Tritium/\(^3\)He measurements as calibration tools in groundwater transport modelling

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Abstract The distribution of \(^3\)H and its decay product \(^3\)He in aquifers can be utilized to calibrate groundwater flow and transport models, either by using the two isotopes as separate tracers, or by using the \(^3\)H/\(^3\)He age. A variety of modelling approaches has been applied to \(^3\)H/\(^3\)He data sets in hydrogeological settings ranging from simple homogeneous sands to fractured bedrock systems. These studies indicate that tritium concentrations alone, as well as \(^3\)H/\(^3\)He data provide powerful calibration targets for groundwater flow and transport models. \(^3\)H/\(^3\)He data are also sensitive to longitudinal and transversal dispersion and to large-scale mixing and can be used to quantify these processes.

INTRODUCTION

The standard method for calibration of groundwater flow and transport models is to adjust the boundary conditions and hydraulic parameters until the model can reproduce one or more physical states of the system. In the case of a flow model, this is typically done by minimizing differences between measured and predicted hydraulic head data. In many groundwater systems, calculated hydraulic heads are not very sensitive to changes in the distribution of hydraulic conductivity in the system. Also, hydraulic heads are often only available for a snapshot in time and may not be representative for the "average" state of the system.

If transport of contaminants is of concern, at least one other parameter, the dispersion coefficient is required, which cannot be determined from hydraulic data alone. Therefore, many authors have suggested the use of calibration targets other than hydraulic head data, such as directly measured streamflow gain/losses or groundwater flow velocities (e.g. Anderson & Woessner, 1992). Recent developments of tracer techniques, such as \(^3\)H/\(^3\)He, chlorofluorocarbons (CFCs), \(^85\)Kr, etc. provide another calibration target for groundwater flow and transport models. They can be applied as directly measured concentration fields or as apparent ages or residence times derived from their measured concentrations. In this contribution we focus on a brief review of the use of \(^3\)H and \(^3\)He data for calibration of groundwater flow and transport models.

\(^3\)H/\(^3\)He DATING

Tritium (\(^3\)H or T), the radioactive isotope of hydrogen, is produced naturally by interaction of cosmic rays with nitrogen and oxygen in the stratosphere. Tritium decays
with a half-life of 12.43 years to the noble gas $^3$He. Starting in the early 1950s, tritium from anthropogenic sources (mainly atmospheric nuclear weapon tests) was added to the atmosphere in significant amounts. After oxidation to $^2$H$^3$HO, tritium participates in the global hydrological cycle. Tritium concentrations in precipitation have been monitored by an international network of stations beginning in the 1950s (IAEA/WMO, 1998) and its delivery to surface waters, groundwater, and the oceans is reasonably well known (frequently called "input function"; Doney et al., 1992). Tritium has been used extensively as a dye for better understanding the dynamics of groundwater flow systems (see Schlosser, 1992 for references on early tritium applications). However, as a consequence of $^3$H decay and mixing, it has become increasingly difficult to quantify groundwater flow dynamics by relating measured $^3$H concentrations to the $^3$H input functions. The simultaneous measurement of $^3$H and $^3$He, allows us to compensate radioactive decay and to determine groundwater $^3$H concentrations as if $^3$H were a stable isotope. In addition, we can determine an apparent age based on the concentration ratio of radioactive mother ($^3$H) and daughter ($^3$He), the $^3$H/$^3$He age $t$:

$$t = \frac{12.43}{\ln 2} \ln \left( 1 + \frac{[^3\text{He}]_{\text{ref}}}{[^3\text{H}]} \right)$$

This method, originally proposed by Tolstikhin & Kamensky (1969), has found many applications in groundwater hydrology during the past 20 years. For recent reviews of basic principles and applications of this technique, see Cook & Solomon (1997), Schlosser et al. (1998), Solomon & Cook (1999), and Schlosser et al. (2000).

TRITIUM AND $^3$HE AS MODEL CONSTRAINTS

Tritium and $^3$He are ideal tracers for groundwater flow and transport, because they move at the same speed as water (except for cases where molecular diffusion becomes important) and because they do not undergo chemical reactions. The bridge between independently constructed groundwater models and $^3$H/$^3$He data has been built only very recently. Most of the earlier $^3$H/$^3$He studies treated the tracer data within a simple conceptual model or in the framework of quantitative tracer models.

The initial discussions and first applications were primarily concerned with the potential of the $^3$H/$^3$He method and its technical limitations (sampling, measurements, geochemistry). They also implicitly assumed that the effects of dispersion can be neglected and that $^3$H/$^3$He ages do reflect the true residence time of a groundwater parcel.

Subsequently, one-box, or "lumped parameter" models were introduced that ignore any spatial variations of hydrogeological parameters in the system (Maloszewski & Zuber, 1983). The exponential model assumes an exponential distribution of residence times in the system. The mean residence time of groundwater in the box is the only parameter in this model. The dispersion model takes the effects of dispersion into account and thus has two parameters, the dispersion coefficient and the mean residence time. In both these models, the $^3$H/$^3$He ratio (or the apparent $^3$H/$^3$He age) depends on the $^3$H input function. The residence time of a groundwater parcel is typically determined by comparison of the measured $^3$H/$^3$He ratios with a set of curves that
show the $^3$H/$^3$He ratio as a function of residence time, time of sampling, and in the case of the dispersion model, also the Peclet number $D/vx$. The Peclet number is a dimensionless measure of the relative importance of dispersion and advection (Maloszewski & Zuber, 1983). This approach is useful when the collected samples are representative of a large fraction of the entire system, e.g. if they were collected from springs in which many flow lines converge, or from wells with long screens that tap large fractions of the aquifer.

The next generation of models that was used to interpret $^3$H/$^3$He data was one-dimensional (1-D) vertical or horizontal advection/dispersion models (e.g. Schlosser et al., 1989; Solomon & Sudicky, 1991; Ekwurzel et al., 1994; Stute et al., 1997). The main motivation for the development of these models was to evaluate the influence of dispersion on $^3$H/$^3$He data. Two processes contribute to dispersion in groundwater: molecular diffusion and mechanical mixing caused by spatial variability of groundwater flow velocities primarily due to subsurface heterogeneity. In small-scale, homogeneous, sandy aquifers the former process may dominate which, in contrast to mechanical mixing, affects $^3$H and $^3$He differently because of their different diffusion coefficients. Homogeneous, unconfined aquifers with simple geometry often have a fairly uniform vertical age structure, which justifies the application of 1-D transport models. An example of this type of model is shown in Fig. 1. A vertical flow velocity of 1 m year$^{-1}$ was assumed and the influence of dispersion on the distribution of $^3$H/$^3$He ages determined. The over- and underestimation of the hydrodynamic age is a result of the dominance of the atmospheric $^3$H concentrations by the nuclear surface test peak in the early 1960s.

![Image](image_url)

**Fig. 1** The effect of dispersion on the $^3$H/$^3$He age in a system dominated by vertical water movement. The vertical flow velocity is 1 m year$^{-1}$ and the calculation was terminated in 1992. A $^3$H input function typical for the northeastern United States was used. Similar 1-D models were published by Schlosser et al. (1989), Solomon & Sudicky (1991), Ekwurzel et al. (1994), and others.

This type of model was used to determine boundaries for the dispersion coefficient in a shallow aquifer in Germany (Schlosser et al., 1989) and on the Delmarva Peninsula in the USA (Ekwurzel et al., 1994). A 1-D, vertical advection/dispersion
model was also used to derive groundwater recharge rates in a shallow aquifer near Sturgeon Falls, Ontario (Solomon et al., 1993). The latter study allowed the separation of the processes contributing to dispersion, mechanical mixing and molecular diffusion, by taking advantage of the different diffusion coefficients of $^3$H and $^3$He. A number of the studies found that the effects of dispersion on tracer transport appear to be of minor importance (e.g. Solomon & Sudicky, 1991; Solomon et al., 1993).

The above-described models were developed to interpret $^3$H/$^3$He data independently of other hydrogeological constraints. Solomon et al. (1992) compared the spatial distribution of $^3$H/$^3$He ages with a previously calibrated groundwater transport model of the Borden aquifer in Canada. Although the $^3$H/$^3$He age profiles were vertically offset from the modelled travel times, the gradients in travel time and $^3$H/$^3$He age compared very well. In another study of a sandy aquifer in the southern New Jersey coastal plain, $^3$H/$^3$He ages were compared to two-dimensional groundwater flow models that were calibrated without using information from large-scale tracer measurements. Steady-state finite-difference groundwater flow models were calibrated at three sites by adjusting horizontal and vertical hydraulic conductivities to match measured hydraulic heads (Szabo et al., 1996). Travel times were then calculated using the particle tracking code and compared to the measured $^3$H/$^3$He (and CFC) ages (Fig. 2). The good agreement between $^3$H/$^3$He, CFC, and particle tracking ages indicates that the influence of dispersion is very small at this site and that, with few exceptions, the groundwater flow model does not require major adjustments. Similar studies were used earlier with CFC and $^3$H (not $^3$H/$^3$He) concentrations as calibration targets (Robertson & Cherry, 1989; Reilly et al., 1994; Katz et al., 1995).

![Fig. 2](image_url)  
**Fig. 2** Comparison of simulated travel times and apparent $^3$H/$^3$He ages for the Kirkwood-Cohansey aquifer system, southern New Jersey coastal plain, USA (from Szabo et al., 1996).

In recent studies, attempts have been made to merge hydrogeological models and $^3$H/$^3$He data in more complex systems. Particle tracking ages derived from independently calibrated three-dimensional flow models for two sites in a hydrogeologically complex buried-valley aquifer in Ohio compared reasonably well with $^3$H/$^3$He ages. The agreement decreased with depth (Sheets et al., 1998). Selected
conceptual and parameter modifications (porosity, transmissivity) to the models resulted in improved agreement between $^3$H/$^3$He ages and simulated travel times and also between measured and simulated heads and fluxes. In a study of contaminated groundwater in Cape Cod, Massachusetts, $^3$H/$^3$He data were found in good agreement with particle tracking ages, except for an area where groundwater flows through a pond, which as a consequence of $^3$He re-equilibration with the atmosphere reset the $^3$H/$^3$He clock. The apparent disagreement between the different approaches therefore allowed the identification of an additional process resulting potentially in additional contamination (Dunkle-Shapiro et al., 1999).

The first attempts to calibrate fractured rock groundwater flow models demonstrate some of the difficulties encountered in these even more complex environments (Cook et al., 1996; Sudicky et al., 1997). Typical problems encountered in fractured systems are their extreme heterogeneity, double porosity causing differences between hydrodynamic and $^3$H/$^3$He age, and geochemical complications due to the presence of other $^3$He sources ($^3$He originating from the Earth's mantle, and in situ production).

OUTLOOK

The integration of $^3$H/$^3$He (and CFC) data in hydrogeological models is only at its beginning. Most studies so far looked at very simple, typically fairly homogeneous sandy groundwater flow systems under quasi-steady state conditions. Since $^3$H/$^3$He ages are the result of processes taking place along the entire flow path of a water parcel, relatively few $^3$H and $^3$He measurements can lead to a highly constrained groundwater flow and transport model. However, the potential of this approach needs further exploration, in particular in hydrogeologically complex environments, and also under transient conditions. Uncertainties introduced by double or poorly constrained porosities, for example, may complicate the application of this technique. It would also be very useful, if capabilities to simulate tracer distribution such as $^3$H and $^3$He were built into forward- and inverse computer codes that are widely used by the scientific and engineering community.

REFERENCES


