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I^{131}

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AEC-GE RICHLAND, WASH.

Contamination of milk after an accidental atmospheric fallout of iodine-131.

R. J. Kirchmann, R. R. Boulenger

"Radioactive contamination in gaseous media," Saclay Symposium,

November 12-16, 1963.

I. Introduction

One of the ways in which contamination of the atmosphere can occur is a reactor accident, which was the case at Windscale (G.B.) in October, 1957.

This accident, during which it has been estimated[1] that 27,000 curies of I^{131} and other radioelements were emitted into the atmosphere, has revealed that, from the viewpoint of the food chain, one of the most important of the fission products in the case of a reactor incident was radioactive iodine, I^{131}.

This conclusion cannot be denied when we consider, especially: the fission yield (3.1%), the very high absorption factor (af = 1.0)[2] after ingestion by the animal, the secretion rate in milk, and finally the half-life (8.05 days) which permits it to be transferred from the animal to man.

Milk, a major part of the diet of young infants, is the main carrier of I^{131}.

The installations at the Centre d'Etude de l'Energie Nucleaire at Mol, are located in a region where the major agricultural resource is dairy products; two thousand producers deliver to 3 dairies, located at least 10 km from nuclear installations. In a circle of 15 km radius, the animal estimated production has been 75 million liters in 1962, about 20% of which was sold as milk for consumption.

Two series of experiments, made in 1962 and 1963, have been done under natural conditions in order to obtain results valid to the agricultural pursuits of this region. These tests enabled us to verify if the figures relative to concentration of I^{131} in milk, extablished during the Windscale accident were also valid for the region adjacent to the C.E.N. installations.

These experiments are also aimed at furnishing information on the influence of such factors as: height and nature of the pasture, season during which contamination occurs, meteorological conditions (especially rain).

II. Experimental Conditions

For the tests done in 1962, we have used a permanent pasture for 3 cows, the pasture divided into 6 sections of about 20 acres each, according to the so-called "rotating" method. This procedural method, standard in developing agricultural operations, has had the consequence that the height of the pasturage at the time of contamination was different for each of the sections.

In 1963 our tests were made in a "temporary" type meadow of mixed flora. For the first experiment, we placed each of the cows in an individual pen and fed it daily a known ration of contaminated grass. In the second experiment, the cows pastured freely in an uncontaminated meadow, but each day we gave them a small, known quantity of fresh, highly contaminated grass.

The cattle used are of the type common in Campine; we have selected our different cows according to their milk production so as to obtain maximum individual differences.

The radio-iodine used was elementary I^{131}, carrier-free in CH_3CH_2OH solution (for certain experiments, the solution also contained 3.4% CCl_4).

This mother solution is then diluted with demineralized water (1:1000) before being atomized.

Meteorological conditions: daily record (temperatures in the sun and in shade, relative humidity, and rainfall) is kept.

Conduction of the Experiments

Contamination

The radioactive solution is sprayed by means of a hand sprayer equipped with a row of four beaks, mounted on a sled which is pulled by a jeep. Personnel wear anti-gas masks, rubber gloves and boots

during the treatment. The personnel go through an installation
for measuring ingested gamma radiation before and about 24 hours
after each experiment during which the radio-iodine is sprayed; no
activity of the thyroid gland has been detected in these persons.

Sampling

Grass: Hand-mowing or use of a motorized cutter, according
to the type of experiment and the surface to be sampled, with
removal of a surface greater than or equal to 1 m^2.

Milk: Milking twice daily (8 and 18 hrs), which is present
practice on the farm.

Treatment and Measurement of the Samples

Grass: The harvest is weighed, then an aliquot is oven dried
to determine the quantity of dry grass material per unit of
surface.

We introduce 500 g of fresh material into a hermetic
flask (3 ℓ), special for gamma spectrometry, and add 10 ml of
formaldehyde to stop any fermentation.

Milk: Production from each milking is measured; we take 3
liters, to which we add 10 ml of formaldehyde as preserving
agent. Measurement by multichannel gamma spectrometry is
done as soon as possible, for the reasons given above.

Results

Contamination of the Grass

Direct retention: the measurement of I^{131} in the air sampled
part (mowed or cut) has let us calculate the percentage of the
atomized I^{131} directly retained in this part of the plant. The
results obtained are shown in Table I.

Evolution in time of specific activity: Two samples for the evolution of specific activities (in microcuries I^{131}/kg dry material) are shown in graphs 1 and 2. We verify that the effective half-lives of I^{131} in the grass (per gram of dry material) are 4.5 $\pm$ 0.72 days during the first experiment (1963) and 6.0 $\pm$ 0.93 days during experiment 2 (1963); the corresponding biological half-lives are, respectively, 10.2 $\pm$ 2 days and 23.5 $\pm$ 4.3 days.

Contamination of the Milk

Time required for the appearance of I^{131} as a function of the moment of contamination: graph 3 shows the evolution of the relative concentration of I^{131}, in relation to the maximum observed peak, as a function of the moment of the day in which contamination occurred.

In our tests, we have assumed that placing the livestock at a predetermined time in a contaminated pasture was equivalent at this moment to a pasture occupied by the livestock.

a. I^{131} activity secreted in the milk: Shape of the secretion curves: placing 3 cows (A,B,C) in contaminated pasture in June, July, and September, 1962, has produced milk with I^{131} content per liter as shown in graphs 4, 5, and 6.

Rainfall (date and height in mm) is also shown in the graphs.

The secretion curves obtained in the special cases of the first two experiments in 1963 are given in graphs 1 and 2.

b. I^{131} activity in milk, expressed in percent of the daily ingested activity. Given the practical impossibility of determining the quantity of fresh grass consumed daily by each of the cows, in the case of the free pasture experiment, we made in 1963

two types of experiments under conditions which let us know the quantity of I^{131} ingested daily.

The results are given by graphs 7 and 8.

Discussion of the Results

Contamination of the Grass

Table 1 shows the large variations in the percentage of I^{131} directly retained by the harvested grass. Two reasons for these observed differences are, probably, the height of the grass at the time of spraying and the technique for harvesting the samples.

Concerning the influence of the grass height at the time of contamination, a preliminary experiment seems to indicate that no linear correlation exists between the quantity of fresh material per unit of surface and the specific activity of the dried material. The exact nature of the correlation curve will be determined by later experiments.

In another way, calculation of the correlation between the quantity of fresh material per unit of surface and the harvested activity per unit of surface, shows that the better correlation coefficient is obtained for an exponential type function (graph 9).

Concerning the harvesting technique, results show that hand-mowing (cut to soil level) permits far better recovery of the radio-isotope than cutting with a power mower.

So we verify that the sampling procedure may have a direct effect on the results when they are expressed as activity per unit of harvested surface (microcurie I^{131}/m^2).

Evolution of the specific activity of grass from the different pens does not appear either to be influenced by the fact of the

presence of livestock or related to atmospheric precipitation, in
any apparent way.

The effective half-life of I^{131} in the grass, obtained under
our experimental conditions in 1963, was in the magnitude of 5 days.

Contamination of the Milk

The secretion curves observed in our experiments result from
a combination of experimental curves obtained in controlled sur-
roundings by various authors, and especially Lengemann[2].

The experimental curves described in the literature are of
two types:

a. Oral administration of a "single" or "unique" dose. Graph 10
    shows the concentration curve for I^{131} in cow's milk obtained
    in this case. If we examine the upper curve (physical decay
    corrected), it seems that the half-life of I^{131} in milk was about
    1.5 days during the 3 or 4 days following administration of the
    radio-element, so this period was about 3 days. However, accord-
    ing to Glascock[3], this period of slow excretion only becomes
    apparent after 9 days instead of 3 or 4 days. According to Loutit[7]
    this change in the slope of the excretion curve was due, mainly,
    to the iodine combined with the proteins.

b. Daily oral administration of I^{131} over a long period of time.
    Graph 11 shows the concentration curve for I^{131} in cow's milk
    obtained in this so-called "multiple-dose" type of experiment.

    The upper curve (physical decay corrected) indicates that
    we approach an equilibrium with the contaminated ration after
    about 7 days.

These two types of experiment constitute relatively simple
cases. In our tests, undertaken in natural surroundings, conditions

were much less favorable than in those above, for different para-
meters came into play simultaneously.

Examination of graph 6 shows an interesting fact: cow (B),
placed in the contaminated pasture two days after her companions,
did not show a peak for I^{131} secretion in her milk. One explana-
tion, which requires verification in future tests, was that the
I^{131} retained by the plant was incorporated and metabolized,
which could make it less available for the animal than the I^{131}
in freshly sprayed solution.

On the other hand, we note that the secretion curves show
fluctuations which, in general, occur simultaneously for all
three cows. The rises observed could have resulted from an in-
crease in the specific activity of the grass following uptake of
I^{131} deposited in the "mat".

As to I^{131} activity secreted in the milk, in percent of in-
gested dose, graphs 7 and 8 show that the observed values lie
between 0.22 and 0.6% of the daily ingested dose per liter of
milk. Results obtained during our 1962 experiments were also between
these extremes.

Moreover, results from graph 6 would seem to indicate that
there is no systematic difference between morning and evening
treatments.

III. General Conclusions

Results concerning the excretion of I^{131} in milk are slightly lower than
those found by other researchers[6], if we consider the results relative to
secreted activity expressed as a percentage of the ingested activity.

Values reported during the Windscale accident, to whit that a deposition
of 0.4 μ I^{131}/m^2 on the grass has produced milk containing 0.065 μCi I^{131}/l,

... not been observed during our 1962 experiments. Maximum values were

.0056 µCi I^{131}/ℓ for a deposition of 0.1 µC/m², which corresponded to

.0224 µCi I^{131}/ℓ for a deposition of 0.4 µCi/m².

In 1963, the maximum observed value was 0.024 µCi I^{131}/ℓ for a deposition of 0.62 µCi I^{131}/m², and a specific activity of the grass of 0.77 µCi I^{131}/kg of dry material.

Two reasons could be advanced to explain this difference:

a. The Windscale accident occurred in October, that is to say at a time of year when the grass is no longer growing very much and is less thick (in dry material/m²), which leads to a higher specific activity than in our experiments made in the summer and at the start of autumn.

b. While the chemical and physical nature of the I^{131} emitted during the Windscale accident is uncertain, there was not more than 5% of activity collected near installations associated with particles larger than a micron[1]. Moreover, results obtained by different methods have indicated that 80% of the activity due to I^{131} was retained in the grass. Moreover the work of Thomas [8] and Chamberlain[9] and others, have shown that factors of accumulation in plants for iodine in vapor form could be very important. But in our experiments we have contaminated the pasture with I^{131} in finely atomized aqueous solution.

The shape of the secretion curves has shown us that secondary phenomena probably intervene, such as: modification with time of the form of I^{131} in the plant, washing away by rain, and taking up the I^{131} again from the "mat".

These factors should be studied separately by experiments under controlled conditions.

	Date	Deposited Activity $\mu Ci\ I^{131}/m^2$	Activity of Harvested Grass		% Directly Retained in Harvested Part	Remarks	Natu of Spec:
			$\mu Ci\ I^{131}/m^2$	$\mu Ci\ I^{131}/kg\ MS$			
	6/14/62	0.043(*)	0.42×10^{-2}	0.026	9.7	(*)Balance of I^{131} used was not made	1 x
		0.043	0.97×10^{-2}	0.010	22.5	Hand Cut	1 x
2	7/19/62	0.10	1.60×10^{-2}	0.153	16.0		Aver
		0.10	4.27×10^{-2}	0.073	42.7	Hand Cut	4 x
3	9/7/62	0.045	0.02×10^{-2}	0.126	0.44		
		0.045	0.21×10^{-2}	0.017	4.6	Machine Cut	1 x
4	9/25/62	0.085	0.09×10^{-2}	0.033	1.0	Machine Cut	1 x
5	10/1/62	0.10	0.58×10^{-2}	0.388	5.8	Machine Cut	1 x Aver
1	7/11/63	0.62	0.12	0.77	19.3	Hand Cut	10 x
2	8/12/63	112	23.4	119	20.8	Hand Cut	1 x
3	9/25/63	0.66	0.60	1.57	90.9	Hand Cut (High Grass)	1 x
		0.66	0.09	1.17	13.6	Hand Cut (Low Grass)	1 x

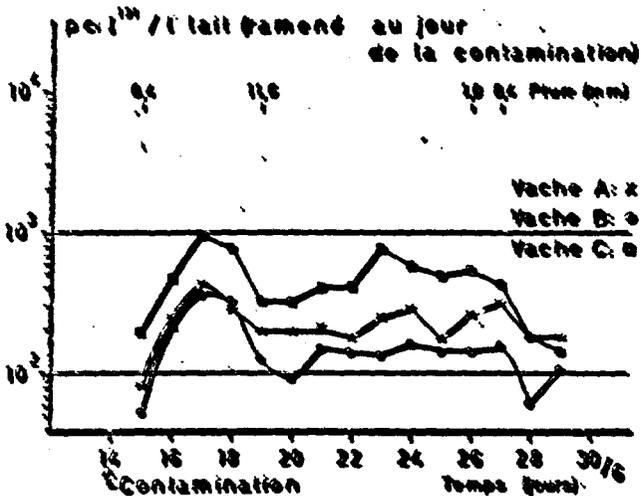TABLE I - DIRECT RETENTION OF I^{131} BY GRASS
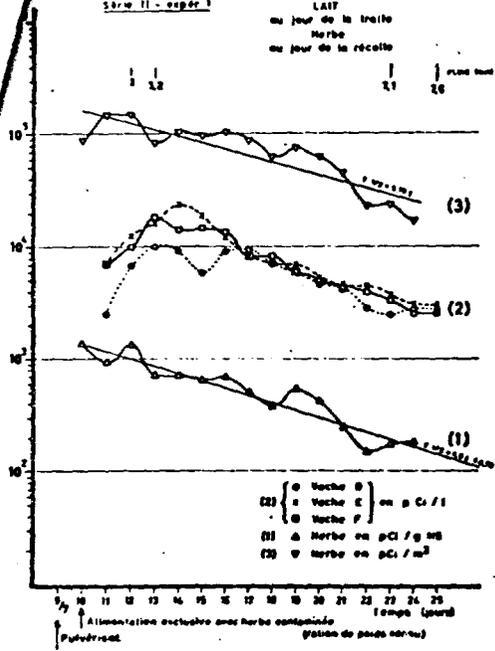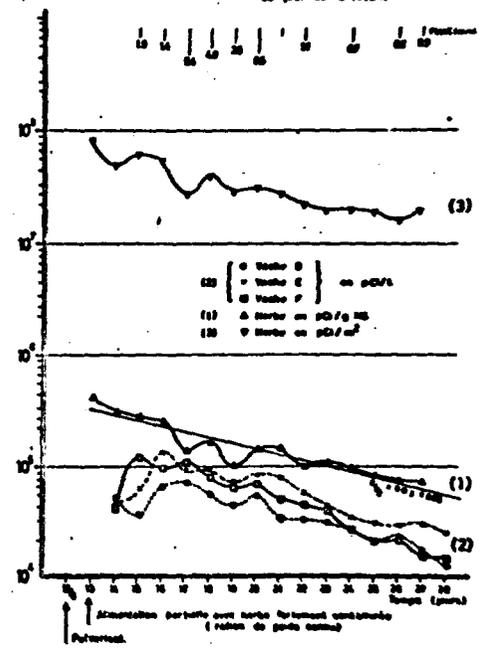
Graphique 1

Graphique 2

Evolution de la concentration relative d'I[131] dans le
lait, en fonction du moment de la journée auquel
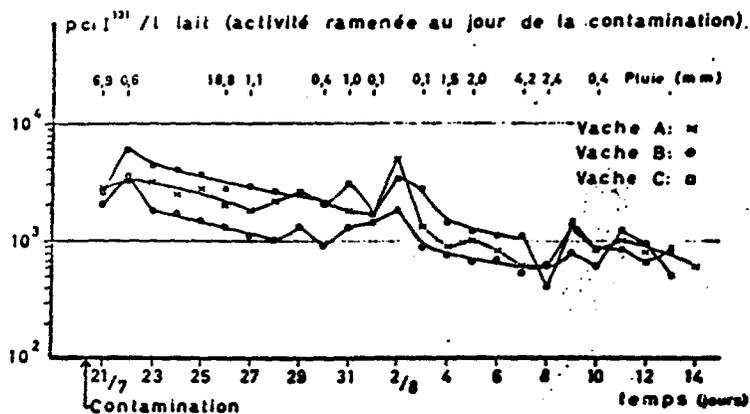a lieu la contamination. (heures de traite: 8 h et 18 h)
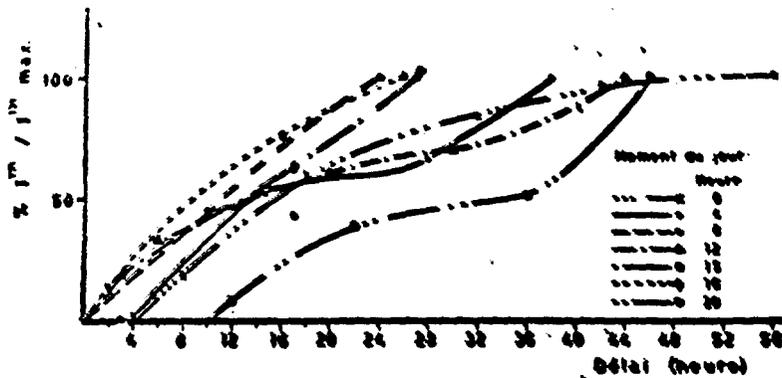
Graphique 3

Graphique 4

Activité spécifique de l'herbe, au jour de la contamination
0,113 µ Ci I[131] / kg matière sèche
0,029 µ Ci I[131] / m²

Graphique 5

Activité spécifique de l'herbe, au jour de la contamination
0,072 µ Ci I[131] / kg matière sèche
0,012 µ Ci I[131] / m²
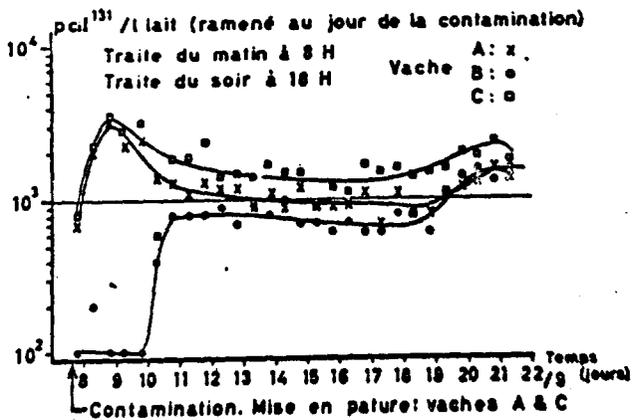
Graphique 6

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Expér. ^{131}I (1963)

Série II - expér. 1

Activité sécrétée par litre de lait /

Activité ingérée

Graphique 7

Expér. ^{131}I (1963)   Série II - expér. 2

Activité secrétée par litre de lait /.

Activité ingérée

Graphique 8

Influence de la hauteur de l'herbe sur $\left[\begin{array}{l}\text{l'activité / m}^2 \text{ de l'herbe récoltée (I)}\\ \text{l'activité / kg matière sèche de l'herbe}\\ \hspace{3cm}\text{récoltée (O)}\end{array}\right]$

Dépôt uniforme de
0,66 µCi ^{131}I / m²,
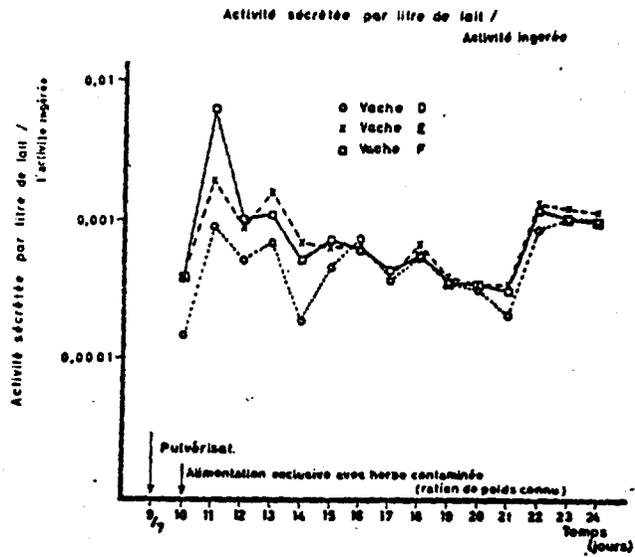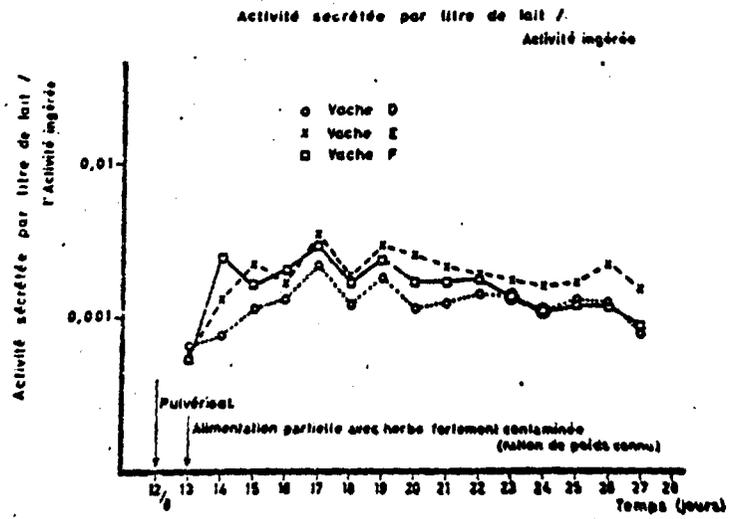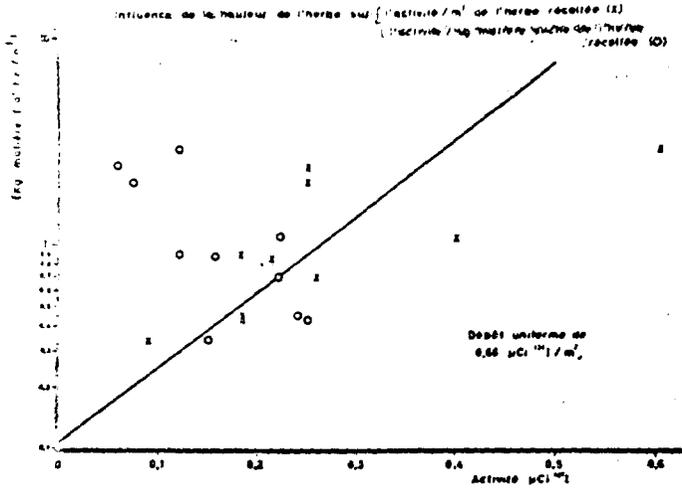
Graphique 9

Secretion of I^{131} in cow's milk

(d'après F.W. Lengemann and C.L. Comar)

Decay corrected o—o
Decay not corrected □—□

Percent of single
dose I^{131} per liter

Days
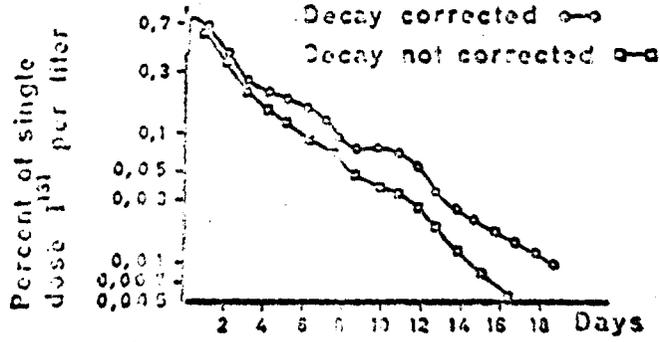
Graphique 10

Concentration of I^{131} in cow's milk

(d'après F.W. Lengemann and C.L. Comar)

Graphique 11
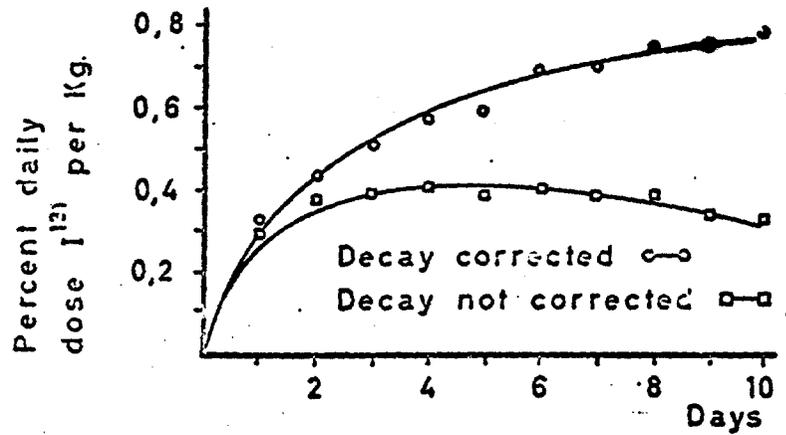
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