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Estimated Radiation Doses from Ingestion of Tritium-Containing Consumer Products Made with Hydrocarbons from Nuclearly Stimulated Natural Gas Wells

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OAK RIDGE NATIONAL LABORATORY
OPERATED BY UNION CARBIDE CORPORATION • FOR THE U.S. ATOMIC ENERGY COMMISSION

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ABSTRACT

Commercial-scale use of nuclear explosives to increase production of natural gas could result in the introduction of hydrocarbons containing tritium into petrochemical feedstocks. Most petrochemical products are not for human consumption, but this report considers radiation doses that could be received from ingestion of several products hypothetically produced from natural gas containing 1 pCi of tritium/cm^3 of gas. One of the highest estimated whole-body doses (0.9 millirem/year) was from ingestion of sufficient ethyl alcohol to maintain a concentration of 0.15%, the intoxication level, in the body water throughout the year. A slightly higher dose (1.0 millirem/year) was estimated for ingestion of a synthetic protein supplement. Estimated whole-body doses from ingestion of other products ranged from 0.5 millirem/year for an individual eating 1 lb/day of a hydrogenated fat or oil product to 0.005 millirem/year for a person taking eight aspirin tablets per day. We conclude that ingestion of products made from tritiated hydrocarbons resulting from use of nuclearly-stimulated natural gas could conceivably be a significant exposure pathway for some individuals, but it seems unlikely to become the most important pathway.

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INTRODUCTION

Previous reports[1-7] have dealt with potential radiation doses (hereafter referred to as doses) from hypothetical uses of natural gas resulting from application of nuclear explosives to increase gas production. The potential doses considered in these reports were primarily due to exposure to airborne HTO, ^{85}Kr, or $^{14}CO_2$ in gas combustion products, but potential doses from ingestion of several products that might be manufactured with tritiated hydrocarbons separated from nuclearly stimulated natural gas were briefly considered in one publication.[1] Some of the information reported there is included in this report for convenience. Further consideration of this subject is given in this report. Tritium is the only one of the three long-lived isotopes present in the nuclearly-stimulated natural gas that is considered likely to be incorporated in consumer products in sufficient amount to warrant investigation.

Tritium Concentration in Source Gas

It has been reported[8] that the Gasbuggy and Rulison wells were both created by weapons-type nuclear explosives. The Rio Blanco project[9] was the first to use nuclear explosives (3-30 kt) specifically designed to increase gas production from low-permeability rock formations. The initial tritium concentration in dry gas from the top Rio Blanco cavity was 28 pCi/cm^3, approximately one-sixth of the initial Rulison concentration.[10] From this it can be deduced that, if the total volume of gas flowing from the cavity during the lifetime of the well is 28 cavity volumes (approximately 1.5 x 10^9 ft^3), the average tritium concentration in the product gas would be 1 pCi/cm^3. The long-range production capability of Rio Blanco's separate cavities remains to be established, but it seems likely that more than 28 cavity volumes would be produced over a 20- to 30-year period. For this study we have adopted this figure (1 pCi/cm^3) for the total tritium concentration of the gas used to manufacture the different products considered. Subsequent developments may require a higher or lower value, but it should be easy to determine whether the change in assumed tritium concentrations will change the conclusions of this report.

Where the chemical composition of the gas is required in our calculations, the Rio Blanco pre-shot gas composition reported by Smith[10] is used. This is (in volume %): 94, CH_4; 3.5, C_2H_6; 0.7, C_3H_8; 0.4, C_4H_{10}; 0.15, C_5H_{12}; 0.2 $> C_5H_{12}$; others, 1.00. We assume that the tritium is distributed among the various hydrocarbons in proportion to their hydrogen content.

Sources and Uses of Petrochemical Hydrocarbons

Although it is somewhat outdated, Fig. 1 gives an approximate indication of the disposition of natural gas hydrocarbons. Among the changes that have occurred relative to this 1956 disposition of hydrocarbons are increased recovery of ethane, resulting from the increasing demand for ethylene in the petrochemical industry, and reduced production of carbon black from natural gas. Since refinery gases constitute an alternate source of hydrocarbons used in the petrochemical industry, it is of interest to examine the relative quantities of hydrocarbons from the two sources. Table 1 shows data

Table 1. Sources of Hydrocarbons Produced in the United States in 1972[11]

Hydrocarbon Product	Production (1000's of barrels)		Per Cent of Total from Natural Gas
	Natural Gas	Refineries	
Ethane	100,691	9,197	91.6
Liquefied gases	344,045	121,182	74.0
Propane	218,039	94,062	69.9
Butane	88,924	18,613	82.7
Mixed propane-butane	3,535	6,428	35.5
Isobutane	33,547	2,079	94.2

for the calendar year 1972, the latest information currently available.[11] It is obvious from the data in Table 1 that a large fraction of petrochemical feedstocks comes from natural gas. Table 2 shows data on production of major petrochemical products in the years 1967-1970.[12] The last column in the table gives the fraction of the product made from petrochemicals. Most of the components in Table 2 are intermediate products. Ethanol is the only compound listed that is commonly ingested by people, but other ingestible materials can be made from some of the intermediate compounds.

Dose Estimations

Ethanol[1]

Although most of the alcohol used in beverages is produced by fermentation, it seems likely that, if ethanol is manufactured from tritiated hydrocarbons, some of the products could be ingested by people.

As was stated earlier, we assume that the gas from the nuclearly stimulated well contains 1 pCi/cm^3 of tritium, its ethane content is 3.5 vol %, and that tritium is distributed among the hydrocarbons in proportion to their hydrogen content. This means that the ethane, with 1.5 times the hydrogen content of methane, would contain 5.1% of the total tritium or 0.051 pCi/cm^3 of the unfractionated gas. The specific activity of the separated ethane is calculated to be 1.444 pCi/cm^3 at STP or 1.08 x 10^3 pCi/g which corresponds to 650 pCi/g of C_2H_5OH or 510 pCi/cm^3 of liquid.

The definition of the alcoholic concentration in human blood at the intoxication level varies somewhat, but 0.15% seems to be a commonly used value. This is equivalent to 64.5 g for standard man[13] with 43 kg of

TABLE 2—Major Petrochemicals—Capacity and Production [12]
(All figures in millions of pounds)

			PRODUCTION				
	CAPACITY Estimated		Actual	Preliminary, Estimated			Percent Petro-chemical
	1969	1970	1967	1968	1969	1970	
Hydrocarbon Intermediates							
Aliphatic							
Ethylene	17000	18000	12200	13500	15000	16500	100
Propylene & Mixtures	7000	9000	5400	7000	8600	11000	100
Butylene & Mixtures	4300	4500	3650	4000	4400	4600	100
Butadiene	3200	3500	2670	2960	3300	3500	100
Acetylene	1700	1700	1430	1450	1400	1400	60
Aromatic							
Benzene	9200	9300	7100	7000	8200	9300	90
Cyclohexane	3200	3200	1780	2030	2500	2800	100
Cumene	2000	2500	1000	1300	1500	1700	95
Ethylbenzene	4800	5400	3600	4250	4700	5300	90
Naphthalene	900	900	880	860	800	800	42
Styrene	4900	5500	3280	3700	4000	4300	90
Toluene	5500	6500	4380	5070	5700	6400	97
Xylene	4400	4400	3480	3770	3800	3900	99
Polymers							
Polyethylene	5500	6000	3760	4540	5300	6000	100
Polypropylene	1000	1400	650	880	1150	1400	100
Polyvinyl Chloride & Copolymers	3400	3900	2080	2430	2900	3450	90
Styrene Polymers	3000	3500	2370	2720	3000	3350	90
Synthetic Rubber	4500	4800	3820	4270	4500	4700	90
Aromatic Chemicals							
Chlorobenzene	690	690	490	560	590	620	60
Nitrobenzene	400	430	360	380	400	410	60
Phenol	2000	2500	1240	1370	1500	1700	96
Phthalic Anhydride	950	1300	720	750	850	950	60
Aliphatic Chemicals							
Acetaldehyde	1800	1800	1400	1500	1600	1700	80
Acetic Acid	2000	2400	1500	1600	1700	1800	99
Acetic Anhydride	1800	1800	1560	1650	1700	1800	99
Acetone	1500	1500	1220	1270	1300	1350	85
Acrylonitrile	1100	1400	670	1020	1050	1100	100
Butanols	900	900	500**	510**	510**	510**	85
Carbon Disulfide	900	900	700	740	770	800	85
Carbon Tetrachloride	900	900	710	760	850	900	95
Ethanol	2400	2400	2200	2200	2200	2200	95
Ethyl Chloride	750	750	640	640	640	640	100
Ethylene Dichloride	8000	8500	4300	4800	5400	6000	100
Ethylene Glycol	2430	2700	2140	2200	2400	2600	95
Ethylene Oxide	3400	3500	2310	2610	2800	3000	99
Fluorocarbons	725	750	500	550	600	650	100
Formaldehyde (100%)	1850	1850	1370	1520	1650	1750	98
Glycerol	430	430	360	370	380	390	70
Isopropanol	2350	2350	1800	1900	2000	2100	100
Maleic Anhydride	230	270	170	180	210	230	30
Methanol	4900	5500	3430	3850	4300	4600	99
Methyl Chloride	450	450	260	310	340	360	90
Methylene Chloride	350	350	240	270	270	270	100
Oxo Alcohols	850	950	570	620	640	690	100
Perchloroethylene	750	750	530	630	700	750	80
Propylene Glycol	410	410	280	300	320	340	100
Propylene Oxide	1000	1200	810	960	1000	1100	100
Trichloroethylene	920	920	490	520	550	580	50
Urea	8000	8500	4360	4860	5250	5800	99
Vinyl Acetate	900	1600	610	710	790	850	75
Vinyl Chloride	4100	4300	2130	2640	2900	3200	90
Ammonia	30000	30000	24400	24200	23600	24000	99

** *n*-butyl & isobutyl only

body water or 82 cm^3 of ethanol. Daily ingestion of this quantity of alcohol would result in a daily tritium intake of 0.042 μCi. This would give an annual whole-body dose of 0.9 millirem. It is interesting to compare this estimated hypothetical tritium dose with that from drinking water. A recent report[14] gives a value of 0.3 nCi/liter (0.3 pCi/cm^3) for the average tritium concentration in all United States drinking water samples tested in the third quarter of 1973. If one assumes that all water intake (3.0 liters/day for standard man, including that in food) has the same tritium concentration as the drinking water, the resulting annual whole-body dose would be 0.02 millirem, 2% of that estimated from drinking tritiated ethanol. The maximum reported tritium concentration in drinking water[14] (4.8 nCi/liter), under the same assumptions as above, would give an annual whole-body dose of 0.3 millirem.

Ammonia Used as Fertilizer[1]

It was reported in 1969[15] that the cost of making a 1000 ft^3 of hydrogen from natural gas was $0.21 as compared to $4 by electrolysis. It seems safe to assume, therefore, that all synthetic NH_3 used in fertilizer comes from natural gas. It was also reported[15] that 8.75 x 10^{11} ft^3/year, representing 35% of the total hydrogen production, was used to make ammonia. Since two volumes of hydrogen are produced from one volume of methane, assuming 100% conversions, 4.4 x 10^{11} ft^3/year would be required to produce this amount of hydrogen. This is equivalent to 5.8 x 10^{11} ft^3/year of NH_3 or 2.8 x 10^{10} lb/year. The calculated tritium concentration in the NH_3 product, assuming 1 pCi/cm^3 of tritium in the natural gas, is 0.45 μCi/lb, and the total quantity of tritium is 1.3 x 10^4 Ci.

For the calculation of tritium absorption in fertilized food, we assume an ammonia application rate of 100 lb/acre-year, an annual rainfall of 40 in. (4000 m^3/acre), uniform dilution of tritium and grain production of 100 bushels per acre at 60 lb/bushel. We also assume that the grain contains 20% moisture in addition to the water required for synthesis of carbohydrates. The calculated concentration of tritium contributed to the water by application of the tritiated ammonia is 0.01 pCi/cm^3. The fraction of water molecules in carbohydrates is approximately 0.6, so the calculated total tritium concentration in the grain is 0.0068 pCi/g or 3 pCi/lb. Ingestion of 1 lb/day of grain would result in an annual whole-body dose of 0.0001 millirem. As noted above, the average tritium concentration in United States drinking water is 0.3 pCi/cm^3, 30 times the concentration calculated in water from use of tritiated fertilizer.

Hydrogenated Fats and Oils[1]

Another potential use of hydrogen made from natural gas containing tritium is for the hydrogenation of foodstuff. It was reported[16] that 10^9 ft^3 of H_2 was used in the hydrogenation of fats and oils in 1967. If we assume that the natural gas has a tritium concentration of 1 pCi/cm^3 and that oleic acid with a molecular weight of 282 is a representative unsaturated compound, we can calculate the tritium concentration in the hydrogenated product. Since two volumes of H_2 are produced from one volume of CH_4, assuming 100% conversion, the calculated tritium concentration in the hydrogen is 0.5 pCi/cm^3 or 0.56×10^4 pCi/g. Since 1-g mole of H_2 (2 g) is consumed in hydrogenating 1 mole (282 g) of oleic acid, 1 g H_2 = 141 g of oleic acid, and the calculated tritium concentration in the hydrogenated product is 0.018 µCi/lb. Ingestion of this product at the rate of 1 lb/day would result in a 1-year whole-body dose of 0.4 millirem.

Drugs

Aspirin, acetylsalicylic acid (ASA), $CH_3COOC_6H_4COOH$, is the most used drug on the market, with an estimated United States daily consumption in 1972 of 48 tons,[17] and was therefore chosen as the "worse case" of a drug which might be made from components of nuclearly stimulated natural gas.

Phenol used in the manufacture[18] of salicylic acid, and eventually of ASA, is derived from petroleum[19] and would be unlikely to contain tritium from natural gas. A review of the literature indicated that acetic anhydride may be made in part from natural gas,[19] though the actual pathways are ill defined and variable, depending on petrochemical supply, demand, and economics; and much of the information needed to define the most probable pathway is proprietary. Processes used include catalytic oxidation of natural gas, carbonylation of methanol, and oxidation of acetaldehyde. None of these processes could contribute more natural-gas hydrogen than the three atoms of the methyl group of ASA. Catalytic oxidation was chosen for calculation. The overall equations for oxidation of ethane and preparation of ASA are

$$2\ CH_3CH_3 + 3O_2 \rightarrow (CH_3CO)_2O + 3\ H_2O$$

$$(CH_3CO)_2O + HOC_6H_4COOH \rightarrow CH_3COOC_6H_4COOH + CH_3COOH$$

We assume as before that the tritium content of nuclearly stimulated gas is 1 pCi/cm^3. The specific activity of ethane, as was stated earlier in this paper, is assumed to be 1.5 that of methane, 1.44 pCi/cm^3 for gas containing 1.0 pCi/cm^3, corresponding to 5.4 x 10^3 pCi per gram of hydrogen. Some data have been reported[20] indicating a slightly higher value than that assumed here. As shown above, the acetyl hydrogens of ASA may come from ethane, and this leads to a level of 90 pCi per gram of ASA.

Few quantitative studies have been made of the metabolism of ASA in humans. However, it is well established that rapid hydrolysis occurs, with release of acetate and salicylate fractions.[21] The acetate, which contains the tritium, enters the body acetate pool and, as acetyl coenzyme A, is involved in the anabolic and catabolic steps of the citric acid cycle.[22] Turnover is rapid; over half the carbon of acetate appears as CO_2 in expired air in 24 hr after administration.[23] The tritium is assumed in our dose calculations to be oxidized and to enter the body water. The body water reaches equilibrium with the ASA input quickly because the half-time for elimination is short, about 10 days.[24] In general, organic compounds are excreted more rapidly than water,[25] and it has been shown[26] that a variation in elimination rate makes relatively little difference in chronic exposure.

With a daily dosage of eight ASA tablets (2.6 g), as for mild arthritis, the daily tritium intake would be 240 pCi. This is 67% of the standard man daily intake of 360 pCi in 1.2 liters of drinking water at the average United States concentration[14] of about 300 pCi/liter. The possible whole-body dose from ASA is calculated to be 0.005 millirem/year, less than 1% of the dose from natural ^{14}C in the body, 1.1 millirem/year.[27] The United States consumption rate of 48 tons/day of aspirin[17] is equivalent to approximately 0.64 of one five-grain tablet (0.21 g) for every man, woman, and child in the country. If all aspirin had the specific activity of tritium estimated here (90 pCi/g), which is a highly improbable circumstance, the resulting United States annual population dose from this source would be 90 man-rems. This compares with approximately 2 x 10^7 man-rems/year to the same population from natural background radiation.

Synthetic Proteins

Some 2500 materials are added to foods for flavoring, enhancement of nutritive value, improvement of texture, and for other purposes.[28,29] A number of these additives could be made from natural gas. Information was sought in publications and by contacts with specialists in the FDA and departments of home economics and toxicology concerning source materials used to make synthetic food additives. Most of the available information is qualitative, and much is speculative. United States Food and Drug Administration (FDA) publications, for example, make frequent use of such expressions as "the amount reasonably required" and "good manufacturing practice."

In general, the specific activities of tritium in products made from nuclearly stimulated gas are similar, so that potential human intake of tritium depends on the hydrogen content and the quantities likely to be consumed. The amounts of most additives consumed are small. A potentially significant product is supplemental protein made from methane.[30]

Single-cell organisms, such as bacteria and yeasts, have been grown with natural gas as nutrient, yielding approximately 1 lb of protein per pound of methane.[31] As before, the tritium content of the gas is assumed to be 1 pCi/cm^3, corresponding to 5.4×10^3 pCi per gram of hydrogen. Like a typical protein,[32] the petrochemical product contains about 7% hydrogen,[30] therefore 390 pCi/g. Assuming that a protein supplement might make up 20% of the 92 g/day intake, characteristic of United States adults,[33] the specific activity of the total protein intake would be 78 pCi/g. The tritium intake then would be 7.2×10^3 pCi/day, about 8 times the United States average currently received by adults from 3.0 liters/day of water, again assuming that all water, including that in food, contains 0.3 nCi/liter.

Turnover half-times of proteins in the body vary from less than a day[34] to 6 months or longer,[35] and rates of synthesis and degradation are equal in the adult.[32] Therefore, it is a conservative approximation that the 8.4 kg of body protein[36] will attain the same specific activity, 78 pCi/g, as the constant input. The body burden would then be 6.6×10^5 pCi, and the estimated annual whole-body dose is 1.0 millirem. This is slightly less than that from natural ^{14}C.

Modified Starch

Starch is modified to improve its thickening power and to make it resistant to extended storage, heat, and cold. Modification may be done, for example, by esterification with acetic anhydride, or etherification with propylene oxide. The latter is chosen for calculation because FDA regulations permit use of more propylene oxide than other modifying agents,[29] and the potential content of natural-gas hydrogen is higher in the propylene group than the acetyl.

Propane can be cracked to produce propylene, which is converted to propylene oxide, $CH_3CH(O)CH_2$, by the chlorhydrin process.[37] The six hydrogens in propylene oxide all come from the original propane. Propylene oxide is reacted with starch, yielding a product[38] with about 20% hydroxypropyl content (provided the proportion of propylene oxide is kept to the FDA maximum of 25%). Representing starch as ROH, the equation is[39]

$$ROH + CH_3CH(O)CH_2 \rightarrow ROCH_2CHOHCH_3$$

The product still contains six of the natural-gas hydrogens per hydroxypropyl unit.

Taking the activity of propane as 2 pCi/cm^3, the specific activity of hydrogen is 5.6×10^3 pCi/g. The tritium content of 20% hydroxypropyl starch would then be 120 pCi/g. The maximum consumption of modified starch reported[40] for an infant was 35 g/day, about one-sixteenth being the etherified type. Thus 2 g/day or 240 pCi would be a credible intake. This is comparable to the infant's average tritium intake in water, 270 pCi in 0.9 liter.[41]

Metabolism of the modified starch would yield propylene glycol, which is quickly converted to pyruvic acid.[42] This is part of the citric acid cycle.[22] The hydrogen is oxidized and enters the body water. Equilibrium with the intake of 240 pCi per day would lead to a dose rate of 0.016 millirem/year for an 8-kg infant.[41] Comparable dose rates might result from ingestion by adults of larger amounts of modified starch in other products.

Sorbitol

Sorbitol, $HOCH_2(CHOH)_4CH_2OH$, is used as a sweetener in "dietetic" foods and beverages.[29] It is manufactured by catalytic hydrogenation of dextrose,[43] according to the reaction

$$C_6H_{12}O_6 + H_2 \rightarrow C_6H_{14}O_6$$

Hydrogen can be prepared by steam re-forming of natural gas.[44]

$$CH_4 + H_2O \rightarrow CO + 3\ H_2$$

Thus two-thirds of the hydrogen could contain tritium, and its specific activity would be 3.7 x 10^3 pCi/g. There are 2 g of such hydrogen per formula weight of sorbitol (182), and the corresponding activity would be 41 pCi/g of sorbitol. The FDA permits the presence of up to 7% of this compound in foods for special dietary use. Thus, if one consumed 100 g per day of a product sweetened with sorbitol, the tritium intake would be 290 pCi/day. Sorbitol is oxidized in the body and its hydrogen is added to body water. The annual whole-body dose would be 0.007 millirem.

Regulations governing food additives[45] are much more stringent than those for drugs. The Food Additives Amendment of 1958 ("Delaney Clause") to the Pure Food Law forbids the addition of radioactivity unless permission has been granted. A procedure is prescribed for filing a petition to FDA setting forth details of intended use and safety investigations. However, the amendment also forbids use of a substance which has been shown to produce cancer in man or animal, at any level of administration.

Tritiated compounds, such as water[46] and thymidine,[47] have been reported to cause tumors in rats and mice. No reports were found of carcinogenic reactions to tritiated proteins, starches, etc. However, it is reasonable to believe that such reactions would be observed if tests were made at sufficiently high tritium levels. It is obvious that the law must be changed, perhaps in such a way as to permit reasonable radiation doses--well below natural background--as in the case of effluents from nuclear power plants. Criteria such as maximum permissible annual dose, or concentrations in air and water, have been discussed in connection with an effort to develop MPC's for food.[48] If the law as presently worded were enforced, drinking water could not be added to food because of its measurable tritium content. One must be sure that the benefit to the user, for example by supplementing a protein-poor diet, overbalances the possible slight risk. Dose estimations for all the consumer products considered in this report are summarized in Table 3.

CONCLUSION

We conclude from the results presented in this report that ingestion of products made from tritiated hydrocarbons resulting from use of nuclearly stimulated natural gas could conceivably be an exposure pathway that could result in doses to some individuals comparable in magnitude to those received from direct exposure to combustion products

Table 3. Summary of Estimated Doses from Ingestion of Consumer Products Manufactured with Natural Gas Containing 1 pCi/cm^3 of Tritium

Product	Daily Consumption of Product (g)	Tritium Concentration in Product (pCi/g)	Daily Tritium Intake (pCi)	Estimated Annual Dose (millirems)
Ethanol	64.5	1.08×10^3	4.2×10^4	0.9
Grain-NH_3 Fertilizer	454	0.0068	3	0.0001
Hydrogenated Oleic Acid	454	40	1.8×10^4	0.4
Aspirin	2.6	90	240	0.005
Synthetic Protein	18	390	7.2×10^3	1.0
Modified Starch	2	120	240	0.016
Sorbitol	7	41	290	0.007

of the same gas. It seems unlikely that the consumer exposure pathway will become significantly more important than the direct exposure pathway.

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