Simultaneous identification of a single pollution point-source location and contamination time under known flow field conditions

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Abstract

A theoretical framework is presented that allows direct identification of a single point-source pollution location and time in heterogeneous multidimensional systems under known flow field conditions. Based on the concept of the transfer function theory, it is shown that an observed pollution plume contains all the necessary information to predict the concentration at the unknown pollution source when a reversed flow field transport simulation is performed. This target concentration $C_0$ is obtained from a quadratic integral of the observed pollution plume itself. Backwards simulation of the pollution plume leads to shrinkage of the $C_0$-contour due to dispersion. When the $C_0$-contour reduces to a singular point, i.e. becomes a concentration maximum, the position of the pollution source is identified and the backward simulation time indicates the time elapsed since the contaminant release. The theoretical basis of the method is first developed for the ideal case that the pollution plume is entirely known and is illustrated using a synthetic heterogeneous 2D example where all the hydro-dispersive parameters are known. The same example is then used to illustrate the procedure for a more realistic case, i.e. where only few observation points exist.

Keywords: Point-source pollution; Backward transport; Pollution source identification; Location probability

1. Introduction

Remediation of polluted groundwater is today a major hydrogeological challenge. An important issue in any remediation process is the assessment of groundwater contamination. This involves understanding the dynamics of the aquifer system in which the contamination is propagating. A crucial aspect within such an assessment and remediation process is to identify unknown contamination sources and to understand the release histories that led to pollution. This is important both for the design of adequate remedial measures and for identification of responsibilities, since remediation is usually a costly process.

The issue of pollution source identification has been extensively investigated in the past decades, on the one hand by geochemical fingerprint techniques [(e.g.) 13,23] and on the other hand, by the use of mathematical and simulation approaches. These approaches focus either on the reconstruction of contaminant release histories [(e.g.) 4,10,12,24–27,29] or on the identification of unknown pollution source locations and contamination times [9,16,21] and on historical contaminant distributions [14]. Atmadja and Bagtzoglou [3] have presented a state of the art report on the mathematical methods that have been developed in the field of source identification. They subdivided the existing methods into four major groups, namely optimisation [(e.g.) 11,12,27], analytical [(e.g.) 1] and direct methods [(e.g.) 4,29] as well as probabilistic and geostatistical approaches [(e.g.) 5,16,26,28]. Each of these methods is subject to significant drawbacks and limitations, such as parameter homogeneity, simple flow geometries, high sensitivity to noise, previous knowledge of potential source locations and/or contamination time and history, or excessive computational effort. The source identification

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problem inherently suffers from under-determination due to the loss of information induced by hydrodynamic dispersion in combination with model and measurement errors. Therefore, restrictive assumptions are inevitable to tackle the problem and differ according to the angle from which the problem is addressed. Only few authors have dealt with the source identification problem in multidimensional heterogeneous systems, as for instance Aral et al. [2], Michalak and Kitaniidis [14], Neupauer and Wilson [21] and Neupauer and Lin [15]. In their examples they assume complete knowledge of the velocity field and the transport parameter distributions, as well as the boundary conditions.

The present work follows the line of approaches that address the identification of an unknown single point-source location and contamination time by solving the advection-dispersion equation with a reversed flow field, but keeping the dispersion part positive (i.e.) 5,16,21,28.

In this context, the work of Neupauer and Wilson [16,17] is particularly relevant. They combine travel time and location probability density functions as used in the transfer function theory [11] with an adjoint method for conservative tracers. Subsequently, they extended their approach to transient hydraulic conditions [18] and first-order decay and sorption [19,20], and applied their method to a real case site [21]. Recently, Neupauer and Lin [15] introduced measurement errors and estimation of the source mass in their adjoint method by conditioning the backward probabilities on measured concentrations. A major advantage of the adjoint method is its numerical stability and the reduction of simulation runs, since one single backward simulation can substitute multiple forward simulations. In the present work, the assumptions of perfect knowledge of the flow field conditions and hydro-dispersive parameters are made. The concept of location probability has been extended in this work, yielding both the point-source location and the contamination time with a single reversed flow field simulation. Although simulation of the ‘physical’ shrinkage of a pollution plume is not possible, the proposed method leads to shrinkage of a characteristic concentration iso-contour of the pollution plume during backward simulation, run with the observed plume concentrations as initial condition. This characteristic, or target concentration \( C_0 \) is obtained from the concentrations in the pollution plume and is predicted \textit{a priori}, making use of the concepts of location probabilities. As soon as the target concentration iso-contour reduces to a single point during the reversed flow field simulation, i.e. becomes a concentration maximum, the pollution source location is identified and the simulator clock indicates the time elapsed since contamination took place.

The theoretical approach is shown to yield unique solutions for the single point-source location and contamination time under perfectly known flow field conditions with a single reversed flow field simulation. In reality, a perfectly known flow field and hydro-dispersive parameters will never occur, since data sets are mostly scarce and contain errors. Since the issue of model uncertainty is inherent to any simulation approach of real-world cases (e.g.) 22 and parameterisation is not the topic of this work, we have focused on the development of the theoretical source identification framework. However, the fact that a unique solution is obtained under perfectly known hydro-dispersive conditions may provide a useful criterion with which uncertain parameters could be estimated in a similar way as it was done by Wagner [27] but without prior knowledge of the source location and the contamination time. Although not explored in the present work, a potential future path to follow with this method would therefore be to combine it with inverse methods.

Apart from the problems linked to model parameterisation, the most severe restriction of the presented approach is the assumption of a single point-source pollution with a conservative contaminant. In a pollution plume with different origins, the concepts of the location probability functions cannot be applied in a straightforward way, unless the superimposed signals from the different pollution events can be separated into different fractions with different origins.

In the first section, the mathematical framework is presented, leading to the relationship between the target concentration and the observed pollution plume. The approach is illustrated using a 2D heterogeneous example for the ideal condition that the entire pollution plume is known. Then, the performance of the method is illustrated for the more realistic case where only few observation points exist.

2. Identification of a single pollution source location and time

In this section, the framework for the proposed source identification procedure is developed for heterogeneous multidimensional systems. For this purpose, the main features of the transfer function theory that are used to develop the proposed approach are summarized. According to the transfer function theory [11], the resident concentration distribution resulting from advective-dispersive transport of an instantaneous unit mass-release at any given time, reflects a location probability density function (pdf). The location pdf indicates the fractions of the initial mass-release that are locally found, per unit of pore volume, in the vicinity of any point. Depending on the modelling objectives, forward and backward (or reversed) flow fields can be applied, yielding complementary information as to forward and backward advective-dispersive mass fractionation.

Assuming for simplicity a divergence-free flow field and non-reactive transport, the forward location pdf \( F_q \) with an initial unit-mass input at a point \( x_0 \) is a solution of the transport equation

\[
\phi \frac{\partial F_q}{\partial t} = -q \cdot \nabla F_q + \nabla \cdot (\phi \nabla F_q), \quad F_q(x_0, 0) = \frac{\delta(x - x_0)}{\phi(x_0)}
\]
and the backward pdf $F_{-q}$ with an initial unit-mass input at a point $x_1$ is a solution of the transport equation with a reversed flow field [16,17]

$$\frac{\partial F_{-q}}{\partial t} = \mathbf{q} \cdot \nabla F_{-q} + \nabla \cdot (\phi \mathbf{D} \nabla F_{-q}), \quad F_{-q}(x_1, 0) = \frac{\delta(x - x_1)}{\phi(x_1)}$$

(2)

with the respective boundary conditions

$$\nabla F_{-q} \cdot \mathbf{n} = 0$$

(3)

In (1)–(3), $\mathbf{q}$ is the flow vector, $\phi$ the porosity, $\mathbf{D}$ the dispersion tensor and $\mathbf{n}$ is a unit vector normal to the domain boundaries. The Cartesian coordinates in 1, 2 or 3 dimensions are symbolized by $x$, with the subscripts denoting the different release locations. The upstream mass-release location for forward transport is denoted $x_0$, while $x_1$ denotes the downstream mass-release location for the reversed transport process. The forward location pdf is denoted $F_q(x, t)$ and the backward pdf $F_{-q}(x, \tau)$, where $\tau$ designates backward flow time.

As was formally shown by Neupauer and Wilson [16,17] and later by Cornaton and Perrochet [6,7], the forward pdf $F_q$ evaluated at a downstream location $x_1$ after $t_1$ of forward transport in response to a unit mass-release at an upstream location $x_0$ is identical to the backward pdf $F_{-q}$ evaluated at the upstream location $x_0$ after $\tau_1$ of backward transport in response to a unit mass-release at $x_1$. This leads to the very useful and fundamental property

$$F_q(x_1, t_1) = F_{-q}(x_0, \tau_1)$$

(4)

Eq. (4) is valid for any flow field of arbitrary heterogeneity and macro-dispersive properties and is schematically illustrated in Fig. 1, where the location probability densities $F_q(x_1, t_1)$ and $F_{-q}(x_0, \tau_1)$ are identical, while the two pdfs as such are very different.

According to Jury and Roth [11], if a mass $M_0$ is released at the point $x_0$, the resident concentration $C(x, t_0)$ observed after $t_1$ of forward advective-dispersive transport, is related to the forward location probability density $F_q(x, t_1)$ by

$$C(x, t_0) = M_0 F_q(x, t_1)$$

(5)

The key to the proposed source identification approach lies in the combination of (4) and (5). The relationship given in (4) applies to every single point $x$ within a pollution plume $C(x, t_1)$, given that the pollution plume results from an instantaneous contamination at the point $x_0$ at $t = 0$. To elucidate the concept of the approach, the initially released mass $M_0$ is assumed to be known and only a single observation point $x_1$ with a resident concentration $C(x_1, t_1)$ is considered. The relationship between the forward and backward location pdfs given in (4) indicates that the concentration at the initial release location $x_0$ will be identical to $C(x_1, t_1)$ if the same initial mass $M_0$ were released at $x_1$ and the flow field reversed during the same time-span $t_1$, corresponding to $\tau_1$ of backward flow time, as shown in Fig. 1.

Now, instead of releasing $M_0$ at the downstream observation point $x_1$ in a reversed flow field, the mass that is locally found at $x_1$ and at $t_1$ is released, namely $M_1 = \phi(x_1)C(x_1, t_1)dx_1$, where $dx_1$ is a small control volume around the point $x_1$. Considering that (5) also applies to backward transport, the concentration at the unknown release location $x_0$ after $\tau_1$ of backward transport is

$$C(x_0, \tau_1) = M_1 F_{-q}(x_0, \tau_1)$$

(6)

By evaluating (5) at the point $x_1$, the released mass $M_1$ in (6) can be expressed as a function of the mass $M_0$ that was initially released at $x_0$ by

$$M_1 = \phi(x_1)C(x_1, t_1)dx_1 = \phi(x_1)M_0 F_q(x_1, t_1)dx_1$$

(7)

Introducing (7) into (6) yields, at the original upstream release location $x_0$, the concentration

$$C(x_0, \tau_1) = M_0 \phi(x_1)F_q(x_1, t_1)F_{-q}(x_0, \tau_1)dx_1$$

(8)

Taking advantage of the relationship given in (4), (8) simplifies to

$$C(x_0, \tau_1) = M_0 \phi(x_1)F_q(x_1, t_1)^2 dx_1$$

(9)

Normalized by the initial mass $M_0$, $C(x_0, \tau_1)$ expresses a joint location probability density. Hence, the product $\phi(x_1)F_q dx_1 F_{-q} dx_0$ is the joint probability to find, in the pore space $dx_0$ around the point-source after $\tau_1$ of backward flow time, a particle initially released at $x_0$, given that this

Fig. 1. Three contours of the forward and backward location probability density functions. The forward pdf $F_q(x, y, t_1)$ results from a unit mass-release at $(x_0, y_0)$ and the backward pdf $F_{-q}(x, y, \tau_1)$ results from a unit mass-release at $(x_1, y_1)$. The forward pdf evaluated at $(x_1, y_1)$, $F_q(x_1, y_1, t_1)$, and the backward pdf evaluated at $(x_0, y_0)$, $F_{-q}(x_0, y_0, \tau_1)$, are identical, located on the same bold iso-contour, according to (4).
particle first reaches the intermediate location \( x_1 \) after \( t_i \) of forward flow time.

Upon substitution of (5), again evaluated at the point \( x_1 \), into (9), the concentration at the point-source location \( x_0 \) after \( \tau_i \) of backward flow time can be expressed as a function of the plume concentration \( C(x_1, t_i) \) by

\[
C(x_0, \tau_i) = \frac{1}{M_0} \phi(x_1) C(x_1, t_i)^2 \mathrm{d}x_1
\]  

Eq. (10) reveals the very useful and simple relationship between the concentration that will be detected at the (yet unknown) pollution source \( C(x_0, \tau_i) \) after \( \tau_i \) of backward flow time and the concentration in the pollution plume \( C(x_1, t_i) \). Hence, running a backward transport simulation with the initial condition \( M_1 \delta(x - x_1)/\phi(x_1) \) allows to follow the evolution of the iso-contour of the predicted point-source concentration \( C(x_0, \tau_i) \) during backward flow time. By virtue of (10), the point-source must be located somewhere on this migrating iso-contour.

Running a reversed flow field simulation from only one point in the pollution plume does not yield a unique solution, unless \( M_1 \) is released at the concentration maximum of the plume. To obtain a unique solution for the pollution source location and contamination time with a single reversed flow field simulation, use can be made of the property that the relationship given in (4) applies to all points in the domain.

3. Pollution source identification under ideal conditions

In an ideal point-pollution case, where the entire pollution plume \( C(x, t_i) \) is perfectly known, a unique solution for \( x_0 \) and \( \tau_0 \) is obtained, making use of all the information contained in the plume.

Conceptually, the predicted concentration contour \( C(x_0, t_i) \) as derived from (10) can be back-tracked for any point \( x_1 \) in the pollution plume. Hence, theoretically, several reversed flow field simulations can be run, each one for a different point. Then, the point-source has to be located on the one and only intersection point of all the backward migrating \( C(x_0, t_i) \)-contours, and the time when all these contours intersect in a single point corresponds to the contamination time. An easy way to identify this singular intersection point is to apply the principle of superposition. Instead of running a reversed flow field simulation for a large number of points separately, one single simulation can be performed with the entire plume concentration distribution as initial condition. Then, according to the principle of superposition, the concentration at the unknown point-source \( x_0 \), denoted target concentration \( C_0 \), is simply the integral of the predicted concentrations \( C(x_0, \tau_i) \) in (10) over all possible release locations \( x_1 \), i.e. over the entire flow domain \( \Omega \). This yields the fundamental result

\[
C_0 = \frac{1}{M_0} \int_{\Omega} \phi(x) C(x, t_i)^2 \mathrm{d}x
\]  

in which the dummy subscript 1 has been removed. Eq. (11) shows that the target concentration \( C_0 \) is entirely defined by the integral of the square of the local resident concentrations \( C(x, t_i) \) in the plume observed at \( t_i \) (i.e. the initial condition for the backward transport process), weighted by the local porosity and scaled by the total mass. \( C_0 \) can also be seen as a mass-averaged plume concentration.

Eq. (11) allows a priori prediction of the target concentration \( C_0 \) that will be detected at the unknown pollution source location \( x_0 \) after \( \tau_0 \) of backward flow time, and with the pollution plume as initial condition. Given that the contamination time is unknown too, the unique solution is obtained by back-tracking the migrating and shrinking \( C_0 \)-contour until it becomes a concentration maximum, which corresponds to the point of disappearance.

The above theoretical findings have two implications with respect to simultaneous identification of both the contaminant point-source \( x_0 \) and the time \( t_i \) that has elapsed since the mass-release. Given a perfectly known flow field, the only computational requirement is a classical, advective-dispersive simulation with reversed flow field of an initial contaminant plume \( C(x, t_i) \), a process in which numerical stability is well controlled by any standard simulation code. Secondly, given that the target concentration contour \( C_0 \) shrinks during its backward migration until it reduces to a point at \( x_0 \), the simulation is simply run until the maximum concentration reaches this value. When this is achieved, the simulator clock indicates the time \( \tau_i \) corresponding to the contamination time \( t_i \).

To illustrate this approach, a synthetic 2D horizontal, heterogeneous aquifer is considered. The software FEFLOW [8], offering the options of flow reversal and direct evaluation of (11), is used in the analysis. The domain dimensions are 1000 m \( \times \) 400 m, discretised into 100,000 quadrilateral finite elements of size 2 m \( \times \) 2 m. The simulation is carried out with heterogeneous porosities (in the range 0.2–0.4) and transmissivities (in the range \( 10^{-3} \)–\( 10^{-2} \) m\(^2\)/s) and with uniform longitudinal (\( \alpha_L = 2 \) m) and transverse (\( \alpha_T = 0.2 \) m) dispersions. Fig. 2a and b show the transmissivity and steady-state hydraulic head distributions. As shown in Fig. 2a, the channelised nature of the aquifer indicates a non-multigaussian permeability distribution (high connectivity of extreme values). Fig. 2c shows the pollution plume \( C(x, t_i) \) generated by a forward transport simulation, during \( t_i = 400 \) d, of an initial mass-release \( M_0 = 50 \) kg at a point-source, as well as the high-lighted contour \( C_0 = 8.83 \) mg/l as computed from (11). The ‘unknown’ point-source \((x_0, y_0)\) is indicated on all figures for comparison with backward shrinkage of the \( C_0 \)-contour, shown in Fig. 2d–h. The close-up area shown in Fig. 2e–h is indicated as bold dashed line in Fig. 2a. In each figure, the black area indicates where the concentrations exceed the target concentration \( C_0 \) and the dashed line shows the \( C_0 \)-contour at the next selected time. Fig. 2e–g show the shrinkage of the black area at backward times \( \tau = 0.25t_i, 0.5t_i \) and \( 0.75t_i \), and Fig. 2h shows the area just before disappearance at \( \tau = 0.99t_i \). At \( \tau = t_i \), the \( C_0 \)-contour disappears at the position \( x_0 \) as expected. This result confirms the validity of the approach in systems with
significant and non-multigaussian heterogeneity. Preliminary tests have also shown that the same procedure can be applied to transient hydraulic conditions by symmetrically reversing the time-dependent flow field.

4. Pollution source identification with few observation points

In this section, the flow field and initial mass are still assumed to be perfectly known as in the above example, and the performance of the approach is tested for the case where only a finite number of observation wells exist.

To solve the source identification problem when only a finite number of observation wells exist, the integral in (11) is replaced by a finite sum of (10) over \( n \) observation points used as release locations for backward transport. The target concentration \( C_0 \) is then expressed as

\[
C_0 = \frac{1}{M_0} \sum_{j=1}^{n} \phi(x_j) C(x_j, t_i)^2 \, dx_j
\]  

(12)
where \( C(x_j, t_i) \) is the concentration at the \( j \)th observation point and \( \phi(x_j) \) the porosity in a small discrete volume \( dx_j \) around the \( j \)th observation point. In a numerical model, \( dx_j \) is typically taken as the volume of influence around the node where the \( j \)th observation point is located. Eq. (12) implies that the reversed flow field simulation is run with the local mass detected at the \( n \) different observation points as initial condition. However, no interpolation between the observation points is required since the initial condition for the simulation is simply the mass that is locally found in the same small volume \( dx_j \) around the observation wells as is used for the calculation of the target concentration. At the beginning of the reversed flow field simulation, the \( C_0 \)-contour is discontinuous, concentrated around each observation point. As a consequence, the initial phase of back-tracking the \( C_0 \)-contour leads to migration of isolated solute parcels away from each observation point that eventually merge before the \( C_0 \)-contour disappears (Fig. 3b–e). Using only a finite number of observation wells leads to loss of information with respect to the entire pollution plume, which is why the point of disappearance only approximates the real pollution source location and the time of disappearance approximates the contamination time. To evaluate the quality of the approximation, a forward control simulation should be carried out from the estimated point of disappearance during the estimated time to compare the observed plume concentrations with the simulated concentrations. A large error indicates then that the distribution of the observation points does not capture the main features of the pollution plume geometry well enough as to provide a good approximation of the unique solution. In a real-world situation where a pollution plume has to be characterised, investigations are usually conducted to such an extent that the main features of the pollution plume are captured. The problem of approximating the pollution source in reality will far more depend on the uncertainty of the model parameters.

To illustrate the procedure for only few observation wells, the same example was chosen as in Fig. 2. In Fig. 3a the locations and observed concentrations are

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**Fig. 3.** (a) Positions of observation points within the pollution plume with concentrations [mg/l] and local porosities \( \phi \) [–]. required to calculate the target concentration \( C_0 \) according to (12). (b–e) Evolution of the target \( C_0 \)-contour for selected backward flow times. Area exceeding target concentration is shown in black, dashed line indicates the \( C_0 \)-contour at the next selected time. (f) Pollution plume simulated with a forward control simulation from approximated point-source (indicated as cross) and concentrations at observation points.
shown for eight observation wells. The pollution plume is only shown for the purpose of comparison with Fig. 2. Also indicated in Fig. 3a are the local porosities at the observation wells. The volumes of influence around all observation wells (well nodes) are constant in this example (dx = 4 m³) and the initial solute mass M₀ = 50 kg. With these data, the target concentration C₀ was calculated according to (12) and is C₀ = 0.0164 mg/l, as opposed to C₀ = 8.83 mg/l for the entire plume shown in Fig. 2c.

This target C₀-contour was back-tracked in a reversed flow field simulation using the local mass Mᵣ at each observation point as initial condition. In Fig. 3b–e the backward migration of the C₀-contour is shown for the same times as for the ideal case in Fig. 2. Fig. 3b–c reveal the migration of initially isolated solute parcels that eventually merge as they approach the real point-source location, as can be seen in Fig. 3d–e. The approximated point of disappearance of the C₀-contour, indicated as cross on Fig. 3f, is located at a distance of about 20 m from the true source location (i.e. 4.5% relative to the distance between the concentration maximum in the plume and the point-source location which is about 450 m), and disappears 10 days too late (2.5% with respect to the actual contamination time). A forward control simulation was carried out, releasing M₀ = 50 kg at the approximated point-source during the approximated contamination time 1.025t₁ (410 days). Comparison with Fig. 3a reveals a fairly good reproduction of the entire original pollution plume, despite some significant local errors.

5. Discussion

Although the above example shows that the proposed method yields a good approximation for the point-source location x₀ and the contamination time t₁ even with relatively few observation points, the error between the observed and control-simulated concentrations is locally significant. It would therefore be important to have a measure to assess the concentration errors with respect to the error related to the characterisation of the point-source.

This aspect is particularly important if the presented approach is going to be taken further to application to real-world cases, where errors are already inherent to the data sets and where the initial source mass M₀ and the hydro-dispersive parameters are uncertain. A possible path to follow is to exploit the information that a unique solution is found for both the source location and contamination time for perfectly known conditions. Applying this approach to a model with an imperfect flow field and with data errors and parameter uncertainty will necessarily yield a ‘false’ point-source location and contamination time. Running forward control simulations from these ‘false’ point-source locations during the ‘false’ contamination times and comparing the simulated concentrations with the observed plume concentrations may however allow minimisation of this error by adjustment of uncertain hydro-dispersive parameters with an optimisation method. The main restriction is then linked to the assumption that the pollution plume was generated by a single point-source.

6. Conclusions

A theoretical framework has been presented, allowing simultaneous identification of a single point-source location and contamination time in heterogeneous multidimensional systems, given perfect knowledge of the hydro-dispersive parameters. It was shown that the concentrations observed in a pollution plume contain the required information to simultaneously determine the source location and the contamination time with one backward simulation. The approach consists in backtracking a predicted target concentration C₀-contour until its point of disappearance in a reversed flow field simulation. The corresponding backward flow time indicates the time elapsed since contamination took place. This target concentration C₀ is shown to be a mass-averaged concentration defined by the observed plume values. In an ideal case, where the pollution plume and all hydro-dispersive parameters are perfectly known, the solution is unique for multidimensional systems of arbitrary heterogeneity and requires only one backward transport simulation with the pollution plume concentrations as initial condition. In a case where only few observation points exist, good approximations are obtained, provided the observation points capture the main features of the pollution plume.

The theoretical result presented in this work, valid for perfectly known flow field conditions, may potentially be extended to parameter estimation methods. Applying the approach to imperfect flow field conditions will yield ‘false’ pollution source locations and times. By running forward control simulations from these points, the error on the observed concentration data can be evaluated and the parameters adjusted in an optimisation process.

The proposed approach is so far only valid for single point-source pollution and conservative contaminants, but will hopefully be extended and further developed to handle more complicated source configurations as well as the important issue of parameter and data uncertainty.

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