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RADIOACTIVE CONTAMINATION OF THE ATMOSPHERE BY SUPER BOMBS

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RADIOACTIVE CONTAMINATION OF THE ATMOSPHERE BY SUPER BOMBS

Ernest C. Anderson

INTRODUCTION AND CONCLUSIONS

A super bomb is expected to release many more neutrons than an ordinary bomb. The ratio of the neutrons released may be 10,000 or more. Capture of these neutrons in various substances will give rise to radioactivities and these activities will contaminate the atmosphere and the surface of the earth. What these contaminations are going to be can be influenced to a considerable extent by selecting the materials that go into the construction of the super bomb and by selecting the surrounding in which the super bomb is detonated. In most contemplated detonations a considerable fraction of the neutrons will be slowed down and eventually captured in the nitrogen of the atmosphere. Such capture gives rise primarily to the long lived carbon 14 isotope. It is probably possible to reduce other important radioactive contaminations caused by the super bomb. In most applications, however, creation of carbon 14 is unavoidable.

It will be shown in the following that the activity of the carbon 14 will remain below the tolerance as long as the number of supers detonated remains smaller than 1200, assuming the carbon 14 activity to be present in the chemical species CO_2. It will be shown that while other chemical forms are possible, it is unlikely that they will be more dangerous than CO_2. Generation of nuclear species in the atmosphere, other than carbon 14, will be shown not to be a limiting factor.

Additional contamination will certainly arise due to the fact that all Uranium 235 (or Plutonium 249) found in the neighborhood of the super is likely

to suffer fission due to the great abundance of neutrons released by the super. Some designs of the super which are now contemplated will produce approximately 40 times more fission fragments than a normal atomic bomb. If this is the case, then the number of supers which can be tolerated throughout the surface of the earth will be 40 times less than the number of ordinary bombs due to contamination by fission products. This latter figure may put a sharper limitation on the number of supers which one can detonate than the limitation due to carbon 14.

EVALUATION OF C14 ACTIVITY AS CO_2

We shall arbitrarily define a super bomb as one which releases a thousand times more energy than the more effective stock pile bombs. Thus, we shall assume that the energy released is 4×10^7 tons of TNT or 1.6×10^{24} ergs. The net reaction in the super is $5D \longrightarrow He_4 + He_3 + H_1 + 2n + 25$ MeV. Thus, we see that one neutron is released for every 12-1/2 million volts or for every 2×10^{-5} ergs. The super therefore releases a total of 8×10^{28} neutrons. It is assumed that all these neutrons will give rise to the $N_{14} + n \longrightarrow C_{14} + p$. reaction. Thus we produce 1.33×10^5 gram atoms of Carbon 14.

It is interesting to compare this amount of Carbon 14 with the estimated total amount which is maintained by the cosmic radiation. This amount has been computed (1) and it is estimated that two Carbon 14 atoms are produced by the

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(1) E. C. Anderson, Dissertation, University of Chicago, Department of Chemistry, June 1949.

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cosmic radiation per square centimeter and per second. Using for the half life of Carbon 14 5,720 years, one obtains 5.26×10^{11} Carbon 14 atoms in equilibrium

per square centimeter of the earth's surface. The total number of "natural Carbon 14" atoms throughout the earth's surface is therefore 2.68×10^{30}. We see, therefore, that approximately 33.5 supers will produce as much Carbon 14 as corresponds to the natural activity present on the earth.

The figure of 33.5 supers is, of course, in no direct relation to the number of supers which will produce a tolerance dose of Carbon 14 because

1. The quantity of "natural" radioactive carbon is greatly below tolerance and
2. The radioactive carbon produced by cosmic radiation is apparently distributed in a uniform manner throughout the whole carbon inventory with which atmospheric carbon dioxide can exchange in times short compared to geological periods. On the other hand, the carbon 14 produced by super bombs will remain concentrated for a considerable length of time in a relatively small fraction of this carbon inventory. It is worth while to give in this connection the total carbon inventory[1].

Carbon Inventory

Source	Amount (g/cm^2)
Ocean "carbonate"	7.25
Ocean, dissolved organic	0.59
Biosphere	0.35
Atmosphere	0.12
TOTAL	8.2

To obtain the total inventory, multiply by 5.1×10^{18} the area of the earth in cm^2. This gives us a predicted specific activity of natural radiocarbon which is within 10% of the observed value if we use Yuan and Ladenburg's data

for the cosmic ray neutron intensity[2].

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(2) Yuan, L., Phys. Rev. 74, 504 (1948)

Yuan, L., Bull. Am. Phys. Soc. 24, 9 (1949)

In order to obtain the total number of supers which will produce a tolerance dose of carbon 14, we shall assume

1. That the carbon 14 is rapidly distributed in the atmosphere but is not further diluted by distribution in the ocean or in the biosphere. This assumption may underestimate local carbon 14 concentrations in the atmosphere for relatively short periods of time. On the other hand, within a period of a year or two some of this carbon will surely be distributed in the biosphere and in the ocean and therefore the amount of carbon 14 in the atmosphere is on the whole likely to be less than the amount to which our assumption will lead us.
2. We assume that the concentration of carbon 14 in the human body is the same as it is in the atmosphere. This assumption of course is in contradiction with the previous assumption that the carbon 14 is distributed in the atmosphere alone. It is clear however, that these contradictory assumptions will lead to an over-estimate of carbon 14 in the human body and therefore we shall err on the side of caution.
3. Since carbon 14 is an emitter of soft beta radiation, it is likely to exercise a physiological effect only when it is incorporated in the body.

We assume that the tolerance dose is reached when the carbon 14 in the body releases .3 rep per gram of body per week. This dose has been established as the tolerance dose for both clinical workers and for people working on atomic energy projects.

One rep corresponds to 1.7×10^{12} ion pairs per gram of tissue. A dose of .3 rep per week corresponds to 8.4×10^{5} ion pairs per gram and second. A single beta disintegration of the carbon 14 (maximum energy equals 155 KeV) gives rise to 2000 ion pairs. Thus, we can tolerate 4.2×10^{2} disintegrations per gram of tissue per second. The number of carbon 14 atoms per gram of tissue which will produce such activity is 1.1×10^{14}. The average amount of carbon contained in tissue is 18%. The tissue most rich in carbon is fat, which contains 70%. We shall demand not even fat (the tissue richest in carbon and hence subject to the greatest carbon 14 concentration) shall be exposed to more than the clinical tolerance of 0.3 rep per week. One thus finds that no tissue may contains more than 3.5×10^{22} normal carbon atoms per cubic centimeter. The tolerance dose will not be exceeded if the ratio of carbon 14 atoms to normal carbon atoms remains smaller than 3.1×10^{-9}. We can, therefore, tolerate in the atmosphere 1.02×10^{32} carbon 14 atoms. Since a super produces 8×10^{28} carbon 14 atoms, a total of 1200 super bombs can be tolerated.

OTHER CHEMICAL FORMS OF CARBON 14 ACTIVITY

While natural carbon 14 is apparently confined to CO_2 and carbonates in the inorganic world and enters the biosphere only by photosynthetic and metabolic processes, there is a profound difference between "natural" carbon 14 and the material produced by a "super": namely the average natural carbon 14 atom is some 8000 years old and has had ample time to experience all possible chemical reactions with its environment. In the case of the newly formed "super" carbon 14, however, it is possible that an appreciable amount of the activity might be found in less stable chemical species with other and more effective paths of entering living matter. We shall consider the other chemical species which are thermodynamically capable of formation from the carbon 14 atoms reacting with the atmosphere. (Since the carbon 14 is present originally as iso-

lated atoms, we must consider the free energy of formation of the various chemical species from gaseous carbon. In effect, therefore, the negative free energy of sublimation of carbon is added to the free energy of formation and some thermodynamically unstable species can be formed. It should be noted that the original carbon 14 is produced with a considerable kinetic energy so that it can overcome any chemical potential barriers. Also, because of the "tracer" concentration of the carbon 14, most of the reactions must be considered as resulting in the formation of free radicals (such as O or N) whose later reactions do not affect the primary reaction.) Experiments are planned at this laboratory to determine the actual chemical species in which activity is found after the irradiation of air with neutrons of the appropriate energies.

Carbon Monoxide:

1. $C(g) + O_2(g) \longrightarrow CO(g) + O(g)$ $\quad \Delta F^\circ = -165{,}415$ cal/mole (298°K.)
2. $CO(g) + O_2(g) \longrightarrow CO_2(g) + O(g)$ $\quad -6{,}415$
3. $C(g) + O_2(g) \longrightarrow CO_2(g)$ $\quad -281{,}880$
4. $CO_2(g) \longrightarrow CO(g) + O(g)$ $\quad +116{,}465$

We note that:

a. CO will very probably form according to equation 1.

b. It is unlikely that any CO_2 will form directly by equation 3. (unless a third body is present in the collision) since the CO_2 would be left excited by an amount more than enough to cause dissociation according to equation 4.

c. The CO can eventually be oxidised to CO_2 according to equation 2.

We may therefore expect to find a certain fraction of the carbon 14 activity present as CO immediately after the detonation. Has this carbon monoxide any specific entry into the animal body and will this result in a further limitation in the possible number of supers?

The specific entry of CO is of course into the blood stream through the formation of carboxyhemoglobin. We shall show that the absorption of CO by this mechanism is completely negligible.

It has been shown[3] that the ratio of carboxyhemoglobin (HbCO) to

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(3) Wintrobe, M.M., Clinical Hematology, 2nd Ed., P. 112

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oxyhemoglobin (HbO_2) in the blood is given by

$$\frac{HbCO}{HbO_2} = 210 \frac{(CO)}{(O_2)}$$

where (CO) and (O_2) are the respective concentrations of these species in the inspired air. In other words, CO is competing with, and hence diluted by, the O_2 of the atmosphere. Since the O_2 is present at 700 times the concentration of CO_2 in the air, the resultant effective "specific activity" would be less by a factor of 3.3 for the CO than for CO_2. In the case of CO one carbon 14 enters a molecule of molecular weight 16,000 and, if one wishes to consider radiation damage to blood cells, another dilution factor of 3.4×10^{-4} (the concentration of hemoglobin in blood cells) enters.

If all the carbon 14 produced by one super forms CO, is diluted in the atmosphere and comes to equilibrium with hemoglobin one finds:

$$2.7 \times 10^{7} \ \frac{\text{atoms of carbon 14}}{\text{grams of hemoglobin}}$$

$$9.2 \times 10^{3} \ \frac{\text{atoms of carbon 14}}{\text{grams of blood cells}}$$

This compares with the tolerance dose:

$$\frac{1.1 \times 10^{14} \text{ atoms of carbon 14}}{\text{grams of tissue}}$$

Thus the assumption of carbon monoxide formation would permit the detonation of a much larger number of supers without poisoning the atmosphere. In

the long run the CO will be converted to CO_2 and the calculations of the previous section will apply.

CN and Related Species:

5. $C(s) + N_2(g) \longrightarrow CN(g) + N(g)$ $\Delta F^\circ = -9{,}790$
6. $CN(g) + O_2(g) \longrightarrow CO_2(g) + N(g)$ $-110{,}090$
7.a $CN(g) + H_2O(g) \longrightarrow HCNO(g) + H(g)$ $-22{,}634$
b $HCNO(g) + H_2O(g) \longrightarrow CO_2(g) + NH_3(g)$ $-1[illegible]74$
8. $CN(g) + H_2O(g) \longrightarrow HCN(g) + OH(g)$ $-2{,}260$

From these equations we may conclude:

a. CN can be formed according to equation 5. (Its polymerization to the usual C_2N_2 is very improbable because of the extremely low concentration.)

b. The oxidation and hydrolysis will undoubtedly convert CN to other species as indicated in equations 6, 7 and 8. Equations 6 and 7 return the activity to carbon dioxide, so that only equation 8 gives us a new species: HCN. It is clear that only a fraction of the total activity will reach this species.

Unfortunately no quantitative data are available on the metabolism of HCN. It is certain, however, that most of the cyanide is converted to thiocyanate, which is distributed and excreted similarly to the halogens[4] so that no accumulation in the body is to be expected. It is unlikely that the presence of CN will lead to a sharper limitation on the permissible number of supers than carbon dioxide. This is only a probability and not a certainty. Chemical and biological experiments are planned to determine the exact fate of the carbon 14.

(4) Goodman and Gilman, The Pharmacological Basis of Therapeutics, P. 699

On the whole, these statements on the fate of carbon 14 have a greater degree of reliability than the current discussions on the biological effects of fission products. This is due, in part, to the conservative assumptions that have been made and to an even greater extent it is due to the fact that the behaviour of carbon, which is one of the most common elements in the biosphere, is much better known and understood than the biological behaviour of the numerous fission products. For the latter, the possibility of concentration chains exists and these may give rise to unexpected poisoning effects.

OTHER RADIOACTIVE PRODUCTS

Although the great majority of the neutrons from a super would result in the production of carbon 14, appreciable amounts of other radioactive species may also form. Fast neutrons on N-14 can give rise to H-3 (tritium) and this species might also arise from the super reaction itself. On the basis of the cross-section measured by Cornog and Libby[5] tritium production from nitrogen 14

(5) Cornog and Libby, Phys. Rev. 72, 1046 (1941)

is not likely to be more than 1% of the carbon 14 production, while production from the super can be no more than 1% of the amount of unreacted deuterium. The energy of the tritium beta is only 1/10 that of the carbon 14 beta, but the half life is 500 times less giving a corresponding increase in the specific activity. The maximum effect from tritium is therefore nearly the same as that from carbon 14, assuming the same dilution. However, the concentration of water vapor in the atmosphere is at least 3 times the amount of CO_2 and is probably much more. Also rapid dilution by the water of rivers and oceans is to be expected. Therefore, the formation of tritium is not likely to cause a significant limitation in the number of supers.

A survey has been made of other activities likely to be produced by a burst of neutrons in the atmosphere. Table I summarizes the results of these calculations. Column 4 gives the number of atoms of a given species produced relative to carbon 14 production, Column 5 gives the initial activity of the species relative to the initial carbon 14 activity, and Column 6 gives the relative activity after two days. It will be noted that although the initial activity of some species is very high, this is due entirely to their very short half lives and that after two days all these activities will be reduced to a small fraction of the carbon 14 activity. Furthermore, the long lived activities are all due to rare gas isotopes which cannot be concentrated in the body.

The fast neutrons from the super may give rise to activities which as yet have not been discovered. Such activities are likely to have short lives. A search for such additional activities would seem to be indicated.

There remain to be considered only the activities produced from the materials of bomb construction. Any fissionable material will undergo essentially 100% fission because of the tremendous burst of neutrons from a super. The quantity of fission products can therefore be expected to be about 40 times that from an ordinary fission bomb according to present estimates of design, which assume 80 kg of fissionable material in the fission bomb used to initiate the reaction of the super. If the tolerance of normal fission bombs were known, a factor of 40 below this would seem a reasonable estimate of the limitation on the number of supers. This limit might be lower than the one derived on the basis of the carbon 14 hazard. It should be noted furthermore that the amount of active material needed for the deton-

ation of a super is at the present stage necessarily a crude guess.

Activation of structural or tamper materials used in the bomb construction must also be considered. However, it would seem that a sufficient number of common elements with low cross sections and/or short lived active products are available so that any danger from this source could be easily avoided.

TABLE I

Active Nuclide	Half Life	Radiation	Relative Production	Relative Initial Activity	Relative Activity After 2 Days
C-14	5700 y	β^- 0.150	1.00	1.00	1.00
N-16	8 s	β^- 3.5,10 γ 6.5	4.5×10^{-8}	1000	0
O-19	31 s	β^- 4.1 γ 1.6	7.0×10^{-6}	4.1×10^{4}	0
A-41	110 m	β^- 1.18 γ 1.3	8.7×10^{-3}	2.17×10^{5}	0.013
Br-79	34 h	β^+ 0.9,0.6 γ 0.2	6.8×10^{-10}	1.0×10^{-3}	3.8×10^{-4}
Kr-85	4.6 h	β^- 1.0 γ 0.17,0.37	4.1×10^{-8}	0.45	3.3×10^{-4}
Kr-85	10 y	β^- 0.74	2.7×10^{-8}	1.5×10^{-5}	1.5×10^{-5}
Kr-87	75 m	β^- 4	7.8×10^{-9}	0.31	1.1×10^{-12}
Xe-133	5.3 d	β^- 0.34 γ 0.085	3.0×10^{-9}	1.2×10^{-3}	9.2×10^{-4}
Xe-135	9.2 h	β^- 0.95 γ 0.25	1.2×10^{-9}	6.5×10^{-3}	1.8×10^{-4}
Cs-137*	33 y	β^- 0.8 γ 0.5	7.8×10^{-10}	1.3×10^{-7}	1.3×10^{-7}

* By Beta decay from Xe-137.