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CONFIRMATION OF RADIOACTIVITY IN THYROIDS OF VARIOUS ANIMALS, JULY 15 TO SEPTEMER 10, 1954

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CONFIRMATION OF RADIOACTIVITY IN THYROIDS OF VARIOUS ANIMALS

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Ralph L. Gunther and Hardin B. Jones

Donner Laboratory of Biophysics and Medical Physics
and Radiation Laboratory, Department of Physics
University of California, Berkeley, California

September, 1954

This investigation was undertaken to confirm the results of a similar inquiry by Lester Van Middlesworth of the University of Tennessee.[1] Van Middlesworth found readily measured radioactivity in the thyroids of cattle slaughtered in Memphis, San Francisco, and Boston as part of an apparent widespread contamination by iodine-131. The animals were grown in various parts of the country, as far apart as Florida and the West (probably California). The activity he found had the same decay and absorption characteristics as I^{131}, and was as high as 4.4×10^{-3} μc/g for Florida-raised animals.

In the analysis made at Donner Laboratory, activity of the same order of magnitude as Van Middlesworth indicates for California-slaughtered cattle was initially found in the thyroids of various animals obtained from the School of Veterinary Medicine on the Davis Campus of the University of California, and in the thyroids of cattle raised locally.

METHODS

Most of the thyroids from the Davis animals were whole, but those from a slaughterhouse in the San Francisco Bay area were partly damaged owing to the method of decapitation. The glands or the I^{131} calibrating standard were placed in 50-ml beakers and counted by the same scintillation counter over a period of time sufficient to give approximately 1% accuracy (Tables I and II) or 3-4% accuracy (Table III).

Relative absorption determinations were not made here, but such studies had been made by Van Middlesworth; the purpose of this study was to establish the time-intensity relationships of the concentration of radioactivity in thyroid glands.

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[1] Nucleonics 12, 56 (1954).

The cattle slaughtered locally were reportedly grown near the western part of California, probably in the north, and had been either range-fed or had been given a dry food concentrate for some period up to 100 days in a feed lot before being slaughtered.

DATA RESULTS

Table I

Activity in Animal Thyroids from Davis Campus

Both thyroid lobes are included for samples 1-8, and one for 9 and 10.

No.	Description	Date Killed (1954)	Date Analyzed (1954)	Weight (grams)	Specific Activity[a] (cpm/g)	Specific Activity Corrected to 7-10-1954 ($\mu c \times 10^3/g$)[b]
1	Lamb: Monticello	7/11	7/15	1.4	19	0.16
2	Lamb: Monticello			1.1	7.1	0.059
3	Lamb: from ranch near Davis			1.4	11[c]	(0.08)
4	Sheep: Dixon (54-1079)	7/10	7/15	2.6	6.1	0.053
5	Sheep: Monticello	7/11	7/16	0.95	14[c]	(0.10)
6	Goat: Carmichael (54-1080)	7/10	7/19	6.0	5.2	0.064
7	Pig: Davis (54-1083)	7/10	7/19	3.9	2.2	0.027
8	Dog: Davis (12 yrs. old) (54-1090)	7/12	7/16	0.30	17	0.16
9	Horse: Hayward (54-1081)	7/10	7/16	15.0	2.3	0.021
10	Cow: Davis (54-1092)	7/10	7/16	20.0	1.9	0.017

(a) Above background

(b) This specific activity is expressed as μc/g by assuming activity was due to iodine-131 with an 8.0-day half life, and based on calibration shown in Table II.

(c) Quite dehydrated. Value in parentheses in corrected activity column is estimated value under more normal condition of hydration.

cpm = counts per minute. (Background of 70 to 80 cpm.)

Table II

Activity in Cattle Thyroids Obtained Locally

No.	Analysis Date (1954)	Weight (grams)	Specific Activity(a)		Specific Activity(a) Extrapolated to 7-10-1954 ($\mu c \times 10^3/g$)(b)(c)
			(cpm/g)	($\mu c \times 10^3/g$)(b)	
11(a)	7/22	6.5	15		
11(b)		7.7	9.3	0.065	0.18
12(a)		13.2	6.1		
12(b)		7.4	7.9	0.038	0.11
13(a)		11.8	10.2		
13(b)	7/22	14.4	9.3	0.053	0.16
Repeats of 13(b):					
	7/22	14.4	9.3	0.053	0.16
	7/27	14.4	5.6		0.13
	7/28	14.4	5.2		0.13
	8/9	14.4	2.2		0.16

Counting Standard: (0.15 μc I^{131}) on July 26, 1954; 18.6 x 10^4 cpm/μc(a)

(a) Above background
(b) Assuming I^{131}; each value is the average for a given set of two lobes.
(c) No. 13(b) repeats all on the same lobe.

Runs Nos. 1-10 in Table I are with routine samples from the autopsy room of the School of Veterinary Medicine, and are random samples from farmyard animals brought to autopsy. Runs Nos. 11-13 in Table II show activity of feedlot-fed cows killed at a slaughterhouse in the San Francisco area on July 22, 1954. In Table II each determination represents one lobe, or a part of one; each group of two represents one animal.

The activity in the Davis animal thyroids (Nos. 1-10) is of the same order of magnitude as those from the local slaughterhouse. Variation in the activity of Davis glands was quite large, and may reflect the fact that likely the animals were sick before they came to autopsy. However, the variability of the Davis-obtained samples is approximately the same as that of the Van Middlesworth samples $(\sigma/\overline{n} = 0.7)$, indicating that our sampling was at least as consistent as his.

Activities of all samples Nos. 1-10 were corrected for an 8-day (I^{131}) half-life decay to July 10, 1954 for uniformity, assuming that variation of effects over a few days was not too great. The average for samples 1-10 was

5.3 g weight and 13.6 cpm/g specific activity extrapolated to July 10, with a standard deviation from the mean of the experimental distribution, $\sigma = 9.5$ cpm/g. The average for the local cattle thyroids was 10 g weight per lobe and 9.6 cpm/g specific activity on July 22. Van Middlesworth's standard deviation for a similar set of animals was 90% of the mean value. Assuming approximately this error, our samples (in Table II) had an activity of 9.6 cpm/g ± 8.2 cpm/g. These deviations are, admittedly, enormous, but individual values seem to fall quite symmetrically about the mean ones. It will be seen later that this large sampling error is not as important as it might here appear.

Table III

Activity in Cattle Thyroids Obtained Locally and Slaughtered September 10, 1954

No.	Count (cpm)	Weight (g)	Activity[(a)] (cpm)	Specific Activity[(a)] (cpm/g)
Control	75.3 ± 0.7		0	-------
14	75.4 ± 1.1	11.3	0.1 ± 0.4	-------
15	81.4 ± 2.9	15.3	6.1 ± 3.0	0.4 ± 0.2
16	83.6 ± 2.9	9.5	8.3 ± 3.0	0.9 ± 0.3
17	78.0 ± 2.0	13.1	2.7 ± 2.1	0.2 ± 0.2
18	81.5 ± 2.0	9.9	6.2 ± 2.1	0.6 ± 0.2
19	78.9 ± 3.9	9.6	3.6 ± 4.0	-------
20	82.6 ± 2.7	8.0	7.3 ± 2.8	0.9 ± 0.4

(a) Above background

Background for this experiment was 75.0 ± 0.7 cpm.

Data for samples Nos. 14-20 in Table III were obtained on September 10 from entirely range-fed cows of undetermined origin, probably California. The animals were slaughtered a few hours before this experiment. Methods and techniques employed were the same as before.

It can be seen that the values of activity obtained from individual samples do not, in general, satisfactorily meet the criterion of exceeding 3 times the individual standard deviations. However, the fact that all the samples consistently exceeded background count points to the probability that this difference is significant. We can, accordingly, treat the collection of samples as one cumulative sample, counted over the sum of times of individual determinations. This is justified by the fact that consideration of a number of samples reduces the effect of biological variation on the data, and is valid when, as in this case, each sample is fairly representative of the typical biological situation, and the distribution of activities is reasonably uniform. Computation of results on this basis yields an average specific activity of 0.29 ± 0.09 cpm/g above background. This satisfies the criterion $N \geq 3\sigma$ for statistical significance.

The values obtained for extrapolated (i.e., corrected to July 22, 1954) specific activity of sample No. 13(b) agree with one another within a standard error of 7.4%. This error is considerably less than the standard deviation of a (Poisson) equivalent normal distribution, and therefore probably represents agreement within statistical error. Furthermore, the 18-day repeat value agrees exactly with the original count rate of Sample 13(b), within the instrument error obtained. This tends to support Van Middlesworth's assumption that activity is due to I^{131} with an 8.0-day half life. If this is so, Davis samples (Table I) averaged an equivalent $(0.052 \pm 0.036) \times 10^{-3}$ μc/g specific activity on July 22, 1954; local samples (Table II) averaged an equivalent $(0.073 \pm 0.05) \times 10^{-3}$ μc/g specific activity on July 10, 1954; and local samples from Table III averaged an equivalent $(0.0016 \pm 0.0005^{*}) \times 10^{-3}$ μc/g. These values are based on the instrument standardization made on July 26, 1954. In comparison, Van Middlesworth obtained an average 0.29×10^{-3} μc/g on June 16, 1954, in the thyroids of cattle slaughtered locally.

Recent tests (R. L. Gunther, unpublished) indicate activity per quart of ordinary cow's milk about 20% above background (background of about 75 cpm) which came from some long-lived β-ray-emitting isotope, perhaps K^{40}. Similar results were obtained on a small sample of "stew meat", in which activity was statistically above background. The decay of the radioactivity in the meat was not investigated.

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[*] This is the error of counting, only (standard deviation of the experimental distribution itself averaged 0.0008).

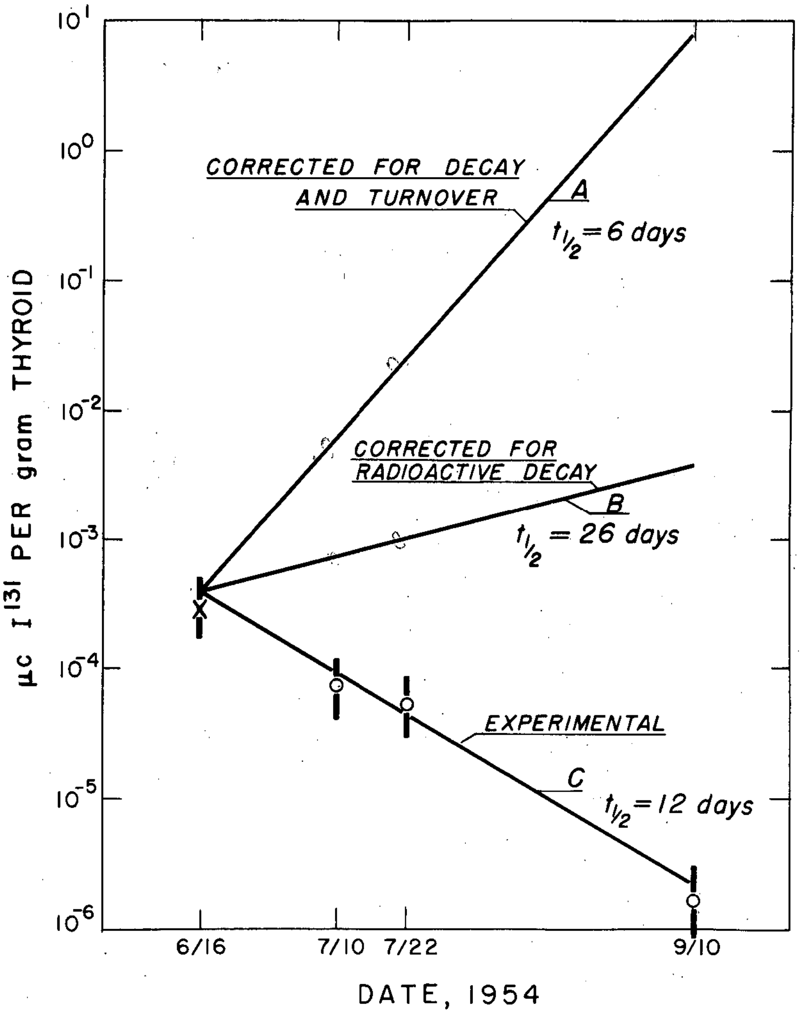
DISCUSSION

The reason for computing a specific activity back to the first day of each set of experiments is based on the assumption of I^{131} activity. This assumption seems to be justified by the fact that the decay of one of the cattle thyroid lobes followed the I^{131} decay within statistical error for more than two periods.

Within the limits of the large deviations involved, the samples of Table I and those of Table II would appear to be approximately equally contaminated, allowing for radioactive decay. Allowance for radioactive decay alone, however, shows a very large discrepancy when, say, the average value of Van Middlesworth's data is compared with that in Table III.

This raises the following point. The wide variance from animal to animal and the relatively small number of animals makes it difficult to draw conclusions concerning the difference in activity in the different samplings of thyroids. However, a rough calculation may be attempted to give a qualitative idea of effects involved. There are not enough experimental observations to interpret Curve C of Fig. 1 as anything but a straight line; it will serve to show the relationships in effect. Curve B is a plot of the activities of Curve C which should have been present in the same animal on June 16, 1954, based only on the 8.0-day half time of of I^{131} decay. If, now, biological turnover is also considered (6- to 12-day period for thyroid iodine), Curve A shows the activity which should have been present in the same animals on June 16, based on an assumed 4.0-day period of over-all iodine disappearance.

Figure 1 shows that the extrapolated value for the activity of the July 22 values is approximately 100 times that found by Van Middlesworth and that of the September 10 values approximately 18,000 times his average value if both radioactive decay and biological turnover occur. As these two effects are undoubtedly taking place at a reasonably constant rate, it is obvious that the extrapolated values fall well outside of experimental or sampling error by Van Middlesworth or us. Inasmuch as the corrected values for decay and turnover show such a large increase over Van Middlesworth's values, then if the material we analyzed is from approximately the same sampling as his, we may assume that from June 16 to September 10 the animals became burdened with an exponentially increasing amount of absorbed iodine.



10^1
10^0
10^{-1}
10^{-2}
10^{-3}
10^{-4}
10^{-5}
10^{-6}
μc I^{131} PER gram THYROID
CORRECTED FOR DECAY AND TURNOVER
A
$t_{1/2} = 6$ days
CORRECTED FOR RADIOACTIVE DECAY
B
$t_{1/2} = 26$ days
EXPERIMENTAL
C
$t_{1/2} = 12$ days
6/16
7/10
7/22
9/10
DATE, 1954
MU-8478

Figure 1

In June we were inclined to believe that food such as grass might be the major source of contamination, due perhaps to fallout of radioactive fission products upon grain that was later ingested. The above analysis of the current data, however, throws doubt on this possibility. First of all, any food concentrate given exclusively to the cattle would be quite old. Second, it can be seen in Table I that the dog absorbed as much iodine as the most active of the lambs and sheep. Furthermore, work in progress bears out the assumption that the contamination seems to be independent of the food experience of the animals. On the other hand, the fact that the activity was found not only in cows, but also in sheep, a horse, a goat, a pig, and a dog indicates the possibility of common contamination. It is difficult to imagine how this might occur through the medium of food, especially at such a consistently increasing rate. Rather, the contaminant may have entered the body through respiratory contact. The most likely source of such material would be airborne fission fragments from the detonation of atomic devices. This would result in continual recontamination as long as the active particles were present in the lower atmosphere, and as long as they continued to be inhaled. The rate of fallout of these aerosols should reach a maximum at some time after their release into the atmosphere, and then gradually diminish. However, the extrapolation described above shows an ever-increasing rate of contamination up to the limit of the sensitivity of our equipment (about 10^{-6} $\mu c/g$ with the size of samples used). The relationship between the variables involved is a true transient equilibrium between the actual contamination and the decay plus turnover. It is possible that the rate of contamination does not increase quite as rapidly from July 22 to September 10 as from June 16 to July 22 if one is justified in drawing Curve C slightly concave downwards, as might be indicated by the trend of the average values per se.

The fact that the rate of absorption of iodine was about 100 times as great on July 22 and more than 10,000 times as great on September 10 as on June 16 might be explained by initial respiration of the heavier particles of the fallout, followed later by smaller aerosols of greater concentration. Stoke's law shows that $F = 6\pi \eta r V$ where F would be the frictional force operating on the aerosols (considered as rigid spheres of radius r) as they move through an atmosphere of viscosity η, with a constant velocity V. We can assume that this relationship is applicable here. At a given value of terminal velocity obtained, the frictional force is balanced by the gravitational force, $F = (4/3)\pi r^3 g\ (\rho_i - \rho_a)$ where g is the acceleration due to gravity, ρ_i is the density of iodine particles, and ρ_a is the density of air through which they move. Then,

$$V = \frac{2r^2 (\rho_i - \rho_a)\ g}{9\ \eta} \quad .$$

It can be seen that particle velocity will vary quite strongly as a function of size, and at the same time will vary as a function of the altitude. Furthermore, as

$$\frac{dV}{dr} = \left[\frac{4\ (\Delta\rho)\ g}{9\ \eta}\right] r \quad ,$$

then, at a given altitude of particles above the earth, a given increase in radius will result in a greater increase in velocity for larger particles than for smaller ones. Therefore a combination of these two effects dictates that as the aerosol falls through air of ever-increasing density, the larger particles fall faster than smaller ones, and particle concentration varies as the height of the particles above the earth. With passage of time, it follows that the aerosol particles present near the earths surface become smaller, more homogeneous in size, and greater in concentration. This is exactly the condition which would fit the observed results. If radioactive decay were not a contributing factor, the above relationships would result in an ever increasing activity in the thyroids of animals which had absorbed the iodine. This condition would correspond to Curve A in Fig. 1. The fact that radioactive decay occurred would bring the increase of activity (with time) down to that of Curve B, were it not for biological turnover which results in a further decrease to Curve C. This tacitly assumes Curve B to represent not only the extrapolation correcting for radioactive decay, but alternatively the correction for biological turnover; the periods for decay and turnover are assumed to be approximately equal (8 days each).

From the above it seems reasonable to conclude that a contamination by air-borne aerosols occurred over a period of time at an (approximately) exponentially increasing rate. This would result in a total dose that was cumulative and due to chronic exposure. It is fortuitous that the highest rates of contamination occur after the isotopic iodine has decayed to a very small fraction of its original activity, thereby resulting in a net decrease in observed activity. It is certainly possible that similar fallout behavior of the longer-lived nuclides would result in considerably greater biological effect if taken into the living system.

Preliminary in vivo tests on human subjects show no activity above background in the vicinity of the thyroids. These determinations, however, were made by equipment which is not sensitive to the small amounts of radioactivity probably present. The human burden when estimated should be approximately one-fifth of the cow iodine burden. This is because the respiratory rate and body size of man are less than those of the cow, whereas the thyroid sizes are approximately the same (ca. 30 g). Thus, the total human thyroid dose is estimated as less than 0.1 roentgen for this entire period from June to September 1954. Therefore the amount of activity found in the animals tested indicates that the dose received by human beings from similar contamination should be within accepted safe tolerances.

SUMMARY

The finding of appreciable quantities of radioactivity in the thyroid glands of cattle by L. Van Middlesworth has been confirmed. This activity follows an 8-day decay, and is probably due to I^{131}. Activity was also found in the thyroids of various animals obtained from the School of Veterinary Medicine on the Davis Campus of the University of California. The contamination is tentatively attributed to respiration of air containing radioactive material. The dose received by human beings from this source is within accepted safe tolerances.

ACKNOWLEDGMENTS

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