics of the plug flow model. This, again, is a non-realistic field condition, but one that has been observed in the laboratory in one-dimensional (axial dispersion) tests. The situation depicted in Fig. 2b is described completely by the so called Ogata–Banks equation (1961) which, in abbreviated form, is expressed as:

$$C_x = (C_0/2)\,[\mathrm{erfc}(x - vt)/2(\alpha_x vt)^{1/2}] \tag{1}$$

where C_x is the concentration at any distance x, C_0 is the source concentration, v is the velocity of the constituent, commonly taken as the velocity of water, t is the time since the experiment began, and α_x is the longitudinal (axial) dispersivity.

Figure 2c is yet a third case which can be referred to as plug flow with transverse spreading (lateral dispersion). Three things are noteworthy here. First, in the absence of longitudinal dispersion, no material travels beyond the advective front. Second, with transverse spreading, the material for the first time is not restricted to a single flow tube but can spread in a direction

transverse to the flow. This condition accounts for the third feature of transverse spreading models where the concentrations are everywhere at a steady state and the steady state is everywhere less than the source concentration, except of course, at the source where $x = 0$. Concentrations less than C_0 are readily achieved in that the mass is equally distributed throughout the chemical enclave, but the volume which occupies the mass increases with increasing distance from the source. Near the source, the mass within a given vertical plane is more concentrated in that it occupies a smaller area (or volume). At some distance x far from the source an equal amount of mass in a given plane occupies a large area (or volume). As concentration is mass/volume, the concentration decreases with distance from the source.

Domenico and Palciauskas (1982) provide a solution to the transverse spreading situation of Fig. 2c. Their model is formulated as a boundary value problem that approximates the spreading of a contaminant plume in both the

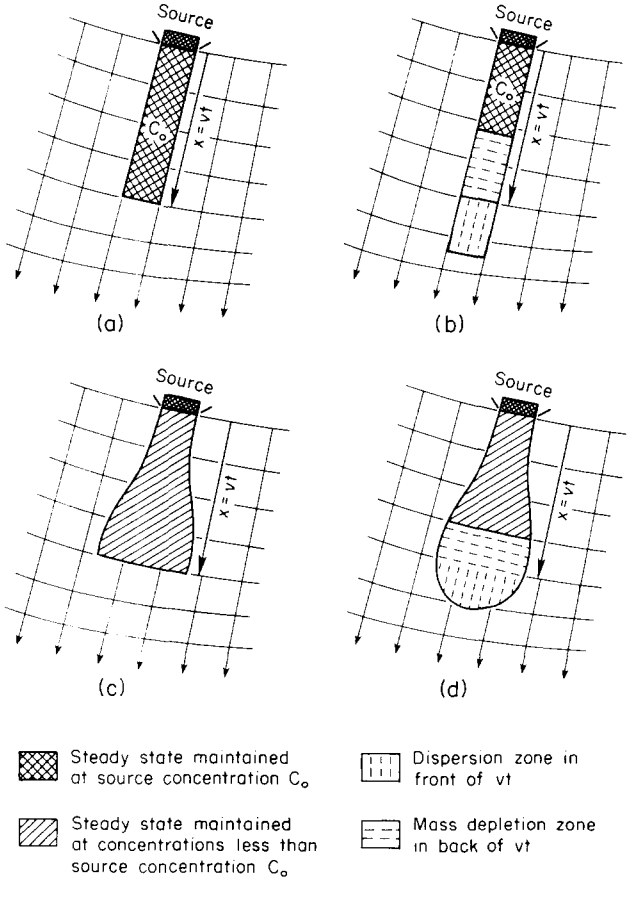

Fig. 2. Idealization of the dispersion process showing: (a) plug flow; (b) longitudinal dispersion; (c) transverse dispersion; and (d) longitudinal and transverse dispersion.

horizontal (y) and vertical (z) directions perpendicular to the prevailing flow path in x. The solution is:

$$C(x, y, z) = (C_0/4)\,\{\mathrm{erf}\,[(y + Y/2)/2(\alpha_y x)^{1/2}] - \mathrm{erf}\,[(y - Y/2)/2(\alpha_y x)^{1/2}]\}\,\{\mathrm{erf}\,[(z + Z/2)/2(\alpha_z x)^{1/2}] - \mathrm{erf}\,(z - Z/2)/2(\alpha_z x)^{1/2}]\} \quad (2)$$

where Y and Z are the source dimensions. If spreading in z can be ignored, the four becomes a two and the error function terms in z are ignored. If spreading takes place in only one direction in z, say vertically downward, $Z/2$ is replaced by Z. This is likely the most common spreading geometry and corresponds to a finite source at the water table in a very thick aquifer. The maximum concentration occurs along the centerline ($y = z = 0$) and is given as:

$$C(x, 0, 0) = C_0\,\mathrm{erf}\,[Y/4(\alpha_y x)^{1/2}]\,\mathrm{erf}\,[Z/4(\alpha_z x)^{1/2}] \quad (3)$$

Note that at $x = 0$, $C = C_0$. Equation (3) is currently in use by the U.S. Environmental Protection Agency to evaluate delisting petitions to exclude certain wastes from the hazardous waste list (Federal Register, 1985).

Finally, Fig. 2d demonstrates a more realistic chemical enclave as might be expected in a field situation. Because of an assumed one-dimensional velocity, this may be referred to as a plug flow model with both longitudinal and transverse spreading (dispersion). Note that this enclave has features common to both the longitudinal dispersion model (Fig. 2b) and the transverse spreading model (Fig. 2c). There is a frontal dispersion zone beyond the advective front caused exclusively by longitudinal dispersion, and a resulting mass depletion zone behind the advective front, this latter zone providing the mass in the frontal portions of the plume. At some distance behind the advective front, depending largely on the value of the longitudinal dispersivity, the enclave is in a steady state. The steady-state concentrations are everywhere less than the source concentration because of transverse spreading, except of course at $x = 0$.

The essential features of this enclave can be captured by combining the longitudinal dispersion and transverse spreading models given in eqns. (1) and (2), or:

$$C(x, y, z, t) = (C_0/8)\,\mathrm{erfc}\,[(x - vt)/2(\alpha_x vt)^{1/2}]\,\{\mathrm{erf}\,[(y + Y/2)/2(\alpha_y x)^{1/2}] - \mathrm{erf}\,[(y - Y/2)/2(\alpha_y x)^{1/2}]\}\,\{\mathrm{erf}\,[(z + Z/2)/2(\alpha_z x)^{1/2}] - \mathrm{erf}\,[(z - Z/2)/2(\alpha_z x)^{1/2}]\} \quad (4)$$

Equation (4) is the extended pulse model of Domenico and Robbins (1985), developed here on the basis of physical arguments as opposed to mathematical arguments employed in the original development. Note that for x less than vt such that the argument of the complementary error function approaches negative two, the steady-state transverse spreading model is recovered, eqn. (2). At the source boundary $x = y = z = 0$, the product of the error functions equals

four and, for times greater than zero, the argument of the complementary error function rapidly approaches negative two, so that the source concentration at the source boundary is maintained at or near C_0 for times greater than zero. For different spreading geometries, the arguments regarding the transverse spreading model apply here as well.

Similar physically based arguments can now be provided to ascertain the manner in which decay may be incorporated in this relatively simple result. The essential features of such an enclave can be captured if the longitudinal dispersion described in Fig. 2b and eqn. (1) can be expressed in terms of both dispersion and continuous decay, and then combined with the transverse spreading component described in eqn. (2). Given such a result, the constituents residing in each vertical plane of the enclave will be characterized by the same amount of decay, with those closest to the source being affected the least, and those furthest from the source being affected the most. The plug flow model with both longitudinal dispersion and continuous decay has been described by Bear (1979):

$$\begin{aligned} C(x, t) &= (C_0/2) \exp \{x/2\alpha_x[1 - (1 + 4\lambda\alpha_x/v)^{1/2}]\} \\ &\quad \operatorname{erfc} \{[x - vt(1 + 4\lambda\alpha_x/v)^{1/2}]/2(\alpha_x vt)^{1/2}\} \end{aligned} \tag{5}$$

where λ is the decay constant or 0.693 divided by the half-life. Note that if the decay constant is zero, the Ogata–Banks (1961) expression is recovered. The extended pulse approximation for a decaying species then becomes:

$$\begin{aligned} C(x, y, z, t) &= (C_0/8) \exp \{x/2\alpha_x[1 - (1 + 4\lambda\alpha_x/v)^{1/2}]\} \\ &\quad \operatorname{erfc} \{[x - vt(1 + 4\lambda\alpha_x/v)^{1/2}]/2(\alpha_x vt)^{1/2}\} \\ &\quad \{\operatorname{erf} [(y + Y/2)/2(\alpha_y x)^{1/2}] - \operatorname{erf} [(y - Y/2)/2(\alpha_y x)^{1/2}]\} \\ &\quad \{\operatorname{erf} [(z + Z/2)/2(\alpha_z x)^{1/2}] - \operatorname{erf} [(z - Z/2)/2(\alpha_z x)^{1/2}]\} \end{aligned} \tag{6}$$

The centerline concentration is expressed as:

$$\begin{aligned} C(x, 0, 0, t) &= (C_0/2) \exp \{x/2\alpha_x[1 - (1 + 4\lambda\alpha_x/v)^{1/2}]\} \\ &\quad \{\operatorname{erfc} [x - vt(1 + 4\lambda\alpha_x/v)^{1/2}]/2(\alpha_x vt)^{1/2}\} \\ &\quad \{\operatorname{erf} [Y/4(\alpha_y x)^{1/2}] - \operatorname{erf} [Z/4(\alpha_y x)^{1/2}]\} \end{aligned} \tag{7}$$

When the argument of the complementary error function approaches negative two, the steady state is achieved

$$\begin{aligned} C(x, y, z) &= (C_0/4) \exp \{x/2\alpha_x[1 - (1 + 4\lambda\alpha_x/v)^{1/2}]\} \\ &\quad \{\operatorname{erf} [(y + Y/2)/2(\alpha_y x)^{1/2}] - \operatorname{erf} [(y - Y/2)/2(\alpha_y x)^{1/2}]\} \\ &\quad \{\operatorname{erf} [(z + Z/2)/2(\alpha_z x)^{1/2}] - \operatorname{erf} [(z - Z/2)/2(\alpha_z x)^{1/2}]\} \end{aligned} \tag{8}$$

Equation (8) reduces to the original version of the steady state when the decay constant is zero. Although there appears to be a contradiction of terms when

a steady state can be achieved for a decaying species, the condition can be realized if the source is continually renewed by new material.

Finally, the steady-state concentration along the centerline is expressed as:

$$C(x, 0, 0) = C_0 \exp \{x/2\alpha_x[1 - (1 + 4\lambda\alpha_x/v)^{1/2}]\} \times \operatorname{erf}[Y/4(\alpha_y x)^{1/2}] \operatorname{erf}[Z/4(\alpha_z x)^{1/2}] \tag{9}$$

A CALIBRATION PROCEDURE

In this section a calibration procedure for determining the pertinent coefficients and parameters from field data is discussed. For simplicity, spreading in the z direction will be ignored although incorporating this particular feature is relatively straightforward. Because the expressions for error functions, exponents, and complementary error functions are long and cumbersome, we will use the following abbreviations:

$$\left.\begin{aligned} \operatorname{erf}(y_i) &= \operatorname{erf}[(y_i + Y/2)/2(\alpha_y x_i)] - \operatorname{erf}[(y_i - Y/2)/2(\alpha_y x_i)^{1/2}] \\ \exp \beta_i &= \exp (x_i/2\alpha_x\{1 - [1 + (4\lambda\alpha_x)/v]^{1/2}\}) \\ \operatorname{erfc} x_i &= \operatorname{erfc} \{x_i - vt[1 + (4\lambda\alpha_x)/v]^{1/2}\}/2(\alpha_x vt)^{1/2} \end{aligned}\right| \tag{10}$$

where the i designates a specified point in the field.

The first problem is to determine the transverse dispersivity α_y and the source size Y. The ratio of two field concentrations at the same x distance from the source but at different y coordinates may be expressed from eqn. (6) in terms of the notation above:

$$C(x_1, y_1, t_1)/C(x_1, y_2, t_1) = \operatorname{erf}(y_1)/\operatorname{erf}(y_2) \tag{11}$$

This result can be iterated immediately for the source size Y and the dispersivity α_y. A second iteration at two different points with a common x distance from the source will provide for unique values of α_y and Y. This is demonstrated in Fig. 3 from the results of three concentration ratios with distances from the source of 4000 cm, 6000 cm, and 8000 cm. The unique values are $\alpha_y = 10$ cm and $Y = 1000$ cm. The procedure is identical to that described for a non-decaying species (Domenico and Robbins, 1985). Examples of this procedure can be found in Domenico and Robbins (1985), Kelley (1985), LaVenue and Domenico (1986), and Fryar (1986).

The next step in the calibration procedure requires that two points be selected in the near field where it is reasonably certain that steady state has been achieved. The points are at two different distances x from the source. The concentration ratio for the stated steady-state concentration expressed in eqn. (8) is:

$$C(x_1, y_1, t_1)/C(x_2, y_2, t_1) = \exp \beta_1 \operatorname{erf}(y_1)/\exp \beta_2 \operatorname{erf}(y_2) \tag{12}$$

This can be further expressed as:

$$(C_1/C_2) \operatorname{erf}(y_2)/\operatorname{erf}(y_1) = N = \exp \beta_1/\exp \beta_2. \tag{13}$$

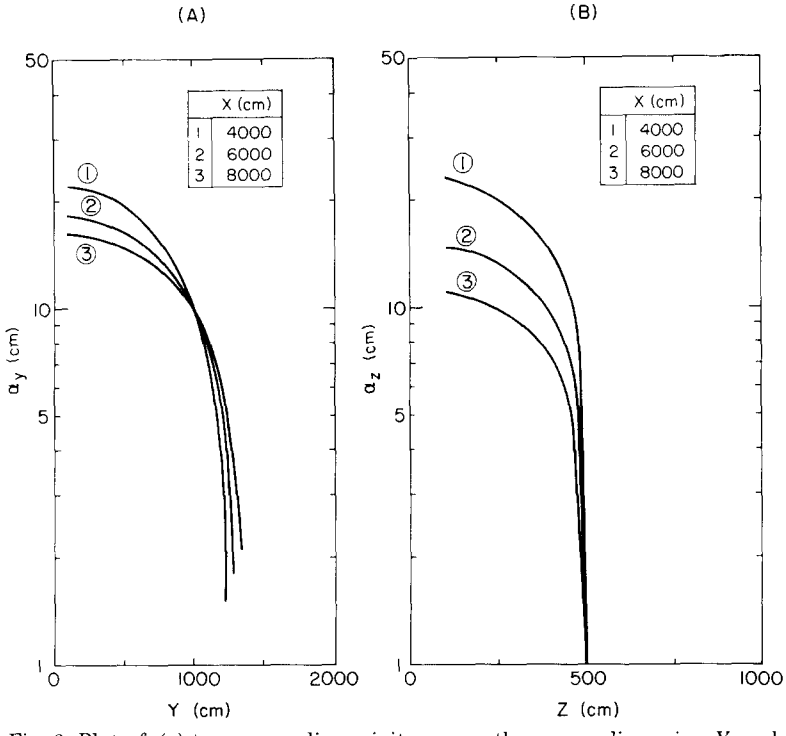

Fig. 3. Plot of: (a) transverse dispersivity versus the source dimension Y; and (b) the transverse dispersivity versus the source dimension Z (after Domenico and Robbins, 1985).

where N is some known number. Normally, N will be close to one or possibly one or two orders of magnitude smaller than one. Substituting for the exponent terms in eqn. (10), eqn. (13) may be expressed as:

$$\alpha_x[\ln N/(x_1 - x_2)]^2 - \ln N/(x_1 - x_2) = \lambda/v \tag{14}$$

In that the first bracketed quantity is a very small number considerably less than one, it is seen that this relationship (and the exponential terms in the solution) are insensitive to α_x. This is not totally unexpected in that steady-state concentrations have been observed to be independent of longitudinal dispersivity (Prakash, 1982). This is further demonstrated with the following example. At a distance of 1600 m for $\lambda = 0.057$ and $v = 250\,\mathrm{m\,yr^{-1}}$, the exponent term is 0.6966 when $\alpha_x = 2$ m, and 0.7019 when $\alpha_x = 100$ m, for a difference of 0.76%. At a distance of 100 m for the same fixed parameters the exponent term is 0.9777 and 0.9781, respectively, for a difference of 0.04%. Ignoring the term involving α_x, eqn. (14) is expressed:

$$v/\lambda = (x_2 - x_1)/\ln N \tag{15}$$

where v is the velocity of the contaminant, and will be equal to the velocity of the groundwater only for unretarded contaminants, and N is given in eqn. (13). For a radioactive species, λ is known and the velocity may be calculated

directly. For a biodegradable organic where λ is unknown, only the combined parameter v/λ is determined here, and other methods are required to obtain the individual parameters.

Given the information above, the source concentration C_0 may be determined with the steady-state eqn. (8) where everything is now known except for α_x. However, any reasonable value of α_x can be placed in the exponential terms (say α_x = three or four times α_y) and the exponential terms will be insensitive to that value.

With the procedures given above, the following may be determined: (1) the source dimension Y; (2) the lateral dispersivity α_y; (3) the velocity of the contaminant v for a radionuclide (or λ/v for a biodegradable organic); and (4) the source concentration C_0. The remaining parameters include time and longitudinal dispersivity. It is convenient now to express a concentration ratio for concentrations along the centerline (x, 0, 0), where the ratio is expressed as the actual concentration, eqn. (7), to the maximum concentration, eqn. (9). This gives:

$$C/C_{\max} = \tfrac{1}{2}\,\mathrm{erfc}\,\{[x - vt\,(1 + 4\lambda\alpha_x/v)^{1/2}]/2(\alpha_x vt)^{1/2}\} \tag{16}$$

For any $C/C_{\max}$ less than one, the enclave is in the unsteady state, and the longitudinal dispersivity is readily determined if the time t is known. A more simple approach is to determine the distance x where $C/C_{\max}$ equals 0.5 so that:

$$x = vt(1 + 4\lambda\alpha_x/v)^{1/2} \tag{17}$$

where all parameters are known except for α_x.

CONCLUSIONS

The methodology presented in this paper may be useful in the analysis of radionuclide contamination or organic transport with biodegradation, where λ is the biodegradation "half-life" value. The calculations are straightforward and easily programmed for microcomputer analysis, and the model can be manipulated to account for several spreading geometries. Most importantly, information on most of the relevant parameters can be obtained from the concentration distribution. As all the information is taken directly off the plume, the method can be applied to a chemically retarded species without any regard to retardation coefficients. Given data at different points in time, field retardation coefficients can be ascertained.

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