

Modelling the tritium release from underground nuclear tests performed at Mururoa and Fangataufa: a conservative estimate based on uncertain data

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Abstract Tritium (^3H) release from underground nuclear tests was assessed by a model chain using conservative estimates on the models and parameters in cases where data are not available. The ^3H inventory of individual tests, analytical expressions for ^3H release from the vicinity of each test location and its migration through the fractured volcanic rocks and overlying highly porous carbonate layers, are described by double and single porosity models building the foundation of nuclide flux calculations to the lagoon or ocean. Despite the scarce data, the model chain delivers reliable results and consistent predictions for ^3H release into the biosphere, overestimating measured data due to conservative but not unrealistic assumptions.

INTRODUCTION

Underground nuclear tests performed at the atolls of Mururoa and Fangataufa left behind a radionuclide legacy within the volcanic rock and overlying carbonate layers. These radionuclides will migrate to the lagoon and the ocean. After the definitive end of the French atomic bomb tests at the atolls, data were made available to the International Atomic Energy Agency (IAEA) and the International Geological Commission (IGC) to allow the investigation of the radiological situation and the long term effects of these underground tests (Perrochet & Tacher, 1997; Fairhurst *et al.*, 1998; IAEA, 1998). The data delivered include, e.g. the number and type of tests, their classification into a yield range, and the area where they were carried out (French Liaison Office, 1996). Estimates were made of their influence on the rocks and the accompanying change of water flow (Fairhurst *et al.*, 1998), the initial radionuclide inventory for individual tests and the nuclide partition between rubble, lava and water (i.e. dissolved) (IAEA, 1998), which are necessary to allow a conservative estimate of the long-term radionuclide release to the biosphere.

The total of 147 underground tests are divided into seven test categories with specific characteristics. Three categories, totalling 137 tests, are relevant with respect to ^3H migration. These are nuclear tests that “behaved as assumed”, tests where the chimney reached the top of the volcanics (CRTV), and tests where the volcanic cover did not behave as assumed (“leaky”). For each category a source term and a related transport parameter set is defined. In total the release of 35 radionuclides to the ocean or the lagoon was estimated in the summing up of releases calculated for individual tests (Hadermann & Pfingsten, 1998). Of special interest is the ^3H release. It is produced partly by reaction in the thermo-nuclear fuel and is also used to boost the

fission yield. ^3H is non-sorbing and there is unequivocal evidence of migration of ^3H from underground into the lagoon, resulting in elevated ^3H concentrations in the lagoon, that are not influenced by former surface tests. ^3H was also measured in boreholes, most of them directly drilled into the test cavities. Therefore, ^3H data are used to establish a conservative model chain for the source term and migration to the lagoon or the ocean. As neither exact depths for the test were given, nor flow paths characteristics are available in any detail, assumptions have to be made to overcome this lack of data. Pre-test hydraulic modelling and sensitivity analysis on the impact of individual tests (categories) (Fairhurst *et al.*, 1998) were used to deduce a conservative approach, applicable to all tests. This included the simplification with respect to: (a) three-dimensional (3-D) dispersion, (b) the time distribution (between 1975 and 1996) when the tests were performed, and (c) the transient upward velocity decrease after the tests. Whereas in the IAEA report (1998), the effects of (a) and (b) were investigated in the framework of a box model and good agreement with measured data was reached, here we estimate their impact in the framework of an advection–dispersion transport model in the carbonates and we quantitatively demonstrate their effects for a few dominant nuclear test for ^3H . For the source term, an analytical expression is deduced describing the radionuclide concentration within the cavity chimney and its temporal development. Transport is described using two different approaches: a single porosity and a double porosity model. The double porosity model seemed more appropriate with respect to the volcanic rock structure and the fractures that were obviously generated in the vicinity of the explosion. It delivers earlier release of non-sorbing radionuclides into the lagoons than was evident from observations. In contrast, for transport in the overlying carbonates, a single porosity model seemed to be more appropriate because of a much higher porosity.

TRITIUM SOURCE TERM ESTIMATION AND TRANSPORT MODELLING

Tritium is dispersed initially in the cavity water of the chimney created by the explosion. It has a single source term, different from other nuclides such as Pu, that are sorbed on the rubble and, in addition, partly incorporated within the lava which forms a meniscus at the bottom of the chimney when the molten rock cools down. For ^3H , we assume a homogeneous distribution within the whole chimney water brought about by convection cells during the initial thermal phase leading to a well-mixed compartment where fresh water enters from the chimney bottom and sides. A net outflow out from the chimney was calculated vertically upward into the carbonates (Fig. 1). The ^3H concentration in chimney water is given by:

$$C = \frac{A}{V\varepsilon} \quad (1)$$

where A is the total activity in the chimney, V the chimney volume and ε is the porosity of the chimney. Conservation of mass gives the change of concentration in the chimney:

$$\frac{dC}{dt} = -\lambda C - \frac{v_D}{h\varepsilon} C \quad (2)$$

where h is the chimney height, λ is the decay constant, and v_D is the time dependent Darcy velocity for water leaving the chimney. The first term on the right describes radioactive decay; the second describes the loss of activity in the chimney by advection. Hydraulic modelling showed that the velocity decreases exponentially from its maximum value shortly after the explosion to a background flow velocity v_{bg} . Depending on the test yield, location and related geohydraulic parameters, an analytical function could be fitted to the calculated transient water flow velocity at the top of the chimney:

$$v_D(t) = v_{bg} + (v_{bomb})e^{-bt} \tag{3}$$

where v_{bomb} is the velocity increase due to the explosion and b a constant describing the relaxation to pre-test flow conditions. The solution to equations (2) and (3) is:

$$C = C_0 e^{\frac{\lambda b h \epsilon t - v_{bg} b t + v_{bomb} (1 - e^{-bt})}{b h \epsilon}} \tag{4}$$

where C_0 is the initial concentration in the chimney water at $t = 0$. The three terms in the exponent are identified as pure radioactive decay, loss by background flow and dissipating flow due to the test. Obviously, the latter two are corrections to radioactive decay and depend on the rate of water flow through the chimney. Then, the total nuclide release rate j from the top of the chimney is calculated from equation (5). It indicates that the total release rate is equal to the product of the volume of water leaving the chimney per unit time and the radionuclide concentration in that volume (R_c , is the chimney radius):

$$j = \pi R_c^2 v_D C \tag{5}$$

Transport modelling requires detailed consideration of the geometrical structure of water carrying features and transport processes (Fig. 1). The volcanics contain many

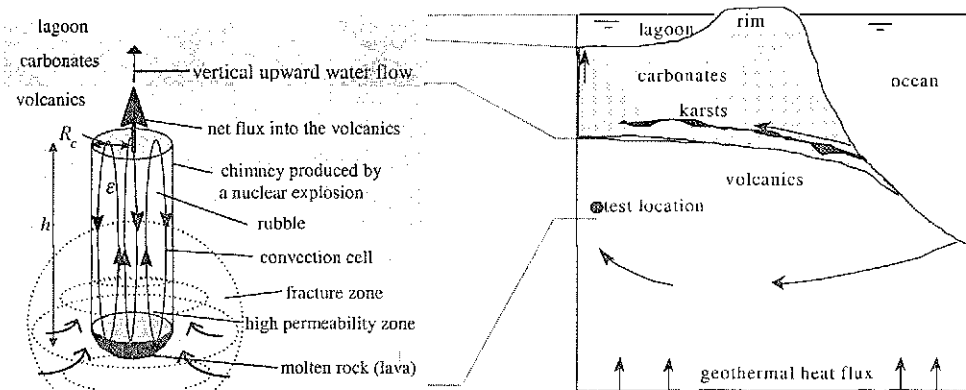


Fig. 1 Concept for the source term calculation for a nuclear test that creates a cavity and chimney containing rubble (crushed volcanic rock with higher porosity and hydraulic conductivity) at the top, and molten volcanic rock (lava) at its bottom. A zone of higher conductivity than the original rock hydraulic conductivity is created around the chimney. Due to differences in hydraulic conductivity, temperature and hydraulic heads, a convection cell develops in the chimney with a net outflow of nuclide contaminated water at its top and a net inflow in the lower part of the chimney (IAEA, 1998). The geometry of the chimney is a function of the test yield.

fractures of varying scales, and transport takes place in a fractured medium. Hence, for transport in the volcanics we use a model describing a double porosity medium (Jakob, 1997). It includes one-dimensional (1-D) advection–dispersion and surface sorption in the fractures, diffusion and sorption within the rock matrix and radioactive decay and build-up for constant and transient hydraulic conditions. Model parameters are longitudinal dispersion in the fracture, the fracture half-width and porosity, the matrix quantities porosity and depth, and nuclide specific parameters such as diffusion constant and decay constant as well as a transient source term. The water velocity within the fracture is given by $v = v_D \epsilon_f^{-1}$ where ϵ_f is the fracture porosity, and can be calculated by the expression $\epsilon_f = n 2b$ where n is the fracture frequency [m^{-1}]. $2b$ represents the hydraulic and transport aperture. Whereas advective transport is usually very fast (small fracture apertures) in such a double porosity system, matrix diffusion presents a powerful retardation mechanism. Provided that the residence time in the geosphere is long compared to the release time, and the rock matrix does not get saturated with the radionuclide in question, the retardation by matrix diffusion, \hat{R} , can be expressed as:

$$\hat{R} = 1 + \frac{2}{3} \tau_0 \left(\frac{L}{v} \right)^{-1} \tag{6}$$

L is the migration distance, and the time shift τ_0 (Hadermann & Heer, 1996) is given by:

$$\tau_0 = \left(\frac{\epsilon_p}{b} \right)^2 \left(\frac{L}{v} \right)^2 \frac{D_p R_p}{4} \tag{7}$$

where R_p is the retardation factor in the matrix that is 1 for ^3H .

Site-specific information on the important parameters is scarce, especially for those characterizing the flow paths. In this context, it is important to note that the solution of transport equations in (Jakob, 1997) is determined by four independent parameters, and it is more or less a matter of choice as to which of the physical parameters to fix, and which to vary. We fix $L/\alpha_L = 10$ (based on a wealth of field experiments, see e.g. de Marsily, 1986), where α_L is the longitudinal dispersivity. Further, the fracture frequency was fixed to $n = 10 \text{ m}^{-1}$ and fracture aperture to $2b = 10^{-3} \text{ m}$, assuming that all water flow is through the fractures. Relatively fast advective transport of radionuclides results. The matrix diffusion coefficient was fixed at $D_p = 10^{-11} \text{ m}^2 \text{ s}^{-1}$, a generic value from the literature. The variables were Darcy velocity and the depth of penetration for diffusion into the matrix (Table 1). The description of transport through the carbonates is seriously limited due to a lack of information on the geometric structure of the flow paths, especially of karsts, and an understanding of the influence of tides in the karst (IAEA, 1998). There are, thus, several possible approaches that could be taken to estimate the flux of radionuclides into the lagoon that are discussed elsewhere (IAEA, 1998). We present here a 1-D porous medium approach for transport in the carbonates to reflect the major flow direction, upwards into the lagoon, and the relative high porosity of 0.3. Then, the total ^3H release was calculated for Mururoa and Fangataufa using yield clusters of 5, 25, 60 and 100 kt tests conducted at nominal depths of volcanic cover ranging between 25 and 250 m because exact depths of individual tests were not given.

Table 1 Calculations performed for ^3H : test categories, related parameters for the base case calculations and their variations in parentheses.

	Test yield of 5, 25, 60 and 100 kt, in total 121 (Cat. 1)		Enee (50 kt), Lycos (87 kt), Megaree (54 kt), Nestor (47 kt) (Cat. 2)	12 CRTV trials, 5 kt each (Cat. 3)
Matrix depth (m)	v_D (m year $^{-1}$) chimney, volcanics	v_D (m year $^{-1}$) carbonates	v_D (m year $^{-1}$) carbonates	v_D (m year $^{-1}$) chimney, carbonates
0.0495 (0.1)	1 (0.1, 10)	1 (2)	1 (2, 20)	20 (2, $v_D(t)$)

Table 2 ^3H measurements in chimney water compared to source term calculations.

Measured in post-test boreholes at test site (Bq m $^{-3}$):				Calculated (Bq m $^{-3}$)
Lycos	Aristee	Boros	Ceto	
5×10^9	1×10^{10}	4×10^8	2×10^{10}	1×10^{10}

RESULTS AND DISCUSSION

Tritium concentrations were available for a few boreholes that were drilled after tests directly into the cavity. Comparing this data to initial concentrations calculated from estimated inventories (IAEA, 1998) together with the source term model (equation (4)) delivers ^3H concentrations that are in good agreement (Table 2). Parameters describing the transient phase (equation (3)) are hard to derive for individual tests, especially the velocity relaxation, therefore, we assumed a constant maximum flow velocity for different test categories. To quantify this approximation, we calculate the release and further transport through the carbonates for a 5 kt test for a dissipating water velocity compared to the related constant, maximum velocity (Fig. 2). The peak maximum is decreased and delayed, depending on the initial velocity value, for the more realistic dissipating velocity case, for which individual test data are hard to quantify. It should be mentioned that dissipation to background velocity will take up to several hundreds of years depending on the test yield.

With the clustering of tests, the exact dates of the original test will no longer be of relevance. Considering that, in the base case, conservative parameters have been

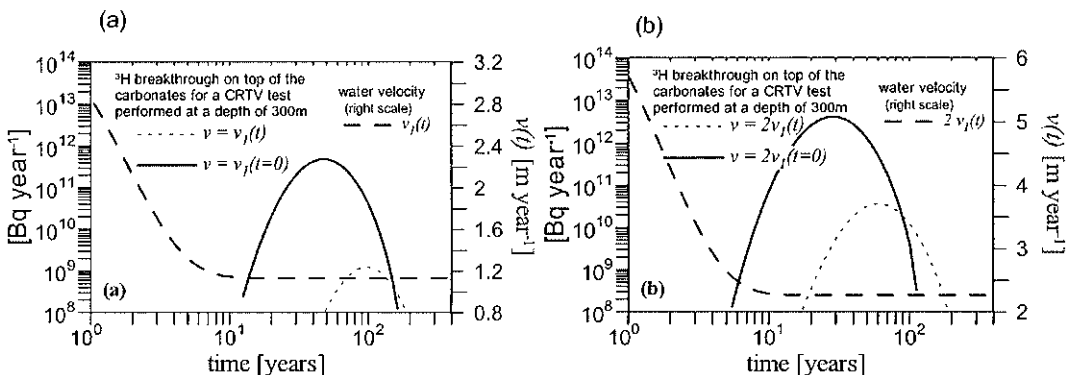


Fig. 2 ^3H breakthrough curves for differently dissipating vertical water flow velocities in the carbonates: (a) $v = v_1(t) = (0.34 + 0.9e^{-0.35t})/0.3$ and (b) $v = 2 v_1(t)$ compared to approximations of constant maximum velocity, $v = v(t = 0)$, for a 5 kt test.

chosen, this procedure leads to overestimation of peak releases. It is conservative for non-sorbing species with relatively short migration times, as for ^3H , because the temporal distribution of tests will smear out the releases to the lagoon (or ocean) and will lower the peak releases. The total release period will be slightly longer for the dominating tests of Cat. 2 and 3 (Fig. 3), taking into account the time distribution of the tests and the result when all test dates are taken as identical. Using the approximations of constant maximum water velocities and all tests performed at the same time, the total ^3H releases are calculated by summing data for 127 tests at Mururoa and 10 at Fangataufa. ^3H releases for the first decades are dominated by Cat. 2 and 3 tests for both Fangataufa (Lycos test, 1989), and Mururoa (12 CRTV and 3 leaky tests). For Fangataufa and Mururoa, the release begins to decrease some years after the end of dominant tests, which is consistent with the experimental measurements of the concentrations in the lagoon (IAEA, 1998). However, the absolute rate is overestimated by one to two orders of magnitude. Lower release rates would be obtained by dissipating or using lower Darcy velocities, which would reduce the flux into the carbonates. However, sensitivity analysis shows that low velocities in the carbonates ($v_D = 1 \text{ m year}^{-1}$) cannot reproduce the present day fluxes into the lagoon: the peak would be both too late and too small. In principle, it would be possible to obtain a better match to the observed flux, resulting in less of an overestimate, by slightly varying the Darcy velocity in the carbonates. This was not done because the model and the parameters are so uncertain and the present results err on the side of conservatism. Overestimation of the fluxes to the lagoon or the ocean is important in the context of a consequence analysis, since this quantity enters the dose calculations. Although direct evidence against which to test the validity of the modelling assumptions and the predictions made is limited, several encouraging confirmations have been established:

- Measured ^3H concentrations in the chimneys are consistent with those calculated by the source term model.

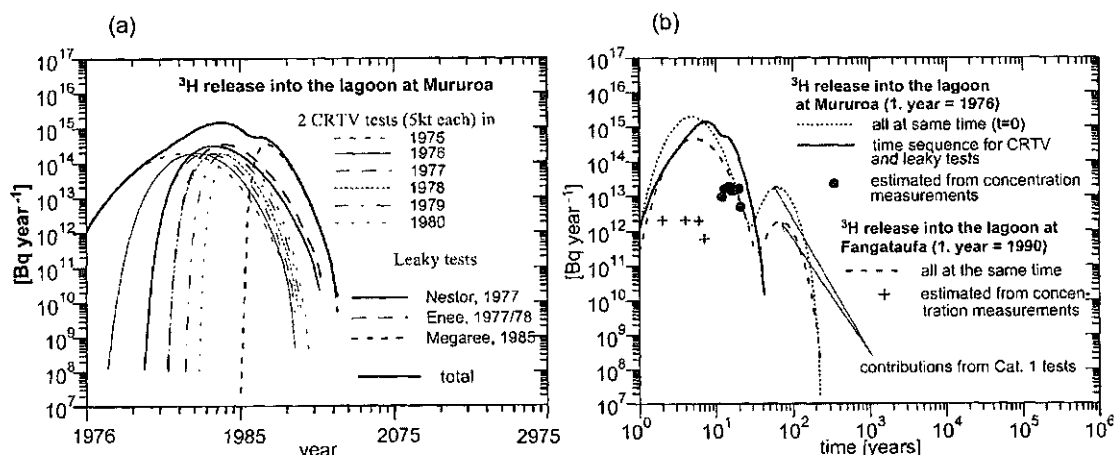


Fig. 3 ^3H breakthrough curves for (a) the time distribution of individual Cat. 2 and 3 tests, and (b) total releases for Mururoa, compared to the conservative estimate "all tests at the same time", and Fangataufa.

- Approximations made for the thermal driven, vertical velocity dissipation and to calculate the total ^3H release as if all tests had been performed at the same time are conservative with respect to peak concentration levels.
- Predicted ^3H inventories in the carbonates and releases to the lagoons are essentially consistent with the concentrations measured.

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