On the conversion of tritium units to mass fractions for hydrologic applications

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We develop a general equation for converting laboratory-reported tritium levels, expressed either as concentrations (tritium isotope number fractions) or mass-based specific activities, to mass fractions in aqueous systems. Assuming that all tritium is in the form of monotritiated water simplifies the derivation and is shown to be reasonable for most environmental settings encountered in practice. The general equation is nonlinear. For tritium concentrations c less than 4.5×10^{12} tritium units (TU) – i.e. specific tritium activities $< 5.3 \times 10^{11} \text{ Bq kg}^{-1}$ – the mass fraction w of tritiated water is approximated to within 1 part per million by $w \approx c \times 2.22293 \times 10^{-18}$, i.e. the conversion is linear for all practical purposes. Terrestrial abundances serve as a proxy for non-tritium isotopes in the absence of sample-specific data. Variation in the relative abundances of non-tritium isotopes in the terrestrial hydrosphere produces a minimum range for the mantissa of the conversion factor of [2.22287; 2.22300].

Keywords: contaminant plumes; groundwater; hydrogen-2; hydrogen-3; mass fractionation; radioactive nuclides; tritium units

1. Introduction

Tritium, a radioactive isotope of hydrogen, is a useful tracer in hydrologic and oceanographic studies [1–3]. Tritium is integral to the water molecule and detectable at exceedingly low levels [4]. Natural cosmogenic processes produce tritium in the upper atmosphere [5]. Rutherford et al. were the first to produce anthropogenic tritium – although initially they did not recognise it as such [6]. Anthropogenic production completely overwhelmed cosmogenic production during the period of atmospheric nuclear-weapons testing (1945–1980) [7]. Nuclear power generation and fuel reprocessing continue to augment natural production [4]. At sufficiently high levels, tritium is a regulated contaminant [8].

Rigorous modelling of tritiated groundwater and soil–water transport by advection–diffusion processes require the conversion of analytical tritium units (TU) to mass-fraction (or mole fraction) units [9] (or at minimum a demonstration of their linear interdependence). Estimating amounts of tritium in contaminant plumes or fluxes of tritium across compliance and other boundaries of interest – the land–atmosphere interface, for example – requires similar conversions to mass (or

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mole) quantities of tritium [10]. The conversion of analytic to mass units is unobvious, nonlinear, and dependent on assumptions that are rarely specified. The full range of activities in pristine to highly contaminated systems requires a general approach. To be useful, the approach must be practical yet rigorous, with well-defined limitations. A thorough review of the literature did not locate any such conversions. This note documents the derivation of a broadly applicable conversion from analytical units to mass-fraction units for tritiated water in the terrestrial hydrosphere.

2. Derivation

We denote the tritium subset of hydrogen atoms by ^3H , the group of protium (^1H) and deuterium (^2H) atoms by $^{1,2}\text{H}$ (for our purposes constituting the ‘non-tritium’ subset of hydrogen atoms), and the complete set of relevant hydrogen atoms by $^{1,2,3}\text{H}$. Heavier isotopes of hydrogen have been synthesised, but are assumed unimportant in hydrologic contexts as their half-lives are shorter than 10^{-21} s. Similarly, we denote the complete set of environmentally persistent oxygen atoms, ^{16}O , ^{17}O , and ^{18}O (see discussion), by $^{16,17,18}\text{O}$ or, for simplicity, O. We thus assume that tritiated water (either $^{1,2}\text{HO}^3\text{H}$ or $^3\text{H}_2\text{O}$) plus non-tritiated water ($^{1,2}\text{H}_2\text{O}$) form the complete set of water molecules ($^{1,2,3}\text{H}_2\text{O}$). We perform calculations for a representative sample of water from the system under consideration, for which the relative abundances of the non-tritium atoms making up the water are evaluated using their standard atomic weights and then further evaluated according to the range of values reported for terrestrial hydrologic samples.

The SI unit of activity is the Becquerel (Bq), a hypothetical number of atoms having an instantaneous decay rate of one per second. Concentrations of tritium from environmental analytical laboratories are most commonly reported in TU [11], where one TU is defined as a particular (SI) number fraction (X) unit (specifically, a particular *isotope number fraction*) equal to one tritium atom per 10^{18} hydrogen atoms [11,12]. The TU arose from Libby et al.’s estimate of the natural abundance of tritium in Norwegian surface water, which was taken to approximate secular equilibrium between cosmogenic production and radioactive decay [13]. On a mass-of-water basis, 1 TU is approximately equal to 0.118 Bq kg^{-1} . The conversion is imprecise due to uncertainty in the decay rate of tritium ($4500 \pm 8 \text{ d}$) [14]. In contrast, the conversion of tritium concentration to mass fraction carries with it the relatively high precision of atomic mass determinations.

The derivation presented here is in terms of atomic quantities, which will not change with further refinements in decay rate. Discrepancies between the relative abundance of the non-tritium isotopes in the water sampled versus those assumed for calculations represent the outstanding source of uncertainty in analysis that follows, unless these abundances are independently assessed. In this latter case, the independently determined abundances of the non-tritium isotopes can be substituted for the terrestrial hydrospheric-water values in the equations that follow. The following derivation is for water only, meaning that the mass of solutes is either negligible or separately taken into account (see discussion). Additionally, measurement errors in any analytically determined quantity amounts (bulk sample mass, for example) are not considered.

Further assuming (see discussion) that

- (1) all tritium exists as monotrithiated water, i.e. as the molecule $^{1,2}\text{H}-^{16,17,18}\text{O}-^3\text{H}$ ($^{1,2}\text{HO}^3\text{H}$),
- (2) deuterium and protium are uniformly distributed throughout all water molecules, i.e. the probability that a given non-tritium hydrogen is either protium or deuterium does not depend on the presence or absence of tritium as the other hydrogen atom in the molecule,
- (3) the relative abundance of the three oxygen isotopes is similarly independent of the distribution of tritium,
- (4) the molar mass of $^{1,2}\text{H}$ in the modelled sample S, $M(^{1,2}\text{H})_S$, is equal to that of hydrogen in water of the terrestrial hydrosphere T, $M(^{1,2}\text{H})_T$, which – corresponding to the 1999 *standard*

atomic weight compilation – has a ‘standard’ value of $1.00794 \cdot 10^{-3} \text{ kg mol}^{-1}$, but ranges at least from 1.007904 to $1.008003 \cdot 10^{-3} \text{ kg mol}^{-1}$ [15],

- (5) the molar mass of $^{16,17,18}\text{O}$ in the modelled sample S, $M(\text{O})_S$, is equal to that of oxygen in water of the terrestrial hydrosphere T, $M(\text{O})_T$, which has a ‘standard’ value of $15.9994 \cdot 10^{-3} \text{ kg mol}^{-1}$ but ranges at least from 15.99906 to $15.99946 \cdot 10^{-3} \text{ kg mol}^{-1}$ [15], and
- (6) the molar mass of ^3H , $M(^3\text{H})$, is equal to $3.016049 \cdot 10^{-3} \text{ kg mol}^{-1}$ [16];

the analysis can proceed as follows:

The molar mass of monotrinitated water ($^{1,2}\text{HO}^3\text{H}$) is

$$M(^{1,2}\text{HO}^3\text{H}) = M(^{1,2}\text{H})_T + M(\text{O})_T + M(^3\text{H}),$$

or, applying the ‘standard’ values cited above

$$M(^{1,2}\text{HO}^3\text{H}) = (1.00794 + 15.9994 + 3.01605) = 20.0234 \cdot 10^{-3} \text{ kg mol}^{-1}.$$

Similarly, the molar mass of non-tritiated ($^{1,2}\text{H}_2\text{O}$) water is

$$M(^{1,2}\text{H}_2\text{O}) = 2 \times M(^{1,2}\text{H})_T + M(\text{O})_T,$$

or, again applying the ‘standard’ values,

$$M(^{1,2}\text{H}_2\text{O}) = (1.00794 + 1.00794 + 15.9994) = 18.0153 \cdot 10^{-3} \text{ kg mol}^{-1}.$$

From its definition, the concentration (isotope number fraction) known as TU equals exactly one tritium atom per 10^{18} hydrogen atoms. Thus, if the water sample S has a tritium concentration $c(^3\text{H})_S$ of 1 TU, one can write:

$$c(^3\text{H})_S = X(^3\text{H})_S = \frac{N(^3\text{H})_S}{N(^{1,2,3}\text{H})_S} = \frac{1}{10^{18}} = 1 \text{ TU},$$

where $N(^i\text{E})_S$ denotes the number of entities (here atoms of specific isotopes or isotope groups) of element E having mass number i (protons plus neutrons) in sample S [12].

Regardless of isotopic species, each water molecule in sample S contains two hydrogen atoms. Thus,

$$2 \times N(^{1,2,3}\text{H})_S = N(^{1,2,3}\text{H}_2\text{O})_S, \text{ from which } \left(\frac{2N(^{1,2,3}\text{H})_S}{N(^{1,2,3}\text{H}_2\text{O})_S} \right) = 1, \quad (1)$$

and

$$2 \times N(^{1,2}\text{H})_S = N(^{1,2}\text{H}_2\text{O})_S, \text{ from which } \left(\frac{2N(^{1,2}\text{H})_S}{N(^{1,2}\text{H}_2\text{O})_S} \right) = 1, \quad (2)$$

in which $N(\text{Q})_S$ specifies the number of atoms or molecules in the specified isotope group *or isotopologue group* Q ($\text{E} \in \text{Q}$) in sample S.

Then, per mole (abbreviated mol) of sample water (containing both tritiated and non-tritiated isotopologues) with tritium isotope number fraction $X(^3\text{H})_S$, the mass of tritiated water, $M(^{1,2}\text{HO}^3\text{H})_S$, is:

$$\begin{aligned} \left(\frac{M(^{1,2}\text{HO}^3\text{H})_S}{\text{mol}(^{1,2,3}\text{H}_2\text{O})_S} \right) &= \left(\frac{N(^3\text{H})_S}{N(^{1,2,3}\text{H})_S} \right) \times \left(\frac{N(^{1,2}\text{HO}^3\text{H})_S}{N(^3\text{H})_S} \right) \times \left(\frac{2N(^{1,2,3}\text{H})_S}{N(^{1,2,3}\text{H}_2\text{O})_S} \right) \\ &\times \left(\frac{N_A N(^{1,2,3}\text{H}_2\text{O})_S}{\text{mol}(^{1,2,3}\text{H}_2\text{O})_S} \right) \times \left(\frac{M(^{1,2}\text{HO}^3\text{H})_S}{N_A N(^{1,2}\text{HO}^3\text{H})_S} \right), \end{aligned} \quad (3)$$

where N_A is the number (the Avogadro constant, tying atomic to macroscopic quantities) equal to the number N of atomic entities per mole [12]. The first term on the right-hand side of (3) is

$X(^3\text{H})_S$, the tritium isotope number fraction pertaining to the sample. The second and third terms are statements of the number of the different types of hydrogen isotopes (isotope groups) in their respective water isotopologues (by Equation (1), for example). The fourth states the equivalency relationship between the Avogadro constant and the mole. The fifth gives the molar mass of the tritiated water in the sample.

Absent sample-specific data, the molar mass of the tritiated water in the sample is approximated using standard values for molar masses of non-tritium isotopes as reported for water in the terrestrial hydrosphere, together with the molar mass of tritium:

$$\left(\frac{M(^{1,2}\text{HO}^3\text{H})_S}{N_A N(^{1,2}\text{HO}^3\text{H})_S} \right) \cong \left(\frac{M(^{1,2}\text{HO}^3\text{H})_T}{N_A N(^{1,2}\text{HO}^3\text{H})_T} \right). \quad (4)$$

Using the values given above, with tritium concentration c expressed in TU, Equation (3) (with (4)) gives the molar mass of tritiated water in the sample:

$$\left(\frac{M(^{1,2}\text{HO}^3\text{H})_S}{\text{mol}(^{1,2,3}\text{H}_2\text{O})_S} \right) = c \times 2 \left(\frac{20.0234 \times 10^{-3} \text{ kg}(^{1,2}\text{HO}^3\text{H})_S}{1 \times 10^{18} \text{ mol}(^{1,2,3}\text{H}_2\text{O})_S} \right). \quad (5)$$

In the same mole of water, the number fraction of non-tritiated water is the complement of the number fraction of tritiated water, i.e. $X(^{1,2}\text{H}_2\text{O}) = 1 - X(^{1,2}\text{HO}^3\text{H})$. Using this fact, the molar mass of non-tritiated water is similarly given by

$$\begin{aligned} \left(\frac{M(^{1,2}\text{H}_2\text{O})_S}{\text{mol}(^{1,2,3}\text{H}_2\text{O})_S} \right) &= \left(1 - \frac{N(^3\text{H})_S}{N(^{1,2,3}\text{H})_S} \right) \times \left(\frac{N(^{1,2}\text{H}_2\text{O})_S}{2N(^{1,2}\text{H})_S} \right) \times \left(\frac{2N(^{1,2,3}\text{H})_S}{N(^{1,2,3}\text{H}_2\text{O})_S} \right) \\ &\times \left(\frac{N_A N(^{1,2,3}\text{H}_2\text{O})_S}{\text{mol}(^{1,2,3}\text{H}_2\text{O})_S} \right) \times \left(\frac{M(^{1,2}\text{H}_2\text{O})_S}{N_A N(^{1,2}\text{H}_2\text{O})_S} \right). \end{aligned} \quad (6)$$

Assuming the previously shown approximation derived from standard terrestrial values, Equation (6) expressed in TU gives for the molar mass of non-tritiated water in the sample:

$$\left(\frac{M(^{1,2}\text{H}_2\text{O})_S}{\text{mol}(^{1,2,3}\text{H}_2\text{O})_S} \right) = (10^{18} - c) \times \left(\frac{18.0153 \times 10^{-3} \text{ kg}(^{1,2}\text{H}_2\text{O})_S}{10^{18} \text{ mol}(^{1,2,3}\text{H}_2\text{O})_S} \right). \quad (7)$$

The mass fraction of tritiated water to total mass of water $w(^{1,2}\text{HO}^3\text{H})_S$ is, by definition

$$w(^{1,2}\text{HO}^3\text{H})_S = \left(\frac{M(^{1,2}\text{HO}^3\text{H})_S}{M(^{1,2}\text{HO}^3\text{H})_S + M(^{1,2}\text{H}_2\text{O})_S} \right). \quad (8)$$

Substituting from Equations (7) and (5), and simplifying, provides the general quantity equation result:

$$w(^{1,2}\text{HO}^3\text{H})_S = \left(\frac{(c \times 20.0234/5 \times 10^{17})}{(c \times 20.0234/5 \times 10^{17}) + ((10^{18} - c) \times 18.0153/10^{18})} \right). \quad (9)$$

The conversion depends nonlinearly on the tritium concentration. However, for tritium concentrations (number fractions $X(^3\text{H})$) greatly less than $X(^3\text{H}) = 1 = 10^{18}$ TU, the mass fraction of tritiated water in sample S, $w(^{1,2}\text{HO}^3\text{H})_S$ is approximated by

$$w(^{1,2}\text{HO}^3\text{H})_S \approx \left(\frac{(c \times 20.0234)}{18.0153 \times (5 \times 10^{17})} \right) \approx (2.22293 \times 10^{-18}) \times c, \quad (10)$$

which is accurate to 1 ppm for $c \leq 4.5 \times 10^{12}$ TU (5.3×10^{11} Bq kg⁻¹).

3. Natural isotopic variation

The relative abundances of the stable isotopes constituting water in the hydrologic cycle are not constants of nature, but instead vary both systematically from location to location across landscapes around the globe (and through time) as well as in response to local geochemical conditions [15]. As already mentioned, the relative abundances of the non-tritium isotopes in a given sample of water can be directly measured and substituted in the final terms of Equations (3) and (6), which generally will result in different, more accurate values for the numerical coefficients appearing in the numerators on the right-hand sides of Equations (5) and (7), and hence (9) and (10). In the derivation of Equations (9) and (10), estimates of the relative abundances of hydrogen and oxygen isotopes in water of the hydrologic cycle served as surrogates in the absence of sample-specific data.

Coplen et al. [15] placed the ‘standard’ values (and standard ranges) of molar masses for the isotopes of oxygen, hydrogen, and other elements in the context of the reported range of variations in the terrestrial hydrosphere. These ranges will likely expand as data accrue from additional locations. The ‘standard’ values that were used to arrive at the coefficients presented here lie within, but not necessarily at the centre of, the ranges for terrestrial hydrospheric water. Substituting minimum and maximum values (as cited in the previous section) for the ‘standard’ values results in a minimum corresponding range in the coefficient (on the right-hand side of Equation (10)) expressed in bracket form as [nominal minimum; nominal maximum] of $[2.22287 \times 10^{-18}; 2.22300 \times 10^{-18}]$. This represents a range in the coefficient (relative to its midpoint) of 58 ppm.

4. Discussion

Given three isotopic species of hydrogen (^1H , ^2H , ^3H) and three environmentally persistent isotopes of oxygen (^{16}O , ^{17}O , ^{18}O), there are 18 distinct isotopic species of water, half of which contain tritium. (In addition to the heavier hydrogen isotopes already mentioned, 10 isotopes of oxygen other than ^{17}O exist, but are also relatively short-lived, with half-lives ranging from $\sim 10^{-21}$ to ~ 122 s.) Rather than accounting for all nine (thirty-nine) tritium-containing species, the problem is made tractable by assuming that all aqueous tritium exists as ^{16}O -, ^{17}O -, and ^{18}O -variants of monotritiated water (assumption (1)). Neglecting minor isotopic variations in hydrogen–oxygen bond dynamics, and assuming that isotopic exchange has proceeded to equilibrium, the fraction of bitritiated water is given by the joint probability that both hydrogen atoms in a given water molecule are tritium, which, to a first approximation is $(c \times 10^{-18}) \times (c \times 10^{-18}) = c^2 \times 10^{-36}$. This is small enough to be ignored.

The assumption (4) that the naturally occurring blend of hydrogen isotopes has a molar mass unaffected by tritium is justified by tritium’s exceedingly low natural abundance. The estimated total natural inventory of tritium in the hydrosphere (here restricted to oceans plus terrestrial water) and the atmosphere is ~ 2.4 and ~ 4.2 kg, respectively [17,18]. These values represent a balance between rates of cosmogenic production in the upper atmosphere, mixing into the rest of the hydrologic cycle (principally the oceans), and radioactive decay. The hydrologic inventory of non-tritium hydrogen is greater than that of tritium, both natural and anthropogenic, by more than 16 orders of magnitude [7,18].

The general expression (9) breaks down at sufficiently high tritium activities. In practice, even the most highly contaminated environments will rarely violate assumption (1). For example, groundwater near the Hanford plutonium–uranium extraction facility, which operated from 1956 to 1990, had a maximum sampled tritium concentration in 1998 of $\sim 1.2 \times 10^6$ TU (reported as 3,870,000 pCi l^{-1}) [19]. The neglect of bitritiated water is justified, as its fractional abundance is

only about 1.4×10^{-24} ($= c^2 \times 10^{-36}$). A potentially more serious issue is the neglect of hydrogen atoms in OH^- and H^+ ions. Most groundwaters are naturally buffered to nearly neutral conditions [20]. At neutral pH, the fraction of tritium in the two ions can be shown to be about $c \times 1.8 \times 10^{-9}$. This fraction, negligible for neutral solutions, grows exponentially as pH deviates from neutrality. Disposed effluents at Hanford included highly concentrated sodium hydroxide and nitric acid solutions [21]. Neglect of these ions would not be justified where such wastes entered the ground. This is in addition to the considerations of solute masses mentioned above.

Three points merit emphasis. First, the relevant mass fraction in hydrologic transport problems is the mass ratio of tritium-bearing solute (here, tritiated water) to total solution (overwhelmingly non-tritiated water) by which the transport takes place. Second, one molecule of water, the transporting substance, contains two hydrogen atoms. Third, the TU is defined on the basis of atoms, not molecules. Mistaking tritiated *water* transport for tritium *atom* transport results in a fractional error equal to the mass ratio of monotritiated water to a single atom of tritium, or roughly $20 \div 3 = 6\frac{2}{3}$. Overlooking the second and third points can produce a factor-of-two error. Compounding the errors can cause transport calculations to be off by a factor of $\sim 13\frac{1}{3}$. Such mistakes are commonly made, with order-of-magnitude consequences.

The conversion from measured specific activities (Bq kg^{-1}) to tritium concentrations (TU) carries with it the uncertainty in the half-life of tritium. The currently recommended value [14] is uncertain to more than 1 part in 600. The uncertainty in applying the linear approximation (Equation (10)) is therefore not limiting.

5. Conclusions

Careful accounting and due attention to assumptions, definitions, and isotopic speciation leads to a general equation for converting between tritium concentrations and mass fractions for hydrologic applications. The assumptions are reasonable for a broad range of environmental conditions. While the general relationship is nonlinear, its linear approximation is accurate to one part per million and applicable to most situations likely to be encountered in practice. The natural variation in the relative abundances of non-tritium isotopes making up hydrologic water increases the uncertainty by about a factor of at least fifty-eight.

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