

BOXMODEL

An Excel-workbook for the interpretation of transient tracer data (^3H , ^3He , CFCs, ^{85}Kr) with the box-model approach.

Original version developed by
Kai Zoellmann

Later additions by
Werner Aeschbach-Hertig

Documentation and final version by
Urs Beyerle

This document provides a general introduction to the use of box-models to interpret tracer data and a documentation for the Excel-workbook "Boxmodel-V3". It is an excerpt from the UNESCO report UNEP/DEWA/RS.02-2, "A Survey of Methods for Groundwater Recharge in Arid and Semi-arid regions", by W. Kinzelbach et al., 2002. The complete report is available at: www.unep.org/water/groundwater.

**A Survey of Methods for Groundwater Recharge in Arid and
Semi-arid regions**



UNEP/DEWA
UNESCO/IHP

This publication was compiled with contributions from:
Wolfgang Kinzelbach, IHW, ETHZ, Zurich, Switzerland
Werner Aeschbach, EAWAG, Zurich, Switzerland
Carmen Alberich, IHW, ETHZ, Zurich, Switzerland
Ibrahim Baba Goni, Dept. of Geology, University of Maiduguri, Nigeria
Urs Beyerle, University of Berne, Berne, Switzerland
Philip Brunner, IHW, ETHZ, Zurich, Switzerland
Wen-Hsing Chiang, S.S. Papadopoulos Associates, Bethesda, MD, USA
Joerg Rueedi, University of Berne, Berne, Switzerland
Kai Zoellmann, ETHZ, Zurich, Switzerland.

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Design layout, editing and production by Salif Diop, Ralph Johnstone, Patrick M'anyi, Dennis Lisbjerg and Audrey Ringler.

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Division of Early Warning and Assessment
United Nations Environment Programme
P.O. Box 30552
Nairobi 00100,
Kenya
Tel : (254 2) 62 41 05
Fax : (254 2) 62 42 69
E-mail: dewainfo@unep.org

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The software included on the CD-ROM is available at: www.unep.org/water/groundwater/

5 Groundwater Dating using Environmental Tracers and Black Box Models

Urs Beyerle, University of Berne, Berne, Switzerland

5.1 Introduction

The age of groundwater – the water’s age or its residence time – is defined as the time elapsed since the water parcel had its last contact with the atmosphere. Water in shallow aquifers can range from days to decades in age, while old groundwater may have residence times from 10^3 to 10^7 years. Groundwater renewal or recharge is unknown for most aquifers, or associated with great uncertainties as a result of the difficulties in its quantification.

5.2 Environmental Tracers

A number of environmental tracers carry information on the age of groundwater. Tritium (^3H) and rare gas tracers such as ^3He , ^{85}Kr and chlorofluorocarbons (CFCs) are used to date groundwater with an age of less than 50 years. For groundwater with ages of a few hundred years, the most useful age indicator is ^{39}Ar , whereas ^{14}C , ^{36}Cl and radiogenically produced noble gases are normally used to date even older groundwater (Table 5.1).

In industrial areas groundwater is often over saturated with CFCs, indicating either

higher atmospheric concentrations due to the proximity to emission sites or direct input of chlorinated substances into the groundwater flow, eg. from dump sites. In addition, there are indications that CFCs decompose under anoxic conditions (especially CFC-11). Also, a retardation due to adsorption on organic aquifer matter may occur. In general, CFC dating is recommended for water age analysis in sandy aquifers far away from industry.

In principle three different kinds of dating methods have been identified. The first set of methods utilises the fact that certain atmospheric trace components, such as CFCs, ^{85}Kr and tritium (^3H), have a time dependent input function (Figure 5.1). Thus their atmospheric concentrations are not constant but vary over the past 50 years.

Industrial production introduces most of the CFCs, especially CFC-11 and CFC-12, to the atmosphere, while ^{85}Kr is released from nuclear reprocessing plants. In the early 1960s, nuclear bomb tests in the atmosphere increased the global tritium inventory by 2 to 3 orders of magnitude. Tritium concentrations have steadily decreased since the nuclear testing of the '60s, but up to today the tritium activity in meteoric water is commonly one order of magnitude greater than the expected

TABLE 5.1: TIMESCALES OF VARIOUS TRACER-BASED DATING METHODS

Tracer	Time scale	Source	Detection
^{222}Rn	days – weeks	^{238}U	α -counting
^{37}Ar	Months	spallation (S)	Low level counting (LLC)
^3H - ^3He	months – decades	S, nuclear bomb testing	Mass spectrometry (MS)
^{85}Kr	months – decades	nuclear power techniques	LLC
CFCs	months – decades	industry	Gas chromatography (GC)
^{39}Ar	10^2 years	S	LLC
^{14}C	10^3 - 10^4 years	S	LLC, MS
$^4\text{He}_{\text{rad}}$	10^2 - 10^7 years	U, Th	MS
$^{40}\text{Ar}_{\text{rad}}$	10^4 - 10^7 years	^{40}K	MS

background level due to tritium production by cosmic rays. If a given water parcel is in gas exchange with the atmosphere at time t , the actual concentration in the water reflects the atmospheric partial gas pressure at this time (Henry's law). As the atmospheric partial pressure of the tracer changes over time, the concentrations of successive water parcels introduced into the aquifer change as well. If mixing processes can be neglected, the concentration of a water parcel basically reflects the time elapsed between the last gas exchange and the sampling date.

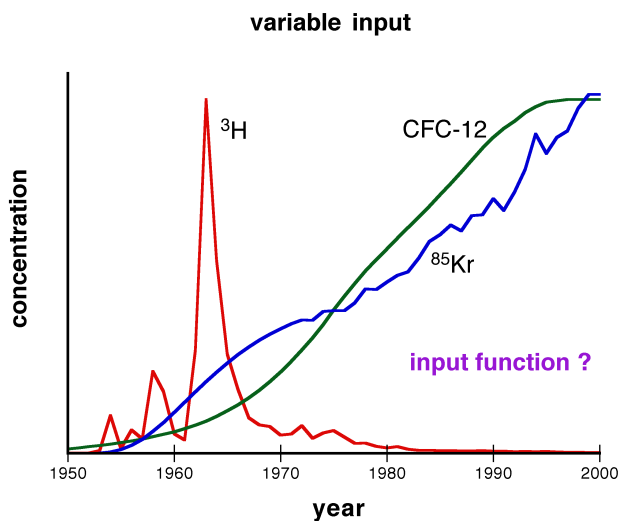


FIG. 5.1: TRACERS WITH VARIABLE INPUT FUNCTIONS

The second set of methods relates mainly to radioactive decay, eg. ^{14}C decays with a half-life of 5,730 years and ^{39}Ar decays with a half-life of 269 years. In order to calculate a groundwater residence time using a radioactive tracer, its initial input concentration should be known (Figure 5.2). In contrast to ^3H and ^{85}Kr , the fluctuations of atmospheric concentrations of ^{14}C and ^{39}Ar are small and their initial concentrations can be estimated.

FIG. 5.2: RADIOACTIVE TRACER CONCENTRATION AS A FUNCTION OF TIME

mother - daughter isotope

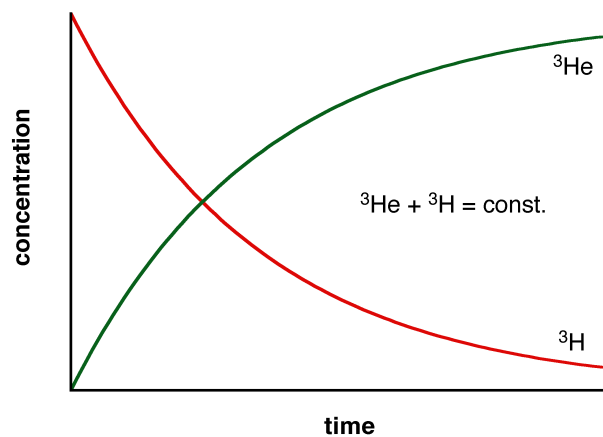
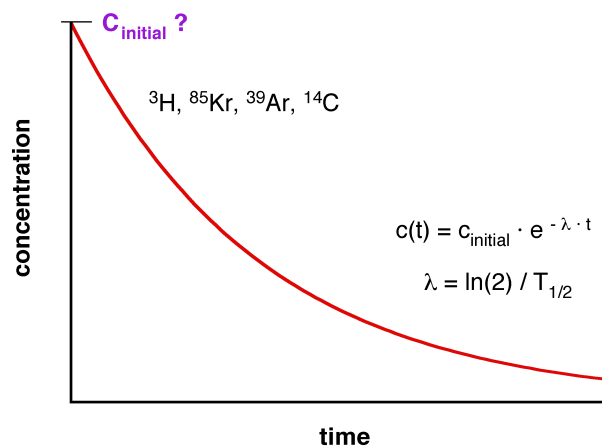


FIG. 5.3: THE 'MOTHER-DAUGHTER ISOTOPES' ^3H AND ^3He

In a closed system, in which all isotopic changes are purely the result of radioactive decay, the ratio of the stable daughter (^3He) and radioactive mother (^3H) is a direct measure for the time elapsed since the water was last in contact with the atmosphere. As this so-called ^3H - ^3He Dating relies only on the radioactive decay, it does not depend on the knowledge of the atmospheric tritium input function (Figure 5.3).

The third set of groundwater dating methods relies on a continuous increase of a particular tracer. For example, the stable noble gas isotopes ^4He and ^{40}Ar have the

radioactive decay



potential to be used for dating very old groundwater. The production of these radiogenic noble gas isotopes is controlled by long-lived nuclides in minerals and rocks, such as U, Th isotopes and ^{40}K . Groundwater ages derived from ^4He or ^{40}Ar concentrations are based on the assumption that the radiogenic noble gases accumulate continuously during groundwater flow (Figure 5.4). The radiogenic noble gases are expected either to diffuse out of minerals within the aquifer or to emanate from deeper strata into the aquifer. In order to obtain groundwater residence times, the respective accumulation rates must be known. They can, however, only be crudely estimated.

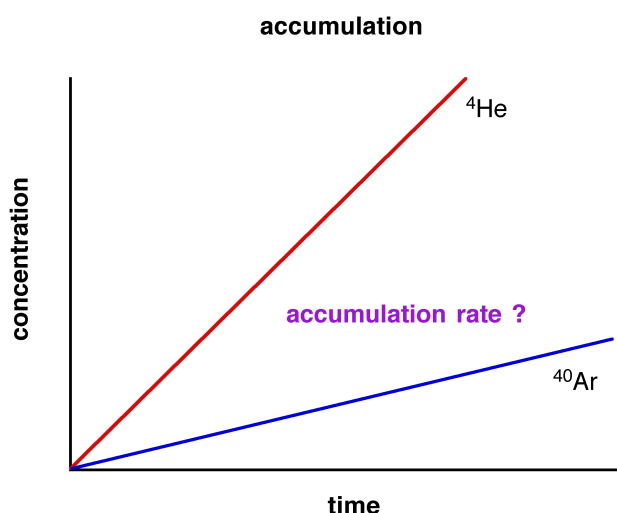


FIG. 5.4: GROUNDWATER DATING WITH RADIOGENIC NOBLE GAS ISOTOPES

The combination of dating methods restricts the mean residence time of an aquifer to a much greater extent than the use of only one technique. Therefore mixing processes in the groundwater can be addressed if several tracers are available. Since the tracers used are both chemically and biologically inert, they take part in the physical groundwater processes only. They can serve as a scale marker for other substances, which undergo additional chemical and biological alterations in the aquifer.

The various dating methods exploit different ways of tracer introduction into the groundwater body. CFCs and rare gas isotopes are dissolved as gases in the water, whereas tritium is bound in water molecules as $^1\text{H}^3\text{HO}$ and is therefore part of the moving water. CFCs and rare gases are therefore influenced by gas exchange to a much greater degree than tritium. In addition, gas tracers only measure the time since the groundwater reached the saturated zone (ie. the start of its complete isolation from the atmosphere), whereas tritium travels as a component of the water molecule through the unsaturated zone.

TABLE 5.2: EXAMPLE OF A GROUNDWATER SAMPLE TAKEN IN JANUARY 2000

Tracer	Concentration	Mean residence time (see Fig. 5.5)
CFC-12	390 ± 10 pptV ⁽¹⁾	15 years
^{85}Kr	25.7 ± 1.1 dpm/cc Kr ⁽²⁾	12 years
^3H	18.8 ± 0.9 TU ⁽³⁾	46, 42, 39, 21, 19 years

(1) pptV = parts CFC per trillion (10^{12}) parts air by volume. An atmospheric CFC-12 concentration of 390 pptV corresponds to 243 pg/kg (= 10^{-12} g/kg) measured in groundwater (at T = 20°C, atmospheric pressure = 954 mbar)

(2) dpm/cc Kr = decays per minute per cm^3 STP Kr

(3) TU = Tritium Unit = 1TU = $^3\text{H}/^1\text{H}$ ratio of 10^{-18}

5.3 Testing the Methods

In Table 5.2, an example of a groundwater sample is given. The sample was taken in January 2000 and concentrations of CFC-12, ^{85}Kr and ^3H were measured. Using the measured tracer data and assuming piston flow conditions, mean groundwater residence times were calculated (Table 5.2 and Figure 5.5). The piston flow assumption implies that the water parcel basically remained unmixed after its entry into the aquifer, and preserved its original identity.

For CFC-12 the measured concentration is given as the equivalent atmospheric concentration linked to it by Henry's law. Assuming no decomposition of CFC-12 (ie. no anoxic conditions), all we have to do is to compare at what time in the past the atmospheric concentration was equal to the measured concentration. We draw a horizontal line at the equivalent atmospheric value of the sample and look for the intersection with the input curve. As the input curve is monotonically increasing, there is only a single intersection.

In the cases of krypton and tritium, we do the same but take into account the decay. This requires a backward extrapolation from the concentration still remaining in 2000 to the original value. As the plots are semi-logarithmic we go back in a straight line, the slope of which is given by the decay constant. At the intersection with the input curve, we find a possible candidate for the time at which the parcel started its travel. Note that for tritium, several solutions are feasible as the input curve is not monotonic.

5.4 The Box Model Approach

The simplest interpretation of environmental tracer data is based on the box model approach (Fig. 5.6).

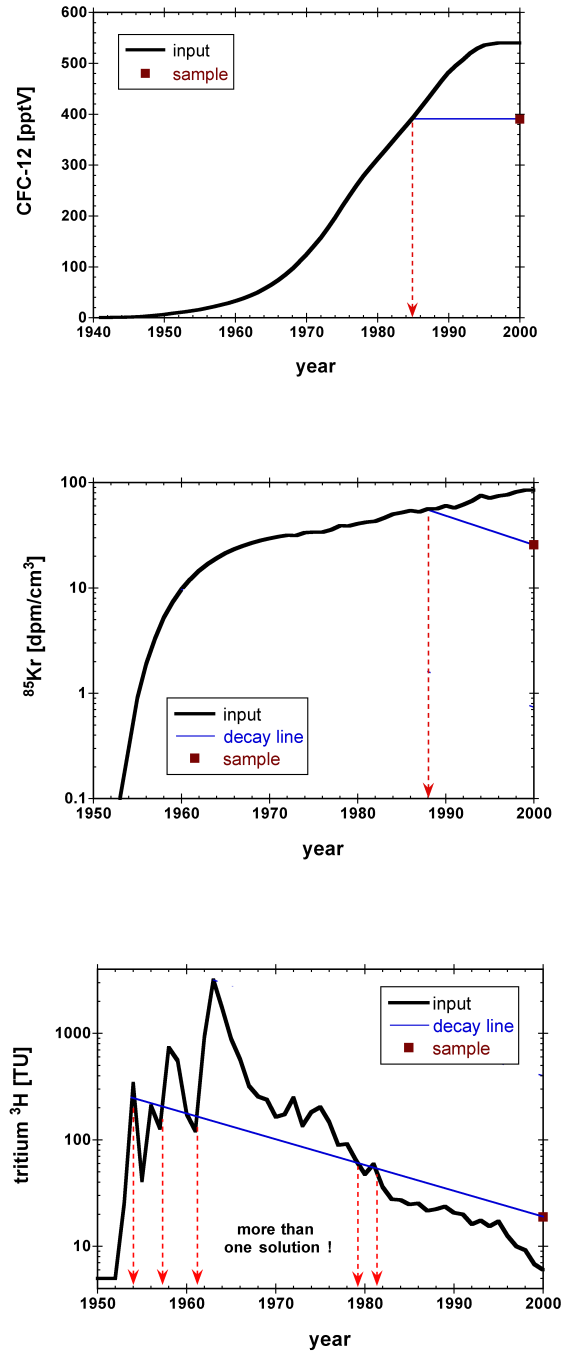


FIG. 5.5: CALCULATING MEAN GROUNDWATER RESIDENCE TIMES USING CFC-12, ^{85}KR , AND ^3H (ASSUMING PISTON FLOW CONDITIONS)

For a given tracer input function, a box model calculates the convoluted theoretical output depending on:

- the transfer function (either for a piston flow, an exponential or a dispersion model, see Fig. 5.7a-c); and

- the parameters of this particular transfer function (mean residence time τ , dispersion parameter δ).

By comparing observed and calculated concentrations, one can identify these parameters.

"black box" or "lumped parameter" models

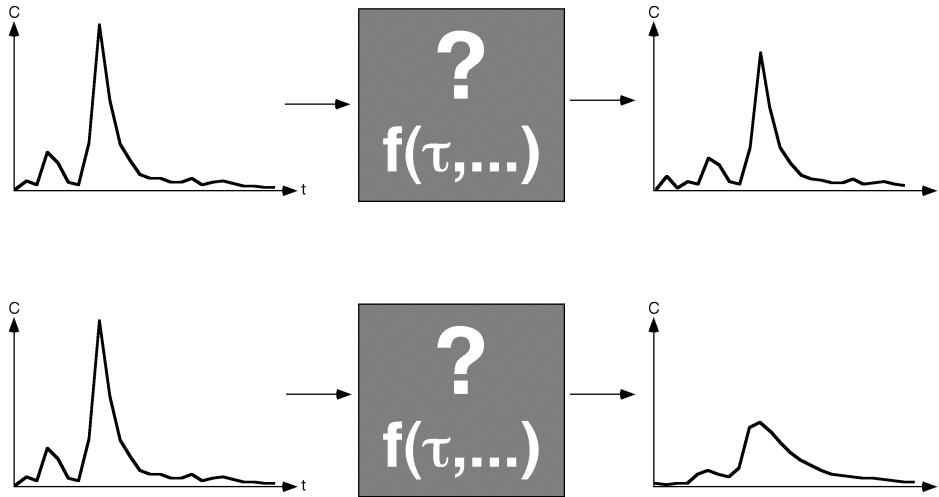


FIG. 5.6: SCHEMATIC PRINCIPLE OF BOX MODELS

Piston Flow Model (PM)

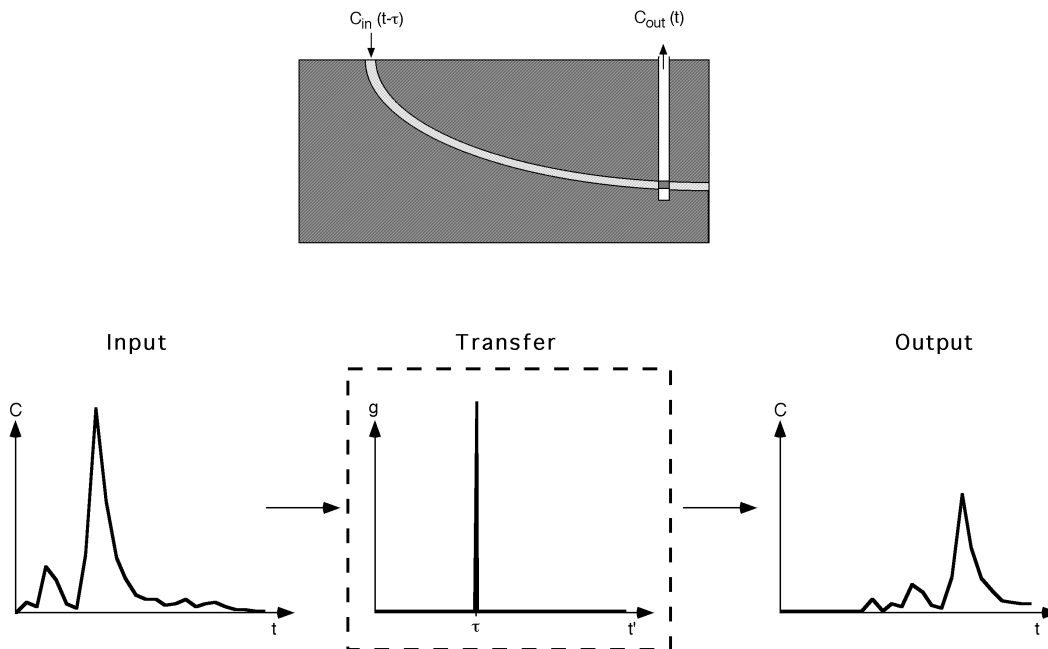


FIG. 5.7 A: PISTON FLOW MODEL AND CORRESPONDING TRANSFER FUNCTION (THE OUTPUT CONCENTRATION IS ADDITIONALLY INFLUENCED BY RADIOACTIVE DECAY)

Exponential Model (EM)

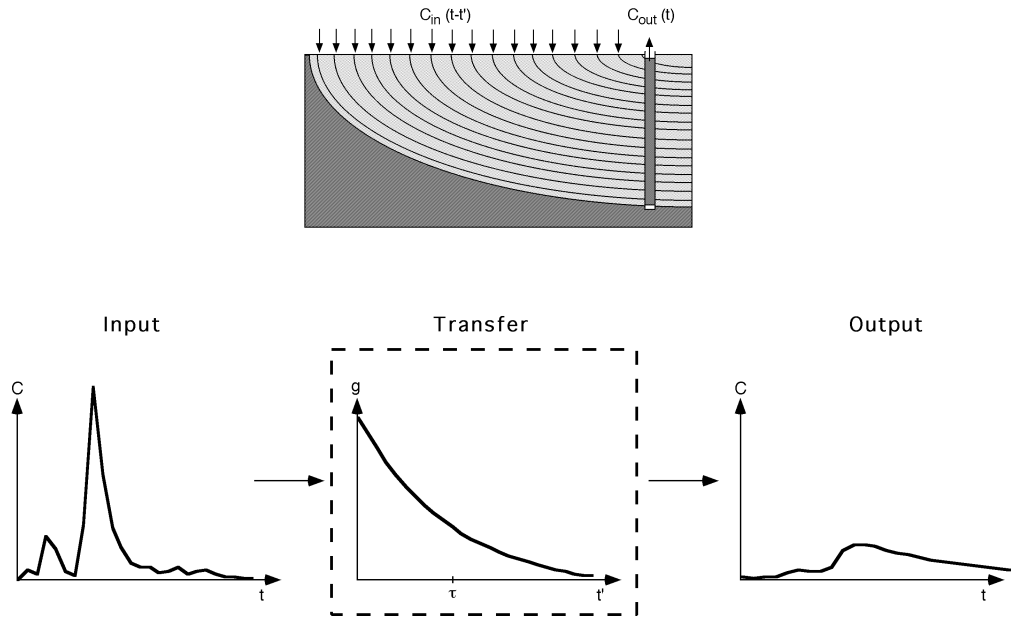


FIG. 5.7 B: EXPONENTIAL MODEL AND CORRESPONDING TRANSFER FUNCTION (THE OUTPUT CONCENTRATION IS ADDITIONALLY INFLUENCED BY RADIOACTIVE DECAY)

Dispersion Model (DM)

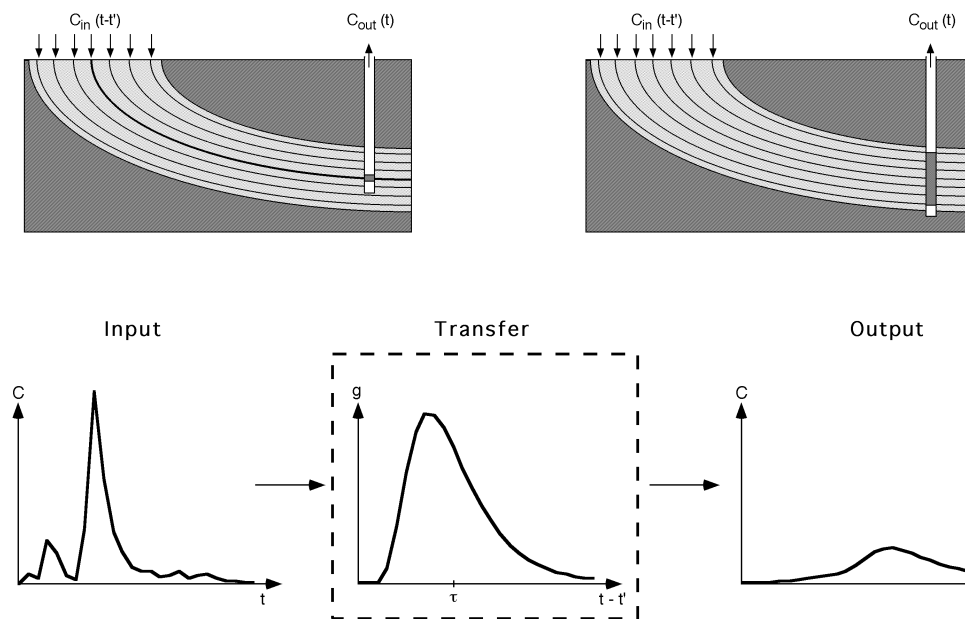


FIG. 5.7 C: DISPERSION MODEL AND CORRESPONDING TRANSFER FUNCTION (THE OUTPUT CONCENTRATION IS ADDITIONALLY INFLUENCED BY RADIOACTIVE DECAY)

The mathematical basis of box models, as well as a spreadsheet for working with them, are provided in the next chapter.

The choice of the box model should respect the general aquifer situation, as characterised by the figures above.

6 Use of Spreadsheets for Age Determination with Tritium and CFCs

Kai Zoellmann, ETHZ, Zurich, Switzerland

Werner Aeschbach, EAWAG, Zurich, Switzerland

The box model approach described in Chapter 5 is implemented with an Excel Workbook, provided as *Boxmodel_V3.xls* in the CD-ROM accompanying this report (or www.unep.org/water/groundwater/). This spreadsheet software allows for the interpretation of environmental tracer data taken from groundwater samples (^3H , $^3\text{He}_{\text{tri}}$, ^{85}Kr and CFCs), based on the box model approach (see Figure 5.6).

The use of the Excel Workbook in the interpretation of measured concentrations of environmental tracers in wells is described below. All you need to run it is a computer equipped with standard Excel software. A simplification used in this Workbook is that the input data are assumed to be constant over the year, which could be unrealistic for systems with very small residence times.

The Workbook consists of 17 separate sheets (INTERFACE, Tr in Graph, CFC in Graph, Kr in Graph, Transfer Exp Graph, Transfer Disp Graph, Output Graph, Tau Graph, Tritium Input, CFC Input, ^{85}Kr Input, Piston Flow Model, Exponential Model, Dispersion Model, Output (tau), Output (t) and CFC Converter) which are described in detail below.

Interface

The Interface sheet contains the following input parameters that can be changed by the user (black bordered cells):

Model Code - Enter the desired transfer function (pm, exp or dm) that is used for convoluting the input function (pm = Piston Flow Model, exp =

Exponential Model, dm = Dispersion Model).

Tau [a] - Enter a value for the mean residence time in years for the pm, exp or dm model. The Transfer Function Graph is calculated for this value only. In the Tau Graph, on the other hand, the output concentrations for the required year are shown as function of Tau for a whole range of Tau values.

Tau Step [a] - This value determines the step width for the Tau Graph in years.

Delta [a] - Dispersion Parameter for the Dispersion Model. This value has no effect if the Piston Flow or the Exponential Model is used.

Tritium Factor - Defines a scaling factor for the tritium input function.

Tracer Code - Enter a code (tr, cfc, kr, he) for the desired tracer:

tr = tritium; cfc = chlorofluorocarbons CFC-11 or CFC-12 depending on the input function provided (see sheet CFC Input); kr = ^{85}Kr ; and he = tritiogenic ^3He (i.e. ^3He which originates from the tritium decay during groundwater flow).

Year of Observation - This value determines the time for which the concentrations are calculated (Tau Graph). This should be the year of your tracer measurement.

C_obs - Enter here the measured tracer concentrations in the same units as the corresponding input function. These values are not needed for the

computation of the other graphs. But the comparison of those lines with the computed graphs allows the age dating, and they are therefore drawn in the same graph as the computed output. Note that the measured CFC concentration of the groundwater sample [eg. in pg/kg] must be transformed to pptV according to the solubility, which depends on the groundwater temperature and the altitude of the recharge area (see CFC Converter sheet) as the input function is given in these units.

Tr in Graph, CFC in Graph, Kr in Graph

These three sheets show the input functions of tritium (^3H), CFC-12 or CFC-11, and ^{85}Kr derived from the Tritium Input, CFC Input and ^{85}Kr Input data sheets.

Transfer Exp Graph, Transfer Disp Graph

These sheets show transfer functions based on the parameters (Tau and Delta) given in the User Interface Sheet.

Output Graph

This sheet shows the resulting output concentration for the desired transfer function as a function of time t (year of observation) for the given parameters Tau and Delta. The intersection of this curve with the observed concentration (red line) should be in agreement with the chosen residence time (Tau). You can obtain this agreement iteratively. But you can also proceed to the Tau Graph for a more convenient method.

Tau Graph (=Residence Time Graph)

This sheet shows the resulting output concentration for the desired transfer function as a function of the parameter Tau (and implicitly Delta) for a given time t (the year of observation). The intersection of this curve with the observed concentration (red line) identifies the 'correct' mean residence time (Tau).

Tritium Input, CFC Input, ^{85}Kr Input

These three sheets contain the data of the input functions of tritium (^3H), CFC-12 or CFC-11, and ^{85}Kr . The tritium concentrations were derived from monthly averaged precipitation and have to be adapted to your local tritium input function. They are given in Tritium Units (TU). The CFC data are given in pptV and are derived from global CFC measurements. If necessary they will have to be adjusted to local conditions. The ^{85}Kr input function can be strongly dependent upon the investigated area and should be modified if local atmospheric concentrations are known.

Piston Flow Model, Exponential Model, Dispersion Model, Output(tau) and Output(t)

These five sheets contain the calculation algorithms providing the data for the different graphs. The user should not change them.

CFC Converter

This sheet allows one to convert the measured CFC concentration in pg/kg in water to the corresponding atmospheric concentration in pptV.

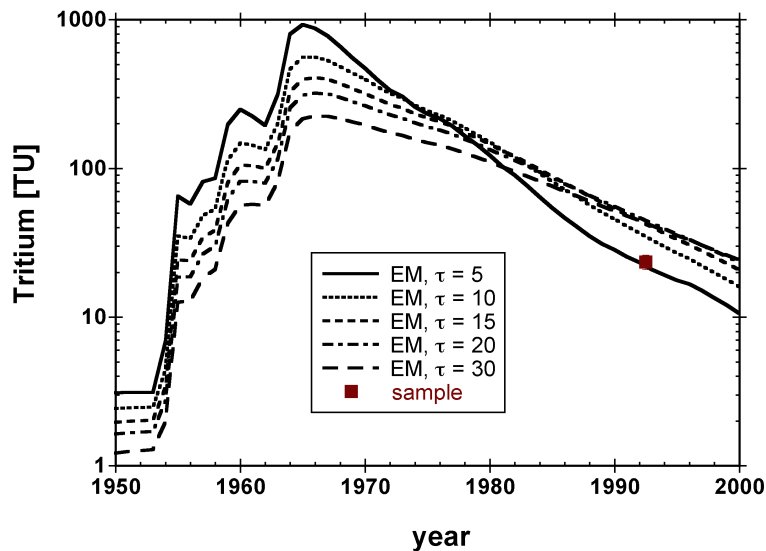
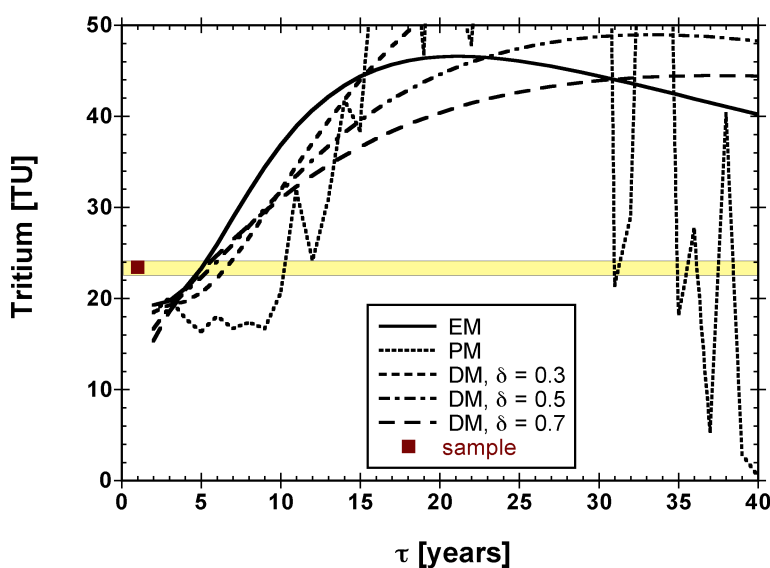


FIG. 6.1: OUTPUT CONCENTRATION OF TRITIUM AS A FUNCTION OF TIME FOR DIFFERENT EXPONENTIAL MODELS (τ = MEAN RESIDENCE TIME)

6.1 Example

As an example, the Output Graph and the Tau Graph for different models (EM, PM, DM) and parameters (τ , δ) are calculated (Figures 6.1 and 6.2) and compared with a sample taken in June 1992 ($^3\text{H} = 23.4$ TU).

FIG. 6.2: OUTPUT CONCENTRATION AS A FUNCTION OF THE MEAN RESIDENCE TIME τ FOR DIFFERENT MODELS AND PARAMETERS (EM = EXPONENTIAL MODEL, PM = PISTON FLOW MODEL, DM = DISPERSION MODEL, δ = DISPERSION PARAMETER)



The Output Graph shows a strong agreement of the exponential model (EM) with the measured sample in the case of a mean residence time $\tau = 5$ years (Figure 6.1). The same result can be obtained from Figure 6.2 (intersection at $\tau = 5$ years between measured concentration and calculated Tau Output function of the exponential model).

If the investigated aquifer is best described with a piston flow model, a realistic residence time would be around 10 years or between 31 and 38 years (Figure 6.2). Assuming a dispersion model (DM with δ between 0.3 and 0.7), a realistic mean residence time would be close to the one calculated using the exponential model, i.e. about 5-7 years.

In general, with one tracer measurement only one parameter can be determined (normally τ). Therefore the type of the model (PM, EM or DM) and eventually additional parameters (eg. δ) should be known. This will require that a conceptual model for the investigated aquifer already exists.

To determine the type of model and additional parameters using tracer data only, either a time series (1 tracer, 1 site, additional measurements over time) or additional tracers (1 site, 1 time, additional tracers such as CFC or ^{85}Kr) will be needed. A third possibility would be tracer data from

additional boreholes (1 tracer, 1 time, more sites). But in such cases, black-box models are less suitable because the residence time (τ) has to be determined for each borehole separately. Therefore a better approach

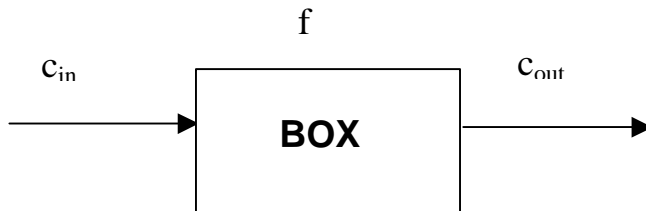
would be to use a numerical flow and transport model. This applies also for strongly time varying recharge rates or cases in which a single average recharge rate is inadequate.

6.2 Mathematics of the Black Box Model Approach

Principle: The input signal $c_{in}(t')$ is convoluted with a transfer function $f(t,t')$ to yield the output signal $c_{out}(t)$.

Assumptions:

- the system is linear;
- with a steady state in its flow properties $f(t,t') = f(t-t')$.



$$c_{out}(t) = \int_{-\infty}^t c_{in}(t') \cdot f(t-t') dt'$$

Plugflow model

$$f(t-t') = \delta(t-t'-\tau)$$

$$c_{out}(t) = \int_{-\infty}^t c_{in}(t') \cdot f(t-t') dt' = c_{in}(t-\tau)$$

Exponential Model

$$f(t-t') = \frac{1}{\tau} \cdot \text{Exp}\left[-\frac{t-t'}{\tau}\right]$$

By including (radioactive) decay, we obtain:

$$c_{out}(t) = \int_{-\infty}^t c_{in}(t') \cdot \text{Exp}[-\lambda \cdot (t-t')] \cdot \frac{1}{\tau} \cdot \text{Exp}\left[-\frac{t-t'}{\tau}\right] dt'$$

If $c_{in}(t) = \text{const on } [t', t'+1]$ (Yearly Input Data), the integral can be converted into a sum:

$$\Rightarrow c_{out}(t) = \sum_{t'=-\infty}^{t-1} c_{in}(t') \cdot \frac{1}{\tau} \cdot \int_{t'}^{t'+1} \text{Exp}\left[-\left(\frac{1}{\tau} + \lambda\right) \cdot (t-t'')\right] dt''$$

$$c_{out}(t) = \sum_{t'=-\infty}^{t-1} c_{in}(t') \cdot \frac{1}{t} \cdot \frac{1}{\frac{1}{t} + I} \left\{ \text{Exp} \left[- \left(\frac{1}{t} + I \right) \cdot (t - t') \right] \right\}_{t'}^{t'+1}$$

The latter equation can be solved for τ (iteratively or graphically).

Dispersion Model

$$f(t-t') = \frac{1/t}{\sqrt{4pd \frac{t-t'}{t}}} \cdot \frac{1}{t-t'} \cdot \text{Exp} \left[- \frac{\left(1 - \frac{t-t'}{t} \right)^2}{4d \frac{t-t'}{t}} \right]$$

The convolution yields:

$$c_{out}(t) = \int_{-\infty}^t c_{in}(t') \cdot \text{Exp} \left[- I t \cdot \frac{t-t'}{t} \right] \cdot \frac{1/t}{\sqrt{4pd}} \cdot \left(\frac{t-t'}{t} \right)^{\frac{3}{2}} \cdot \text{Exp} \left[- \frac{\left(1 - \frac{t-t'}{t} \right)^2}{4d \frac{t-t'}{t}} \right] dt'$$

If $c_{in}(t) = \text{const on } [t', t'+1]$ (Yearly Input Data), the integral can be converted into a sum:

$$c_{out}(t) = \frac{1}{t} \cdot \frac{1}{\sqrt{4pd}} \cdot \sum_{t'=-\infty}^{t-1} c_{in}(t') \cdot \int_{t'}^{t'+1} \left(\frac{t-t''}{t} \right)^{\frac{3}{2}} \cdot \text{Exp} \left[- \left(I t + \frac{1}{4d} \right) \cdot \left(\frac{t-t''}{t} \right) - \left(\frac{1}{4d} \right) \cdot \left(\frac{t-t''}{t} \right)^{-1} + \frac{1}{2d} \right] dt''$$

with the substitutions:

$$a = I t + \frac{1}{4d}$$

$$b = \frac{1}{4d}$$

$$x = \frac{t-t''}{t}$$

$$x_1 = \frac{t-t'}{t}$$

$$x_2 = \frac{t-t'-1}{t}$$

$$c_{out}(t) = \frac{1}{\sqrt{4pd}} \cdot \text{Exp} \left[\frac{1}{2d} \right] \cdot \sum_{t'=-\infty}^{t-1} c_{in}(t') \cdot \int_{x_2}^{x_1} x^{-\frac{3}{2}} \cdot \text{Exp} \left[- a x - \frac{b}{x} \right] dx$$

$$= \frac{1}{4} \cdot \frac{\text{Exp}\left[\frac{1}{2d}\right]}{\sqrt{bd}} \cdot \sum_{t'=-\infty}^{t-1} c_{in}(t') \cdot \left\{ \text{Exp}[2\sqrt{ab}] \cdot \text{Erf}\left[\sqrt{a} \cdot x + \frac{\sqrt{b}}{x}\right] - \text{Exp}[-2\sqrt{ab}] \cdot \text{Erf}\left[\sqrt{a} \cdot x - \frac{\sqrt{b}}{x}\right] \right\}_{\sqrt{x_1}}^{\sqrt{x_2}}$$

with $\text{Erf}[y] = \frac{2}{\sqrt{p}} \int_0^y \text{Exp}(-t^2) dt$

The Boxmodel_V3.xls Excel Workbook does not use this closed solution as the calculation accuracy of Excel is not sufficient to deal with the Erf-Functions.

The integral is solved numerically for each year by numerical integration of the previous integral expression instead.