Reports

Tritium Distribution in Ground Water around Large Underground Fusion Explosions

Abstract. *Tritium will be released in significant amounts from large underground nuclear fusion explosions in the Plowshare Program. The tritium could become highly concentrated in nearby ground waters, and could be of equal or more importance as a possible contaminant than other long-lived fission-product and induced radionuclides. Behavior of tritiated water in particular hydrologic and geologic environments, as illustrated by hypothetical explosions in dolomite and tuff, must be carefully evaluated to predict under what conditions high ground-water concentrations of tritium might occur.*

Tritium from nuclear explosions is not considered as dangerous in the biological sense as many other radionuclides, since it emits only weak beta-radiation and has a half-life of 12 years. For example, the recommended maximum permissible concentration (MPC) for tritium in drinking water for occupational workers continuously exposed (168-hour week) is 3×10^{-3} curies H^3/cm^3 in contrast to 1×10^{-12} curies Sr^{90}/cm^3, or a factor of 10^4 higher MPC for tritium (*1*). However, in the Plowshare Program—the peaceful industrial uses of nuclear explosives, where large underground fusion explosives may be used, tritium may have to be reckoned with as a major long-lived contaminating radionuclide in ground and surface waters.

Basic physical data on reaction products from pure fission and pure fusion explosions have been summarized by Leipunsky (*2*), as shown in Table 1. To recast these data in terms that will illustrate the reaction products formed in a hypothetical underground explosion in the Plowshare Program, we have made three assumptions: (i) that the explosion is a 1-megaton fusion device triggered by a 10-kiloton fission device, (ii) that the explosion will be contained underground in the sense that the fireball and direct neutron flux will not reach the atmosphere, although some reaction products will be vented to the atmosphere by the explosion forming a throw-out crater, and (iii) that the explosive device will be surrounded by borated materials which capture neutrons without producing radioactivity, thereby reducing the neutron flux and the resultant induced activities by one order of magnitude. The environment in which the explosion occurs is assumed to consist of average crustal materials with a porosity of 20 percent and saturated with water. Calculations based on these assumptions are shown in Table 2.

The fission and fusion reaction products postulated in Table 2 for an underground explosion are, proportionately, the same as quoted in Table 1 for atmospheric explosions. However, the induced activities for an underground explosion (*3*) differ considerably from those for an atmospheric explosion, reflecting the chemical composition of the medium around the explosion—that is, rock as opposed to air.

In an underground explosion, the amount of C^{14} induced by neutron reaction with N^{14} would be sharply decreased in comparison with that produced in an atmospheric explosion; any possible radiation hazard to man (*4*) would be correspondingly reduced. This decrease in C^{14} directly reflects the availability of N^{14}, which would be at least two orders of magnitude less abundant in soil and rocks than in air. Furthermore, interaction with crustal materials, particularly water, would reduce the neutron flux available for the $N^{14}(n, p)C^{14}$ reaction by at least one order of magnitude, and neutron shielding of the explosive device would reduce the flux by an additional order of magnitude. Thus, for a nominal 1-megaton fusion explosion underground, the production of C^{14} would be reduced by at least four orders of magnitude to about 15 curies.

Several conclusions may be drawn from the data in Table 2.

1) Activities induced in average crustal material by the neutron flux from the explosion are relatively short-lived, consisting primarily of Al^{28} (2.3-minute half-life), Mn^{56} (2.6-hour half-life), Na^{24} (15-hour half-life), Fe^{59} (45-day half-life), and Co^{60} (5.2-year half-life). These activities decrease very rapidly: four orders of magnitude in the first week. At the end of one year, the induced activities, except for roughly 10^4 curies of Co^{60}, are insignificant compared with the longer-lived Sr^{90} (28-year half-life), Cs^{137} (30-year half-life), and H^3 (12-year half-life).

2) Although the activation product C^{14} is long-lived (5600-year half-life), the total amount produced—15 curies from the fusion neutron flux and a negligible amount from the fission flux, can be considered insignificant when distributed in the large volume of material around an underground explosion.

3) Fission-product activities do not decrease as rapidly as the induced activities, but drop by three orders of magnitude in the first week and by five orders of magnitude in the first year. At the end of one year, Sr^{90} and Cs^{137}, both with about 30-year half-lives, are the principal remaining radionuclides of recognized biological importance. It is assumed that Sr^{90} is the more significant radionuclide. The MPC of Cs^{137} is two orders of magnitude higher than Sr^{90}; Cs^{137} tends to be more firmly held in the solid by exchange mechanisms than is Sr^{90}, and is thus considered less important than Sr^{90}; it is not considered further in the following calculations.

4) Tritium at 6.7×10^6 curies is the predominant long-lived nuclide (12-year half-life) from the fusion reaction, and Sr^{90} at 1.5×10^3 curies is the predominant long-lived radionuclide (28-year half-life) from fission. At the end of one year, the amounts

of H^3 and Sr^{90} are relatively unchanged, whereas the other radionuclides either have been sharply reduced by decay or are present in less than significant amounts.

The distribution of H^3, Sr^{90}, and Co^{60} in a dolomite or similar carbonate rock can now be calculated on the basis of four assumptions, representative of the effects that may be expected (*5*) around a nonventing underground explosion, consisting of 1 megaton of fusion energy release and 10 kilotons of fission release: (i) that the radius of the cavity is 350 ft (approximately 110 m), (ii) that the radius of the outer limit of crushed zone surrounding the cavity is 740 ft (230 m), (iii) that the grain density of dolomite is 2.8, and its porosity (water saturated) is 0.05 (5 percent), and (iv) that radionuclides are distributed only in the crushed zone by direct explosive action, and that post-explosion collapse of the crushed zone into the cavity will not affect the nuclide distribution.

On these assumptions, the mass of solids in the crushed zone will be 1.14×10^{14} grams, the mass of water in the pore space will be 2.1×10^{12}g (or ml), and the total mass will be 1.16×10^{14}g.

Table 1. Reaction products of 1 megaton-equivalent (10^{15} calories) atmospheric explosions [Modified from Table 1, Leipunsky (*2*)].

Pure fission explosion	Pure fusion explosion
Number of fissions	
1.45×10^{26}	
*Number of fusion reactions**	
	1.45×10^{27}
Number of neutrons released	
2.2×10^{26}	1.45×10^{27}
Amount of Sr^{90} (fission product)	
1.47×10^5 c	
Amount of Cs^{137} (fission product)	
1.57×10^5 c	
Amount of C^{14} (activation product) †	
2.34×10^4 c	1.49×10^5 c
Amount of H^3 (residual product) ‡	
	6.67×10^6 c
Total amount radioactive products §	
3.27×10^5 c	6.82×10^6 c
Energy of radioactive products ||	
2.5×10^{25} Mev	7.5×10^{25} Mev

* At 180 Mev energy release for ten single H^2–H^3 reactions, equivalent to prompt energy release of one single fission. † For $N^{14}(n,p)C^{14}$ reaction, assuming for fission air burst that one-seventh as much C^{14} is formed per unit energy as for fusion air burst. ‡ Assuming 1 atom H^3 per 180 Mev energy released. § Only Sr^{90}, Cs^{137}, C^{14}, and H^3 included. || Decay energy of nuclides listed.

Assuming that Sr^{90} is all soluble and uniformly distributed throughout the dolomite in the crushed zone, its initial concentration in the total mass of the crushed zone would be 1500 curies Sr^{90} in 1.16×10^{14}g, or 1.29×10^{11}c/g.

When equilibrium is reached in the exchange of Sr^{90} between the dolomite matrix and the contained pore water, (*6*), the amount of Sr^{90} in the water is expressed by the equation,

$$K_d = \frac{\text{Activity-solid}}{\text{Activity-water}} \times \frac{\text{Volume-water}}{\text{Weight-solid}}$$

where K_d, the distribution coefficient for dolomite (*7*), is 10. The Sr^{90} activity in the water is then 2.8 curies; and the Sr^{90} concentration in the water, 2.8 curies in 2.1×10^{12}ml, is then 1.33×10^{-12}c/ml.

The MPC (168-hour week) for Sr^{90} in water is 1×10^{-12} c/ml, so that the initial Sr^{90} concentration in the contained pore water in this hypothetical example would be about the same as the MPC. For pure dolomite with less clay minerals and other impurities than the "average" dolomite selected, and with large amounts of Ca and Mg ions in the pore water, the K_d for Sr^{90} would be lower, possibly approaching a reduction of as much as two orders of magnitude; the Sr^{90} concentration in water would be correspondingly increased (*8*). Conversely, many dolomites and limestones are relatively impure and would have a higher K_d for Sr^{90}, leading to a lower Sr^{90} concentration.

The only long-lived activity of possible importance induced in average crustal materials is Co^{60} (5.2-year half-life) at 10^4 curies. The cobalt content of carbonate rocks (0.2 to 2.0 parts per million) is at least one order of magnitude less than the average (23 ppm) for the earth's crust (*9*). Thus, about 10^3 curies Co^{60} would be distributed throughout the crushed zone, and its initial concentration would be 9×10^{-12} c/g. Co^{60} is analogous to Sr^{90} in exchange behavior, and after reaching exchange equilibrium between the dolomite and the pore water, its concentration in water would be 9×10^{-13} c/ml.

The MPC (168-hour week) for Co^{60} in water is 5×10^{-10} c/ml, so the concentration is roughly three orders of magnitude less than the MPC.

In a nonventing underground explosion, it is assumed that essentially all tritium would rapidly form tritiated water, either by oxidation or exchange, and that tritium exchange between the tritiated water and the rock matrix would be negligible. The tritium concentration in the pore water, therefore, would be 6.7×10^6 curies H^3 in 2.14×10^{12} ml, or 3.1×10^{-6} c/ml.

The MPC (168-hour week) for H^3 in water is 3×10^{-8} c/ml, so the H^3 concentration in the water (about 1700 acre-feet, or 2×10^6 m^3) in the crushed zone is two orders of magnitude higher than the MPC. This tritium concentration in the water could be reduced only by dilution and mixing with ground water outside the crushed zone, not by exchange mechanisms.

Table 2. Reaction products for an underground explosion, consisting of 1 megaton fusion and 10 kiloton fission.

Source	Fission products (curies)	Induced* products (curies)	Fusion products (curies)
Fission (10 KT)	3.0×10^{9}†	10^5	——
Sr^{90}	1.5×10^3	——	——
Cs^{137}	1.6×10^3	——	——
C^{14}‡	——	Negligible	——
Fusion (1 MT)	——	10^8	——
H^3	——	——	6.7×10^6
C^{14}‡	——	15	——

* Induced gamma-ray activities at 1 hour after detonation (*3*), based on average chemical composition of earth's crust (*9*); induced activities at 1 week would have decayed by roughly 4 orders of magnitude, leaving essentially only Fe^{59} (45-day half-life) and Co^{60} (5.2-year half-life). † Fission-product gamma-ray activity at 1 hour after detonation, decaying at $T^{-1.2}$, or decreasing by 3 orders of magnitude in 1 week, and by 5 orders of magnitude in 1 year (*13*). ‡ Following Leipunsky (*2*) in Table 1, C^{14} from fission and fusion reactions shown separately.

Variations in the chemical composition of the dolomite would not affect the H^3 concentration in the pore water; variations in the water content of the rock would, of course, directly affect the H^3 concentration, as illustrated by

Table 3. Concentration of H^3 in ground water in multiples of MPC (168-hour week).

Type of explosion	Rock type	
	Dolomite	Tuff
Underground explosion	100	10
Cratering explosion at 90% loss of H^3	10	1
Cratering explosion at 99% loss of H^3	1	1/10

a hypothetical explosion in water-saturated tuff or alluvium.

The distribution of radionuclides in volcanic tuff and alluvium, such as at the Nevada Test Site, can be similarly calculated, where the cavity radius would be 520 ft (approximately 155 m), the crushed zone radius 1,100 ft (340 m), the density 2.5, the porosity (water saturated) 0.2 or 20 percent and the K_d for Sr^{90} and Co^{60} at 100; then the concentration of these nuclides in water in the crushed zone would be: Sr^{90} at 5.4×10^{-14} c/ml, Co^{60} at 3.6×10^{-13} c/ml, and H^3 at 2.4×10^{-7} c/ml. The Sr^{90} concentration is two orders of magnitude less than the MPC (168-hour week) of 1×10^{-12} c/ml. The Co^{60} concentration, based on 10^4 curies, since the Co content in the volcanic tuff and associated alluvium is about the same as in average crustal materials, is three orders of magnitude less than the MPC of 5×10^{-10} c/ml. The tritium concentration, which would not be appreciably decreased by any exchange reactions of tritiated water with the tuff medium, is still above the MPC of 3×10^{-8} c/ml by an order of magnitude, and is now contained in 23,000 acre-feet (or about 28×10^6 m^3) of water.

The distribution of radionuclides, where the underground explosion is at the depth of burst for maximum crater dimensions, can be calculated by assuming that, on the average, 10 percent of the total reaction products will escape to the atmosphere (*10*). In the early gas-phase venting associated with cratering, proportionately more of the radionuclides which behave as volatile substances, such as H^3, Kr^{85}, Sr^{89}, Sr^{90}, I^{131}, Cs^{137}, and Ba^{140} are vented than are the nuclides such as Zr^{95}, Ce^{144}, and the rare earth nuclides, which behave in the manner of refractory materials (*11*). Except for the long-lived H^3, Sr^{90}, and Cs^{137}, the "volatile" nuclides are short-lived and do not affect appraisal of long-term contamination of ground water, although these short-lived nuclides do contribute to atmospheric contamination and the related problems of random, localized radioactive fallout (*12*).

Both Sr^{90} and H^3 act as volatiles in the early gas-phase venting associated with cratering; Sr^{90} because of a gaseous nuclide precursor, and H^3 because it is combined in water vapor. It is known that Sr^{90} is enriched in the early stage of venting by a factor of 5 or less (*11*), which, compounded with a 10 percent release, would reduce the ground concentration by somewhat less than one order of magnitude. During the early stage of venting, H^3, although its behavior is not well documented, is probably more volatile than Sr^{90}, so that its ground concentration would be reduced by at least one order of magnitude. If 99 percent of the H^3 became combined in water vapor, a noncondensing gas when associated with the high temperature and pressure of an underground explosion, and were vented to the atmosphere (and this is a possibility) the H^3 concentration in ground water would be reduced by two orders of magnitude.

Under the assumptions made here, the concentration of Sr^{90} in ground water associated with a crater would be somewhat less than that in ground water associated with a nonventing underground explosion and would not be close to the MPC (168-hour week) in most rocks. As shown in Table 3, the concentration of H^3 in ground water associated with craters in dolomite and tuff would also be less than that in water near a nonventing underground explosion by at least one and perhaps two orders of magnitude. If the cratering explosion took place in tuff or alluvium, the H^3 concentration in ground water would be close to, or perhaps an order of magnitude below, the MPC, but in a dense rock its concentration would be equal to, or an order of magnitude above, the MPC (*14*).

Tritium around underground fusion explosions will be present mostly as tritiated water that does not enter appreciably into ion exchange reactions and, hence, will move with the ground water flow. As the tritiated water moves away from its initial explosion-produced distribution, the direction and velocity of ground-water movement and, more importantly, the dilution of the initial concentration of tritiated water can be broadly predicted from general knowledge, possibly to within several orders of magnitude. Dilution primarily by hydrodynamic dispersion might reduce the concentration to less than the MPC (168-hour week) at no great distance from the point of explosion; hydrodynamic dispersion, however, varies widely from little in a homogeneous medium to considerable in highly heterogeneous medium, requiring that each medium be carefully evaluated prior to release of large amounts of tritium.

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References and Notes

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