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"Radioactive contamination in gaseous media,"
Society Symposium, November 12-16, 1963
Translated by [illegible], Hanford Laboratories

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Contamination of milk after an accidental atomospheric 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, established 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

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 ± 0.72 days during the first experiment (1963) and 6.0 ± 0.93 days during experiment 2 (1963); the corresponding biological half-lives are, respectively, 10.2 ± 2 days and 23.5 ± 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

[redacted]

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 surroundings 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, according 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

[redacted]

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

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[8], 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,

ve not been observed during our 1962 experiments. Maximum values were .0056 μCi I^{131}/ℓ for a deposition of 0.1 μC/m^2, which corresponded to .0224 μCi I^{131}/ℓ for a deposition of 0.4 μCi/m^2.

In 1963, the maximum observed value was 0.024 μCi I^{131}/ℓ for a deposition of 0.62 μCi I^{131}/m^2, 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/m^2), which leads to a higher specific activity than in our experiments made in the summer and at the start of autumn.

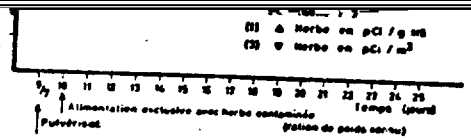
b. While the chemical and physical nature of the I^{131} emitted during the Windscale accident is uncertain, there was not more than 5% of activity collected near installations associated with particles larger than a micron(1). Moreover, results obtained by different methods have indicated that 80% of 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 atomised 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".

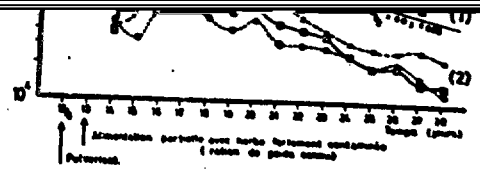
These factors should be studied separately by experiments under controlled conditions.

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

	Date	Deposited Activity $\mu Ci\ I^{131}/m^2$	Activity of Harvested Grass		% Directly Retained in Harvested Part	Remarks	Natu of t 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

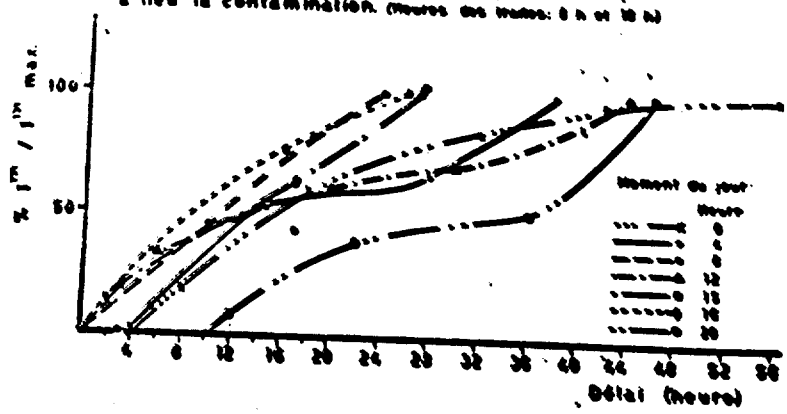


Graphique 1



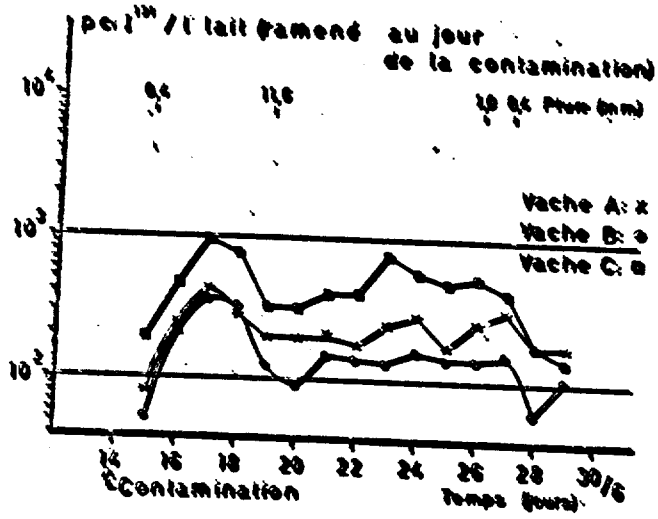
Graphique 2

Evolution de la concentration relative d'I^{131} dans le lait, en fonction du moment de la journée auquel a lieu la contamination. (Heures des traites: 8 h et 18 h)



Graphique 3

Activité spécifique de l'herbe au jour de la contamination
0,010 µ Ci I^{131}/ kg matière sèche
0,007 µ Ci I^{131}/ m²

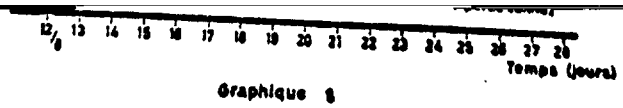


Graphique 4

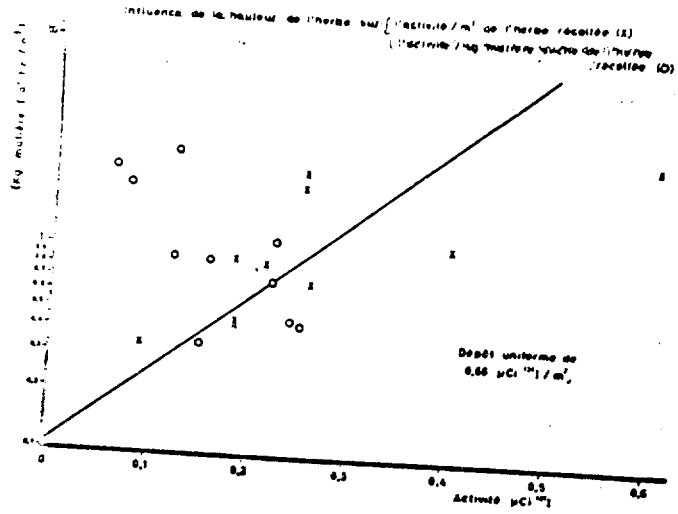
8 9 10 11 12 13 14 15 16 17 18 19 20 21 22/9 (jours)

Contamination. Mise en pature: vaches A & C

Graphique 6

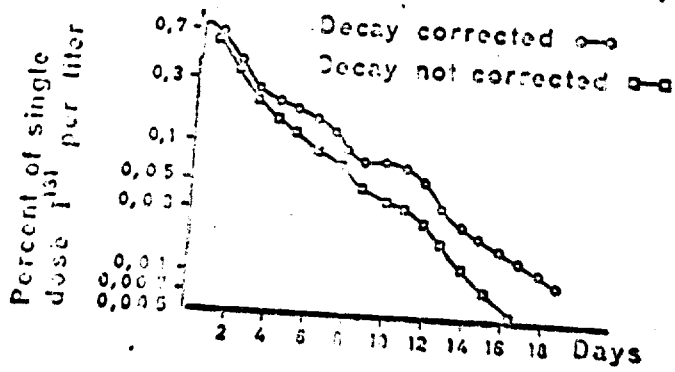


Graphique 8



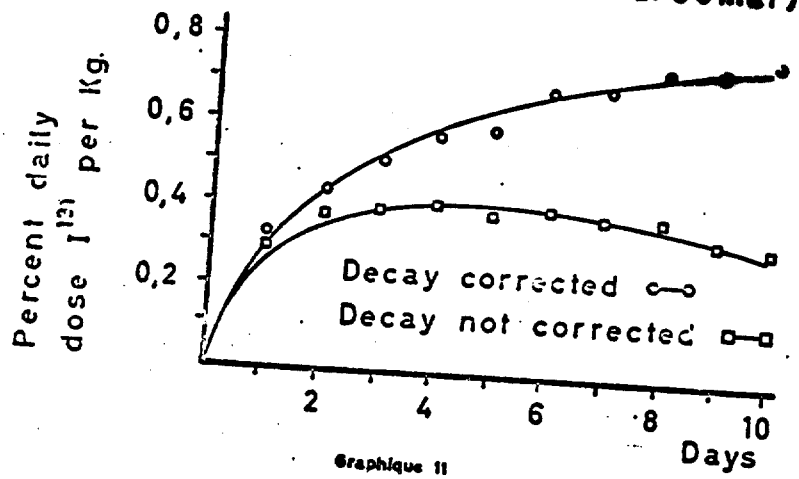
Graphique 9

Secretion of I^{131} in cow's milk
(d'après F.W. Lengemann and C.L. Comar)



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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