A Model of Quality Distribution in Groundwater With Reference to Natural Tritium Concentration

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Abstract

In real groundwater flow systems, mixing of water due to dispersion normally takes place during flow and makes the transport process of tracers very complex. When considering measured tritium concentrations in river waters and groundwaters, most interpretations have been assumed lumped parameter models such as well-mixed reservoir models. The problem on the distribution of fallout tritium in groundwater flow systems has been left unsolved quantitatively so far.

A simple model of groundwater flow is constructed to analyze the distribution of tritium concentration in unconfined aquifers being supplied uniformly by precipitation. The differential equation describing the distribution of tritium concentration averaged over a vertical section of an aquifer includes a term of macro-dispersion owing to inhomogeneity of the medium. Although in real aquifers the actual dispersivity is not known exactly due to its complex process, an inspection of the distribution of average tritium concentration in such an unconfined aquifer reveals extremely small effects of macro-dispersion on the distribution except near the upper edge of flow such as a divide. The tritium concentration averaged over a section in an aquifer can be thus approximated well by assuming that no mixing takes place at all.

On analysis based on the model, it is proved that the distribution of the average tritium concentration is uniform in the flow direction varying with time, if only the amount of effective water stored in the aquifer per horizontal unit area is uniform. The expression of average tritium concentration indicates that the residence ages of waters crossing any vertical section in an unconfined aquifer have an exponentially declining distribution along the section. The expression is in the same form as that of concentration in output water from a well-mixed reservoir with a turnover time. However, the present model, which is concerned with the spatial distribution of tritium in an aquifer, is basically different from the well-mixed reservoir model. The application of this model is checked in groundwaters in volcanic and alluvial areas and also in river waters in Japan. Many tritium data of groundwaters and river waters are consistent with the exponential prediction of the model.

The method of analysis is extended to the problem of the vertical distribution of tritium concentrations through groundwaters to deeper zones. There is a tendency for tritium concentrations to decline exponentially with depth, with the existence of fallout tritium being limited to the depths of a few hundreds meters or less. This finding has been observed in many districts in Japan, and it could be used to infer a vertical flow rate.

1. Introduction

The present study is concerned with the problem of fallout tritium in groundwater flow systems formed through direct recharge of water from precipitation. In most hydrological studies using natural tracers such as stable isotopes, a main purpose is basically the interpretation of systems containing mixed waters from different sources. However, when radioisotopes such as tritium are used for mixed water systems, mixing processes between waters of different residence time within the systems arise as a complex problem.

Many hydrological studies using fallout tritium have been made mainly with river waters, since Kaufman and Libby (1954) made the first survey on tritium content in waters of the Mississippi

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River. Kaufman and Libby postulated a very rapid mixing of the fallout tritium with the whole groundwater body. How well the well-mixed reservoir model fits, when compared with actual concentrations in river waters, was demonstrated typically through long term data from the Ottawa Valley by Brown (1961). The well-mixed reservoir model has been used for the interpretation of tritium concentrations in many river waters in Japan and it has given good fits for the change with time of actually measured concentrations (Tanaka, et al., 1981), though the assumption of quick mixing throughout the whole system may be doubtful.

On the other hand, tritium concentrations in groundwaters have been measured over a wider range in comparison with those in river waters. However, it is noted that the higher concentrations, which have appeared within shallow zones in many cases, showing approximately a uniform distribution in some cases, are similar to the tritium concentrations in river waters measured at the same time. It seems that the well-mixed reservoir model gives also the upper limit of actual tritium concentrations in both river waters and groundwaters in the western part of Japan in the period before or after of 1980. These facts suggest that the tritium concentrations in most waters in hydrological regime are controlled by some kind of mixing process in relation to an exponential distribution of residence time of waters within a system.

Until recently, little attention had been given to the tritium distribution in groundwaters, mainly since tritium concentration may be seen as one indication of complex mixing processes influenced by large scale inhomogeneity of hydraulic properties involved. Actually, the problem on the tritium distribution in groundwater flow systems has been left unsolved theoretically. Recently the problem on solute transport in inhomogeneous media has been investigated with stochastic modeling by several hydrologists (e.g., Freeze, 1975; Gelhar, et al., 1975; Smith and Schwartz, 1981; Dagan, 1982). In the present paper, a theoretical approach to the problem on the spatial distribution and time variation of tritium concentrations in an unconfined aquifer with a uniform input of tritium from precipitation will be made by investigating how far the influence of the dispersion due to inhomogeneity of media on the tritium distribution extends. The proper expression describing the tritium distribution in an aquifer will be shown finally to have a connection with an exponential distribution of residence time of groundwater crossing any section in the aquifer.

2. Theory

Waters which infiltrate into soils from precipitation may move toward stream channels by a variety of routes. If the layer of soil or weathered rock is thick and of uniform permeability, the subsurface waters move vertically to a zone of saturation and then flow along a curving path to a stream channel. Inhomogeneities of permeability may disrupt this simple pattern of flow. A coarse-grained layer overlying on a relatively fine-grained one may carry a large and rapid flow when an aquifer is formed, while a fine-grained layer having no large pores carries a lesser flow. Figure 1 is a schematic representation of groundwater flows in a gently sloping region, where soil layers play a significant role in draining waters.

In most unconfined aquifers, the horizontal dimension is sufficiently large in comparison with the vertical one. The shallow water approximation is usually made, i.e., flow variables such as Darcy's velocity are averaged over a vertical section and their variations only in the horizontal plane are considered. The seepage analysis method has been developed to obtain horizontal flows of groundwater in particular in inhomogeneous unconfined aquifers reducing the general three-dimensional problem to a two-dimensional one (Youngs, 1986). Here, we will consider a similar method also with regard to the transport of fallout tritium which is contained in waters collected through an aquifer. That is, groundwater sampling for tritium measurement is carried out by
collecting water through a well that fully penetrates the aquifer. Since the measured tritium concentrations are averaged over a vertical section in the aquifer, the effect of inhomogeneity in hydraulic properties such as permeability is to increase the effective dispersivity.

When both the specific discharge through a unit area in the vertical section and the concentration of tritium in pore water are separated into the spatial mean and the spatial fluctuation, the convective flux of tritium averaged over the vertical section includes a term which is the average of product of fluctuations in both the specific discharge and the concentration. This term, which represents just the above mentioned dispersion caused by the inhomogeneity of flow field, corresponds to the so-called macro-dispersion (e.g., Freeze, 1975; Bear, 1979; Smith and Schwartz, 1981).

We consider an unconfined aquifer which is supplied with uniform infiltration $\varepsilon_1$ per unit time through a horizontal unit area over the phreatic surface rests on a semi-permeable layer where water leaks vertically at a rate $\varepsilon_2$ through a horizontal unit area uniformly into the underlying layers, as shown in Figure 2. The groundwater in the unconfined aquifer is assumed to flow steadily in the region bounded at the top by the surface $z = \eta_1(x)$ and at the bottom by the surface $z = \eta_2(x)$, where $z$ is the vertical distance measured upward from a horizontal datum level, and $x$ is the horizontal distance measured positively in the flow direction.

On the condition that the slopes of both surfaces are very gentle, i.e., $\partial \eta_1 / \partial x \ll 1$ and $\partial \eta_2 / \partial x \ll 1$ at a position $x$, the horizontal seepage rate $Q$ per unit width at the position is given by
\[
\frac{dQ}{dx} = \varepsilon_1 - \varepsilon_2. \tag{1}
\]

For the investigation of the horizontal distribution of tritium concentration averaged over each section, we integrate the well-known dispersion-convection equation over a vertical section in the aquifer, providing that the equation is satisfied in some microscopic scale where the medium can be regarded as homogeneous. To simplify, we assume that the spatial variation of porosity is sufficiently small in comparison with that of permeability according to Freeze (1975), and that the contribution of pore scale dispersion, which appears in flows through homogeneous media at ordinary scales in laboratory column experiments, is negligibly small as compared with that of macro-dispersion according to Dagan (1982). Still more, it is supposed that the tritium concentration in water leaking downward through the semi-permeable layer is equal to the average concentration over the section. Given these assumptions, the dispersion-convection equation integrated over the vertical section becomes finally

\[
\frac{\partial C}{\partial t} + u \frac{\partial C}{\partial x} = D_M \frac{\partial^2 C}{\partial x^2} - \left( \lambda + \frac{1}{T} \right) C + \frac{C_R(t)}{T}, \tag{2}
\]

as shown in Appendix I, where

- \(C(x, t)\): the tritium concentration in pore water averaged over the vertical section at the position of \(x\) in the aquifer at the time \(t\),
- \(C_R(t)\): the concentration in recharging water at the phreatic surface at the time \(t\),
- \(n\): the effective porosity,
- \(h(=-h_1-h_2)\): the thickness of aquifer,
- \(u(=-Q/nh)\): the pore water velocity averaged over the section,
- \(T(=nh/\varepsilon_1)\): the ratio of the amount of effective water stored in the aquifer per horizontal unit area to the recharge rate through a horizontal unit area of the phreatic surface,
- \(D_M\): the apparent macroscopic longitudinal dispersion coefficient, and
- \(\lambda\): the radioactive decay constant of tritium (\(=5.6 \times 10^{-2}\) y\(^{-1}\)).

With respect to the macroscopic longitudinal dispersion coefficient \(D_M\), which controls the average concentration \(C(x, t)\), we will refer to the theoretical results of stochastic modeling by Dagan (1982, 1984). In Dagan's work, it is seen that the leading term of macroscopic longitudinal dispersivity which is much larger than that of lateral one depends dominantly on the length scale characterizing the inhomogeneity of permeability in a long time approximation, where advective distance is much greater than the characteristic length of inhomogeneity. Given the expression of dispersivity by Dagan, the macroscopic longitudinal dispersion coefficient could be simplified, such as \(D_M = au\), as a first approximation, where \(a\) is the characteristic length of inhomogeneity in permeability (The scale \(a\) expresses the average distance over which permeabilities are correlated. As the most simplified case, it corresponds to nearly such a length in which a medium is composed of blocks of length \(a\) set at random and with no overlap along any direction). We suppose here that the inhomogeneity scale \(a\) is statistically uniform throughout the aquifer.

For a simple inspection of the effect of macro-dispersion term in equation (2), we consider a specified region in the lower stream of a section located at \(x\), where the average pore velocity can be regarded as constant. Providing that the concentration \(C_R(t)\) of recharging water at the surface changes with a harmonic fluctuation of a period \(\tau_R\) around the mean, we can evaluate the contribution of the term to the formation of averaged concentration \(C(x, t)\), as shown in Appendix I.

The result is summarized as follows: the ratio of the dispersion term to the convection one in equation (2) takes a value of \((1-\beta)/2\), where \(\beta = |1 + 4aD_M/u^2T|^{1/2}; \alpha = 1 + \lambda T\) or \(\alpha = 1 + (\lambda + 2\pi i/\tau_R) T\) for the mean or oscillation component, respectively, and \(i\) is the imaginary unit. It is thus seen that the dispersion term in equation (2) can be ignored, if \(|\beta - 1| \ll 2\). On additional conditions: \(T < 1/\lambda\) and \(\varepsilon_1 > \varepsilon_2\), this is equivalent to the inequality: \(uT/a > 1/2\) or \(x/a > 1/2\) for the mean component of \(C_R(t)\), and equivalent to the inequality: \(x/a > (\sqrt{2} \pi/3)(T/\tau_R)\) for the fluctuation component in the case of a short period, where \(x\) denotes the distance from an impervious edge in the...
upper stream such as a divide. The dispersion term in the equation (2) may be ignored, therefore, except near the edge, as long as the characteristic length $a$ of permeability inhomogeneity is of a similar order as the thickness of aquifer, and $\tau_R$ is not so short in comparison with $T$. Thus, it could be assumed that the contribution of the macro-dispersion term is negligibly small over almost the whole region even with actual variations in $C_R(t)$. Then equation (2) for the average concentration would be re-written as

$$\frac{\partial C}{\partial t} + u \frac{\partial C}{\partial x} = - \left( \lambda + \frac{1}{T} \right) C + \frac{C_R(t)}{T}. \quad (3)$$

For the convenience of integration of equation (3), we will suppose such an aquifer that $T$ is nearly constant in the flow direction, that is, the amount of effective water stored in the aquifer per horizontal unit area is approximately uniform in the direction.

On the initial condition of concentration distribution:

$$C = C_0(x) \quad \text{at} \quad t = 0, \quad (4)$$

the solution is obtained as follows:

$$C(x, t) = \int_0^t C_R(t - \tau) \cdot \exp \left\{ - \left( \lambda + \frac{1}{T} \right) \tau \right\} / T \cdot d \tau$$

$$+ C_0(\xi) \cdot \exp \left\{ - \left( \lambda + \frac{1}{T} \right) t \right\}, \quad (5)$$

where $\xi = f(x, t, u)$ is derived from the integral equation

$$\int_\xi^x \frac{1}{u(x)} \, dx = t, \quad (6)$$

and the expression of $\xi$ is determined by the functional form of $u(x)$. For example, if $u = x/T'$, then $\xi = x \cdot \exp (-t/T')$, or if $u$ is nearly constant, then $\xi \approx x - ut$. The initial value of $C_0(x)$ is constant or a function of $x$ depending on the hydrological state in the initial period, but its value is limited in a definite range without a very singular high value. Then the effects of any term including $C_0(\xi)$ diminishes exponentially according to the factor of $\exp \left\{ - \left( \lambda + \frac{1}{T} \right) t \right\}$ independently of the functional form of $f(x, t, u)$. Therefore, it can be seen that the average tritium concentration $C(x, t)$ does not depend on the space variable $x$ as the time proceeds, as long as $T$ is uniform in the flow direction.

Furthermore, it is especially noted that the apparent transit time of water required to arrive at any section after departing from the phreatic surface has an exponentially declining distribution with the mean of $T$ along the section. The mean age of waters passing across any section is also the mean residence time of waters in the upper reach of the aquifer. It is recognized thus that the water recharged nearby a section contributes dominantly to the average tritium concentration of groundwater at the section, and that the contribution of recharge water to the tritium concentration of groundwater decreases as the distance in the upper stream direction from the section increases. Therefore, it can be seen that the boundary condition at the upper edge does not affect the average tritium concentration in any section sufficiently far from the edge.

In the case of a uniform distribution of concentration, the expression (5) coincides with that of the well-mixed reservoir model, since $T$ is nearly equal to the so-called turnover time defined as the ratio of the amount of water stored in the whole system to the sum of total input or removal rate of
water passing through the system, owing to the assumption of a nearly uniform thickness of the aquifer in this model. However, the present model describing the spatial distribution of tritium concentration is different in the physical meaning from the well-mixed reservoir model. For example, the tritium concentration in river water, which is a reflection of groundwater nearby the river, is influenced dominantly by the water recharged in the vicinity of the river in the case of the present model, whereas it is a consequence of turnover through the whole drainage system in the case of the well-mixed reservoir model.

3. Tritium concentrations in river waters in Japan

River discharge is usually separated into direct runoff and baseflow components in order to treat it simply. In Japan, samples of river waters for tritium measurement are often collected in time except immediately after rainfall so that the data are closely related to the baseflow draining regional groundwaters.

Figure 3 shows the tritium concentrations in river waters which have been measured so far in Japan (Takahashi, et al., 1965, 1969; Inoue, et al., 1978; Chaya, et al., 1978; Tanaka, et al., 1981; Kitaoka and Yoshioka, 1984). The stepwise line in the figure stands for precipitation whose concentrations are obtained by averaging the monthly TU values at Tokyo and at Pohang by the International Atomic Energy Agency, Vienna (1969—1982) with weights of 2 and 1, respectively, i.e., the TU value of precipitation = \[ 12 \times \text{TU value at Tokyo} + \text{TU value at Pohang} \] / 3.

The input for \( C_R(t) \) of recharge water in equation (5) is used after multiplying tritium concentrations of precipitation by monthly weighting coefficients of infiltration recharge, which are

![Fig. 3](image-url)
estimated using average years of monthly precipitation and evaporation by a simplified method (Kitaoka and Yoshioka, 1984). Though the annual allotment of the recharge varies considerably with localities in Japan, it does not affect so significantly the output values estimated by this model in the wet climate of Japan. The output curves with \( T = 2y, 5y, 10y, 20y, 50y \) and \( 100y \) in Figure 3 are obtained by calculating monthly based on ten year averages for the precipitation and evaporation at Tokyo.

It can be confirmed from this figure that the actual TU values of river waters fit well within the envelope curve of the output curves obtained for various values of \( T \). Actual variations of tritium concentrations in individual rivers with time can also be checked to agree nearly with some model output curves in many cases, since the TU values in time series for each river decreased linearly on a log scale with time during the 1970's in many regions in Japan (Tanaka, et al., 1981). It is noted that the mean residence times of water in many river basins have values within a relatively narrow range of several years (about ten years at the longest). This range coincides approximately with that of time constants of tank corresponding to baseflow estimated for many river basins through runoff analyses by tank models (Sugawara, et al., 1974). The time constant of runoff through a tank is equivalent to the turnover time of water in the tank.

It is considered from these facts that this simple model can approximately represent actual runoff processes of tritium, and that the residence time of waters passing through a drainage basin must have a distribution similar to an exponential function reflecting hydrological characteristics of the drainage system.

4. Examples of uniform distribution of tritium concentrations in unconfined aquifers

(1) Upland area in the western foot-hills of Aso volcanoes

Volcanic areas have particular hydrological features such as the shortage of runoff waters in the upper elevations and the abundance of spring waters in the lower regions. In the western foot-hills of the Aso volcanoes, Kumamoto (see Figure 4), there is a triangular upland area with low stream density and many large springs around the upland area (Kayane, et al., 1987). It is clear from this evidence that much supply of water from precipitation and active groundwater flows may occur in the wide upland area. The main water-bearing strata in this area are said to be in the formations consisting mostly of Aso pyroclastic flow deposits.

Water samples for tritium measurement were collected in this area in November, 1984, form 11 stations for river waters, 7 for spring waters and 13 for groundwaters. The sampling stations are shown partly in Figure 5(a), in which the piezometric water level above sea level surveyed by Kumamoto prefectural government is also shown. A divide of groundwater flow system exists a little
apart from the surface divide in the drainage area of River Goshi (Kayane, et al., 1987). Except near the groundwater divide, it is seen from the contour map of water table that the direction of main groundwater flow is southwestwards for the large spring zones in which the spring with the highest discharge is Ezu-ko, indicated as SI in Figure 5(a).

TU values for groundwaters and spring waters in this area are in the range of $2.1 - 19.6$ TU as plotted in Figure 5(b). It is noted that the area is separated clearly into the zone with lower TU values in groundwaters and that with higher ones. The groundwater divide is in the zone with lower TU values, where the groundwater may be in a confined state isolated from the surface zone by the local existence of an overlying consolidated sedimentary layer cut by some faults. For contrast, the direction of a wide belt-like zone with TU values higher than 15 TU (as shown by hatching in the figure) corresponds to the direction of main groundwater flow inferred from the map of water table.

Fig. 5 Groundwater level above sea level (m) surveyed by Kumamoto prefectural government and sampling points (a), and distribution of tritium concentration (TU) (b), in the western foot-hills of Aso volcanoes, Kumamoto. ●: wells, ○: springs, □: rivers. Dotted region in figure (a) indicates groundwater divide. Hatched and dotted regions in figure (b) indicate regions in above 15 TU and in below 10 TU, respectively, for groundwaters and spring waters.
Figure 6 shows the sectional distribution of TU values in groundwaters from wells and spring waters collected within the rectangular region drawn in Figure 5(b) with a projection on the AA'-section whose direction coincides nearly with that of the main groundwater flow. The depth of collecting wells is restricted to the range of the Aso pyroclastic flow deposits over an underlying stratum which acts as an impervious layer shown by shading in the upper figure. The TU values in groundwaters and spring waters in the main flow region are found within a range of about 16.0 ± 3.5 TU. The fluctuation exceeding the accuracy of measurement may be caused by the differences of zones collecting groundwaters, since the pyroclastic rocks are stratified as a result of several volcanic eruptions. Even the uncertainties on the data, the TU values in groundwaters can be regarded as being distributed uniformly over the wide area as a whole.

The total amount of groundwater discharge through springs and wells, most of which are distributed in the lower region of the upland, was estimated at about $8.4 \times 10^8$ m$^3$·y$^{-1}$ by the above mentioned survey in 1985. The amount can be converted roughly into the discharge of 1.3 m·y$^{-1}$ per unit horizontal area, if the drainage region of groundwater is assumed to enclose an area of 25 km × 25 km based on the water table map in Figure 5(a). The groundwater system may therefore be in a state of mass-flow equilibrium, since the annual precipitation is about 2.0 m·y$^{-1}$, and according to Shimotsu, et al. (1979) the evapotranspiration is about 0.7 m·y$^{-1}$ in this area. Such situations wherein the groundwater system is mainly recharged by infiltration from the ground surface and the thickness of aquifer is roughly uniform may cause the nearly uniform distribution in tritium concentration in groundwaters to form over the wide upland area from the point of view of the present theory.

(2) Alluvial fan in Shimada City

Figure 7 shows the area investigated in Shimada City, Shizuoka (see Figure 4), where alluvial deposits brought by the flood of the River Ooi have a thickness of about 50 m on average. In this
TU values in this area are found within a narrow range of 10—15 TU for groundwaters and that of 9—10 TU for river waters. Figure 8 shows the sectional distribution of TU values projected on the AA'-line drawn in Figure 7, whose direction coincides roughly with the main direction of groundwater flows except the upper and lower regions in this figure. The collecting zone of groundwaters is restricted to the alluvium in this area. The uniform distribution of TU values in groundwater is seen more clearly in this area, though somewhat lower TU values are found only nearby the River Ooi implying the local intrusion of river water into the aquifer.

The TU range of groundwaters corresponds to the mean residence time of about 3 y in error by
less than about 1 y, extrapolating model output curves in Figure 3. Supposing an aquifer with a uniform thickness of 40 m and with porosity of 0.3 being supplied almost by uniform recharge intensity from the ground surface, the mean residence time of 3 y can be related to the annual recharge of the order of 4 m·y⁻¹ per unit area over the surface from the present model.

The value of infiltrating recharge estimated by this method exceeds considerably the annual precipitation of about 2.4 m·y⁻¹. Though the condition of estimation is not always satisfactory, this difference of recharge suggests that the infiltration from irrigation may be large. In fact, irrigation waters introduced from the river spread widely over the paddy fields during the rice-growing season, and the ranges of annual variations of groundwater level in some observation wells in this area are about 4 m (as the difference between the highest and lowest levels in a year). If the annual variation of groundwater level is assumed to be caused only by a harmonic variation of infiltration, the amplitude of groundwater level is given by \( A_R/(\pi n_0) \), where \( A_R \) : the amplitude of infiltration intensity [m·y⁻¹], and \( \omega = 2\pi \) [y⁻¹]. The value of \( A_R \) estimated by this method is about 3.8 m·y⁻¹ under \( n = 0.3 \). As the infiltration in the drought season can be considered to be negligibly small, the amplitude of the infiltration intensity may be approximately equal to the annual mean of infiltration. This rough estimation of recharge from the annual variations of groundwater level coincides thus with that obtained using the tritium method.

After all, the uniform distribution of tritium concentration in this area may be owing to the fact that the main recharge to groundwater in this alluvium is due to the infiltration through the surface from both precipitation and irrigation, from the standpoint of the present model.

5. Discussion on vertical distribution of tritium concentrations in groundwaters to deep zones

Tritium concentrations in groundwaters sampled from shallow to deep zones in the western part of Japan in the last decade, are plotted against the depth below the surface in Figure 9. The depth used for plotting is taken to be that of the middle point of a screen collecting water. Open circles and triangles in this figure are waters from Beppu hydrothermal field (see Figure 4), where an alluvial fan composed of volcanic deposits spreads over the area of about 25 km². The triangles represent high temperature waters spouting out through boreholes with boiling from deep zones, and the open circles are other groundwaters including hot waters discharged without boiling in the field. Solid circles are waters including hot waters in other fields: San’in, Rokko, Oita, Kumamoto and Shimada areas (see Figure 4), the last two of which have been discussed in the previous section. Tritium concentrations of spring waters flowing out naturally or through horizontal boreholes at the foot of hills in Beppu and Rokko areas and of rain waters collected at Beppu from 1976 to 1981 are also shown in this figure.

Despite the distribution of sampling locations across the western part of Japan, TU values of non-boiling groundwaters lie within an exponentially decreasing range with depth, as drawn by a dashed line in this figure. The major exceptions are the high temperature waters from Beppu which show relatively high TU values even in deep zones. These waters appear to be anomalous, and will be discussed in another paper.

Considering the rest of the data in Figure 9, we will suppose that a confined aquifer is recharged uniformly from an overlying aquifer through a thin horizontal semi-permeable layer. So far as such a condition is satisfied over a relatively wide region, the model for an unconfined aquifer described in section 2 can be applied to approximate the average tritium concentration in the confined aquifer. The tritium concentration \( C_i(t) \) in the \( i \)-th aquifer, numbered from the unconfined aquifer, is thus given in an expression similar to equation (3)

\[
\frac{dC_i}{dt} = - \left( \lambda + \frac{1}{T_i} \right) C_i + \frac{C_{i-1}}{T_i},
\]  

(7)
where $T_i = n_i H_i / \epsilon_i$; $n_i$ and $H_i$ the porosity and thickness of the $i$-th layer respectively, and $\epsilon_i$ the rate of vertical recharge per unit area from the $(i-1)$-th layer to the $i$-th one.

To simplify, it is first supposed that a system composed of two layers, the first (unconfined) and second aquifers, exists on a horizontal impervious layer. In such a situation, the average concentration $C_2(t)$ in the second aquifer is obtained as

$$C_2(t) = \frac{T_2 C_2(t) - T_1 C_0(t)}{T_2 - T_1}, \tag{8}$$

where $C_0(t)$ is the solution of equation (7) replacing $C_{i-1}$ by $C_R(t)$, that is, $C_0(t)$ is obtained as $C$ of equation (5) by replacing $T$ in the equation with $T_i$, ignoring the second term in the right hand side. This replacement is an imaginary condition such that the $i$-th layer is to be recharged by precipitation directly. It is seen from equation (8) that if $T_2 > T_1$, then $C_2 \sim C_{02}$, whereas if $T_1 > T_2$, then $C_2 \sim C_{01}$. If a layer with a high value of $T$ exists generally in some depth, tritium concentration is thus lowered at or below the layer. As discussed in section 3, most of river waters seem to contain waters mainly originating from the first layer which is supplied by meteoric waters directly. Here, the contribution of second layer being recharged from the first layer to the tritium concentration in discharged water will be inspected, supposing that the discharged water is a mixture of waters through both the first and second layers. As an approximation, if the leakage rate $\epsilon_2$ is in proportion to the difference between average heads of adjacent aquifers and is uniform along the interface, the ratio of horizontal
seepage flow is $Q_2/Q_1 = K_2/K_1$, and that of vertical flow is $e_2/e_1 = K_2/(K_1 + K_2)$, where $K_i$ is the transmissivity of the $i$-th layer. Then the tritium concentration of discharged water $C_D = (C_1Q_1 + C_2Q_2)/(Q_1 + Q_2)$ can be obtained using the relationship (8) as:

$$C_D = (1 - \gamma) C_{01} + \gamma C_{02},$$

where $\gamma = [1 + (k_1/k_2 - n_1/n_2)(H_1/H_2)]^{-1}$; $k_i$: the average hydraulic conductivity of the $i$-th layer. For instance, if the hydraulic conductivity in the first layer is only one order greater than that in the second one under the situation that the porosity and thickness are not so different with each layer, the tritium concentration in water discharged from the first layer contributes dominantly to that of the total discharged water. It is inferred, therefore, that tritium concentrations in many river waters may be more or less determined by the water passing through a permeable layer existing probably in shallow zones.

In many cases, TU values in river waters and groundwaters have gradually changed with time within a small range, though those in rain waters have fluctuated rapidly over a wider range. Such a behavior can be seen by giving some kind of fluctuations to $C_\rho(t)$ in equation (3). For instance, when the concentration in precipitation varies with a harmonic function of an angular frequency $\omega$ with time around its mean, the concentration ratio of the first layer to the precipitation is $(1 + \lambda T_1)^{-1}$ in the mean and is $(1 + \lambda T_1)/(1 + (1 + \lambda T_1)^2 + (\omega T_1)^2)$ in the amplitude. Thus the amplitude of the concentration oscillation in precipitation diminishes and falls off more rapidly for large $\omega$. If the concentration is given by a Fourier series, the higher harmonics disappear more rapidly in deep layers.

So, provided that the tritium concentration in the $m$-th layer is sufficiently smoothed with time, then the concentration in $(m+i)$-th layer is expressed as follows:

$$C_{m+i}/C_m = \prod_{j=m+1}^{m+i} \left[ 1 - \frac{\lambda n_j H_j}{\epsilon_j} \right],$$

since the concentrations between adjacent layers in such zones have a relationship: $(C_i - C_{i-1})/C_i = \lambda n_i H_i/\epsilon_i$, as obtained from equation (7). If a formation composed of many stratified layers is taken as a continuous medium with vertical variations, the relationship can be expressed as

$$\frac{dC(z)}{dz} = -\lambda \frac{n(z)}{\epsilon(z)} C(z),$$

which is independent of horizontal seepage flows in the medium. Then the concentration at the depth $z$ in the continuum is expressed as

$$\frac{C(z)}{C(z_m)} = \exp \left[ -\lambda \int_{z_m}^{z} \frac{n(z)}{\epsilon(z)} dz \right],$$

which is equivalent to equation (10), where $z_m$ is the depth of the lower boundary of the $m$-th layer. Thus the tendency of exponential decrease of concentrations with depth can be seen in such a region where the value of $\epsilon/n$ is nearly constant.

The dotted line in Figure 9 is the representation of equation (12) calculated with the condition that $n = 0.3$, $\epsilon = 1$ m·y$^{-1}$ and $C_m = 25$ TU at the depth $z_m = 150$ m, since the formation boundary occurs at about this depth in the southern part of the Beppu area. The calculated curve seems to coincide approximately with the dashed line representing the upper limit of measured TU values at each depth. As the dotted line is obtained under the condition that the infiltration rate is relatively high and the tritium rich zone is relatively thick, the line would indicate approximately the upper limit of TU values at each depth in most cases of stratified formations. It can be considered thus that...
the existence of fallout tritium must be limited within shallow zones in depth of several hundreds meters or less in most districts in Japan.

However, in some regions, tritium is not always detected even in shallower zones. For example, values of about 1 TU are measured at the depth of 100 m in the Tertiary in Oita Plain. In the Beppu alluvial fan, tritium can hardly be detected even at depths less than 100 m along the coastal region, but just behind the coastal area, a conspicuous horizontal distribution from low to high TU values are found in hot waters collected at similar depths. The model discussed above may not be applied to waters that have been in a confined state isolated from overlying aquifers. In such a situation, the horizontal tritium distribution may be predominantly controlled by cross-flow from another aquifer in the upper reach where tritium concentration varies with time. The macro-dispersion during flow may play a significant role in forming the tritium distribution in such the confined condition.

6. Conclusion

In the present paper, we have discussed the distribution of tritium concentration in groundwater in a somewhat limited case where water in an aquifer with nearly uniform thickness is recharged uniformly through the upper surface from above. We have seen that there is little effect on the distribution of average tritium concentration over a vertical section due to macro-dispersion owing to inhomogeneity of the medium. The tritium concentration in groundwater during flow can therefore be treated relatively simply. Under such a flow situation, the model predicts (a) the uniform distribution of tritium concentration in the flow direction (which is observed in some groundwaters in volcanic and alluvial areas), and (b) the exponential distribution of apparent transit time of water crossing any section through the aquifer. Furthermore, the tendency of exponentially decreasing of tritium concentrations with depth in a stratified formation (which has been measured in many areas) can be modeled by applying a similar method of analysis to a more than one layer system.

The modeling shows that the tritium concentration averaged over any section in an unconfined aquifer is in the same form apparently as that for the output water from a well-mixed reservoir. However, the present model is basically different from the well-mixed reservoir model. For example, the tritium concentration of river water is a consequence of a turnover time throughout the whole drainage system in the case of the well-mixed reservoir model. However, in the model presented here, the river water tritium is a consequence of groundwater flow with a mean residence age in the aquifer near the river, that is, it is dominantly influenced by the water recharged in the vicinity of the river.

Although the modeling assumptions and conditions are relatively simple, the predicted exponential distribution in residence time of water is consistent with the actual distribution of tritium concentration in many river waters and groundwaters. The measured tritium concentrations in river waters and groundwaters in Japan have never exceeded the upper limit predicted by the modeling. This model may be widely appreciable to tritium transport in groundwater and runoff processes.

However, for the problem of the horizontal distribution of tritium concentration observed in a confined aquifer in the Beppu hydrothermal field, more regional phenomena with tritium must be determined correctly including high temperature waters which contain tritium significantly through deeper zones in the upper region of the field. Many individual problems may need to be solved in regard to mixing processes during groundwater flow due to each hydrological characteristics in particular regions.

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Appendix I: Equation for tritium concentration averaged over a vertical section in an unconfined aquifer.

In a steady state of groundwater flow in an unconfined aquifer of unit width, the continuity equations of water and tritium in a microscopic scale are given by

\[ \frac{\partial q_x}{\partial x} + \frac{\partial q_z}{\partial z} = 0, \]  
(A1-1)

\[ \frac{\partial nc}{\partial t} = - \left( \frac{\partial j_x}{\partial x} + \frac{\partial j_z}{\partial z} \right) - \lambda nc, \]  
(A1-2)

respectively, where \( c \) is the concentration of tritium in pore water, the components of \( (q_x, q_z) \) and \( (j_x, j_z) \) are the volumetric discharge of water and the tritium flux, respectively, crossing a unit area of vertical and horizontal sections in the aquifer per unit time. The tritium flux vector \( (j_x, j_z) \) is usually given as the sum of convective flux vector \( (j_{cx}, j_{cz}) \) \( = (c q_x, c q_z) \) and dispersive one \( (j_{dx}, j_{dz}) \) in a homogeneous medium.

The integrations of (A1-1) and (A1-2) over the vertical section of the aquifer are

\[ \frac{\partial}{\partial x} \int_{\eta_2}^{\eta_1} q_x \, dz - q_x |_{\eta_2}^{\eta_1} \frac{\partial \eta_1}{\partial x} + q_x |_{\eta_2}^{\eta_1} \frac{\partial \eta_2}{\partial x} + q_x |_{\eta_1}^{\eta_2} - q_x |_{\eta_1}^{\eta_2} = 0, \]  
(A1-3)

\[ \frac{\partial}{\partial t} \int_{\eta_2}^{\eta_1} nc \, dz = - \frac{\partial}{\partial x} \int_{\eta_2}^{\eta_1} j_{cx} \, dx + j_{cx} |_{\eta_1}^{\eta_2} \frac{\partial \eta_1}{\partial x} - j_{cx} |_{\eta_1}^{\eta_2} \frac{\partial \eta_2}{\partial x} - j_{cx} |_{\eta_1}^{\eta_2} + j_{cx} |_{\eta_1}^{\eta_2} \] 
\[ - \frac{\partial}{\partial x} \int_{\eta_2}^{\eta_1} j_{dx} \, dx + j_{dx} |_{\eta_1}^{\eta_2} \frac{\partial \eta_1}{\partial x} - j_{dx} |_{\eta_1}^{\eta_2} \frac{\partial \eta_2}{\partial x} - j_{dx} |_{\eta_1}^{\eta_2} + j_{dx} |_{\eta_1}^{\eta_2}, \]  
(A1-4)

respectively. Defining the average of any scalar \( A(z) \) over the vertical section as

\[ \overline{A} \equiv \frac{1}{\eta_1 - \eta_2} \int_{\eta_2}^{\eta_1} A(z) \, dz, \]  
(A1-5)

\( A(z) \) is assumed to be separated into the average and the fluctuating parts, such as \( A(z) = \overline{A} + A'(z) \). Then the average of integrand in the first term in the right hand side of equation (A1-4) becomes \( \overline{j_{cx}} = q_x \bar{c} + q_x C' \). The second term in this representation can correspond to the macroscopic dispersion. According to Dagan (1982, 1984), after a long period of time, where advective distances are much greater than the characteristic length of inhomogeneity of medium, the macro-dispersion apparently approaches the Fickian type, and the macroscopic longitudinal dispersivity is at least one order greater than that in pore scale dispersion. Then we would express the term \( q_x C' \) by \( nDM(\partial C/\partial x) \) and ignore all the terms relating to the pore scale dispersion \( (j_{dx}, j_{dz}) \) in equation (A1-4), where \( D_M \) is the apparent macroscopic longitudinal dispersion coefficient. To simplify furthermore, it is assumed that
\[ \partial \eta_1 / \partial x < 1 \text{ and } \partial \eta_2 / \partial x < 1. \] Putting boundary conditions at the top and bottom as follows:

\[ q_1 | \eta_1 = - \varepsilon_1, \quad jC_1 | \eta_1 = - \varepsilon_1 C_R \text{ at } z = \eta_1, \]

\[ q_2 | \eta_2 = - \varepsilon_2, \quad jC_2 | \eta_2 = - \varepsilon_2 \bar{c} \text{ at } z = \eta_2, \]

then equations (A1-3) and (A1-4) are reduced to

\[ \frac{\partial q_2}{\partial x} = \frac{\varepsilon_1 - \varepsilon_2}{\eta_1 - \eta_2}, \]

\[ \frac{\partial \bar{c}}{\partial t} + \frac{n q_2}{\partial x} = D_M \frac{\partial^2 \bar{c}}{\partial x^2} + \frac{\varepsilon_1}{n (\eta_1 - \eta_2)} (C_R - \bar{c}) - \lambda \bar{c} \]

respectively. The average concentration \( \bar{c} \) is indicated by the symbol \( C \) for the convenience of description in section 2.

**Appendix II: Evaluation of the dispersion term in equation (2)**

When the surface concentration \( C_R(t) \) in an aquifer varying with time is given by a harmonic oscillation around a mean: \( C_R = A_R \exp(i\omega t) \), the average concentration \( C(x, t) \) over a vertical section in the aquifer can be assumed in the type: \( C(x) = A(x) \exp(i\omega t) \), since equation (2) is linear on the condition that \( u \) is constant. Then the equation (2) can be separated into two ordinary differential equations for \( C_m(x) \) and \( A(x) \). Both the equations are in the same form as

\[ \frac{\partial y}{\partial x} = D_M \frac{\partial^2 y}{\partial x^2} - \frac{\alpha}{T} (y - y_\infty), \]

where \( y_\infty = C_{Rm}/A \) and \( \alpha = 1 + \lambda T \) for \( y = C_m \), or \( y_\infty = A_R/A \) and \( \alpha = 1 + (\lambda + i\omega)T \) for \( y = A \). The general solution is given as

\[ y - y_\infty = \text{const} \cdot \exp \left[ (1 - \beta) ux / (2D_M) \right], \]

where \( \beta = [1 + 4\alpha D_M / u^2 T]^{1/2} \). The ratio of each term in (A2-1) can be got by substituting the solution into it as follows: \( 2 = (1 - \beta) + (1 + \beta) \) in order of the terms in the equation. Therefore, if \( | \beta - 1 | \ll 2 \), i.e., \((1 + \lambda T)D_M / u^2 T \ll 2 \) for \( y = C_m \), or \((1 + \lambda T)D_M / u^2 T + (\omega D_M / 3u)^2 \ll 2 \) for \( y = A \), then the contribution of the macrodispersion term can be ignored.

**References**


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