THE ACCUMULATION OF RADIOACTIVE IODINE IN THE THYROIDS OF GRAZING ANIMALS SUBSEQUENT TO ATOMIC WEAPON TESTS

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Summary

Radioactivity due to ^{131}I was found in thyroid glands collected from sheep and cattle depastured on areas in various parts of Australia subsequent to atomic weapon tests conducted during 1956 at Monte Bello and at Maralinga. Thyroids from cattle were found to contain up to 830 mμc ^{131}I (37 mμc ^{131}I/g of tissue) and from sheep, up to 144 mμc ^{131}I (70 mμc ^{131}I/g of tissue). The uneven degree of contamination of pastures was emphasized by the fact that some of the highest concentrations of ^{131}I were observed in glands collected from individuals of flocks and herds grazing on terrain 1500–2000 miles distant from the site of the explosions. As the tests proceeded, fluctuations in the ^{131}I content of the thyroid glands of grazing stock indicated that many areas received repeated dressings of radioactive debris.

Contamination of Adelaide and its environs with fall-out from the third Maralinga explosion provided an opportunity to establish unequivocally that extremely little if any of the considerable amount of ^{131}I that became concentrated in the thyroids found its way to the glands via the lungs; and the occasion rendered possible a detailed study of the rates of rise and fall of ^{131}I in the thyroid glands of grazing animals in relation to the degree of contamination of the pastures.

The value of the ^{131}I in the thyroid glands as a measure of the hazards imposed by contamination of terrain by troposphere fall-out supervening on atomic weapon tests is considered in light of the findings.

The speed with which grazing dairy cows gather radioactive debris that has been deposited on their pastures, as indicated by the rise and fall of the ^{131}I in their thyroid glands, is discussed in relation to the hazard entailed in the passage of bone-seeking radioactive constituents of fall-out via milk to human populations.

I. INTRODUCTION

The primary purpose of this investigation was to assess the usefulness of the ^{131}I concentration in the thyroids of grazing animals as an integrating measure of the degree of hazard entailed in the contamination of terrain by residua from atomic explosions. When it was undertaken, knowledge of the chemical state in which iodine isotopes exist in the air-borne debris was uncertain, and interpretation of the chain of events responsible for the accumulation of radioactivity in the thyroid tissue of the higher animals was confused. Most, if not all, of this iodine was thought to persist for a considerable period either as a gas or as an aerosol, and thus to find its way to the thyroids via the lungs. Were this so, the radioactivity of the iodine components of atomic fission residua from nuclear weapon tests might itself become

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a serious health hazard, as the amounts likely to be delivered to human foetal thyroids could be dangerous. Alternatively, were the radioactive iodine found in the thyroids of grazing animals derived from solid particles deposited on the pastures and absorbed via the alimentary canal, the amount accumulated in the thyroid gland would provide a useful index of the capacity of grazing animals to assimilate and to concentrate within animal products the longer-lived radioactive constituents of fall-out. The ability of grazing dairy cows, for instance, to gather the fall-out deposited over relatively large areas and secrete into their milk some of its components in a concentrated and easily assimilable form, is perhaps the most important single incident in a number of events that concentrate ^{90}Sr and other bone-seeking radioactive constituents, and so enhance the risks associated with these particular isotopes.

Some aspects of the danger of ^{90}Sr accumulation within the human skeleton have been considered in the report of the British Medical Research Council published as a White Paper on Hazards (1956). This document aimed to assess the possible untoward effects that fall-out from U.S.A. and U.S.S.R. megaton nuclear weapon tests may have on the inhabitants of the British Isles, and so risks imposed by radioactive material settling from the stratosphere were the primary consideration.

The degree of contamination of any part of the earth's surface by deposition of widely dispersed radioactive material from the stratosphere, however, is quite small when compared with the degree of contamination of certain areas in Australia with radioactive material deposited from the troposphere subsequent to the recent weapon tests.

The results of a survey of the amounts of ^{131}I found in the thyroid glands of animals grazing in various parts of Australia (Fig. 1) that are reported in this paper indicate that extensive areas of Australia have been contaminated, and that some of the more heavy precipitations occurred on terrain situated over 1500 miles from the site of the explosions, in areas more or less thickly populated.

Published Reports of Radioactivity in Thyroid Glands

In September 1954, Van Middlesworth (1954) reported that thyroid glands taken from cattle grown in various parts of U.S.A. and slaughtered at Memphis, at San Francisco, and at Boston were radioactive and that the radioactivity had the same decay and absorption characteristics as ^{131}I. The highest concentration observed was $4 \cdot 4$ mμc ^{131}I/g in a gland from one of a group of range-fed cattle raised in Florida; and the average of glands from animals slaughtered in Tennessee on June 16, 1954, was $0 \cdot 29$ mμc ^{131}I/g.

Shortly afterwards, Gunther and Jones (1954) reported from the Radiation Laboratory at Berkeley measurements which confirmed the observations of Van Middlesworth. The concentrations of ^{131}I in the thyroids measured there, extrapolated to June 16 on the assumption of an 8-day half-life, ranged between $0 \cdot 13$ and $1 \cdot 4$ mμc ^{131}I/g with a mean of $0 \cdot 56 \pm 0 \cdot 288$ mμc ^{131}I/g, and it was concluded tentatively that ^{131}I, originating from nuclear weapon tests and air-borne in the interim as aerosols, had been absorbed by these animals via their lungs.

Iodine, in gaseous form or in aerosol suspensions, is known to be absorbed through the lungs with extraordinary efficiency, and the possibility that ^{131}I found its way into the body via this route was strengthened by the Californian observations which indicated a similar concentration of ^{131}I in all thyroids that were tested, irrespective of whether they were from carnivores or from grazing herbivores.

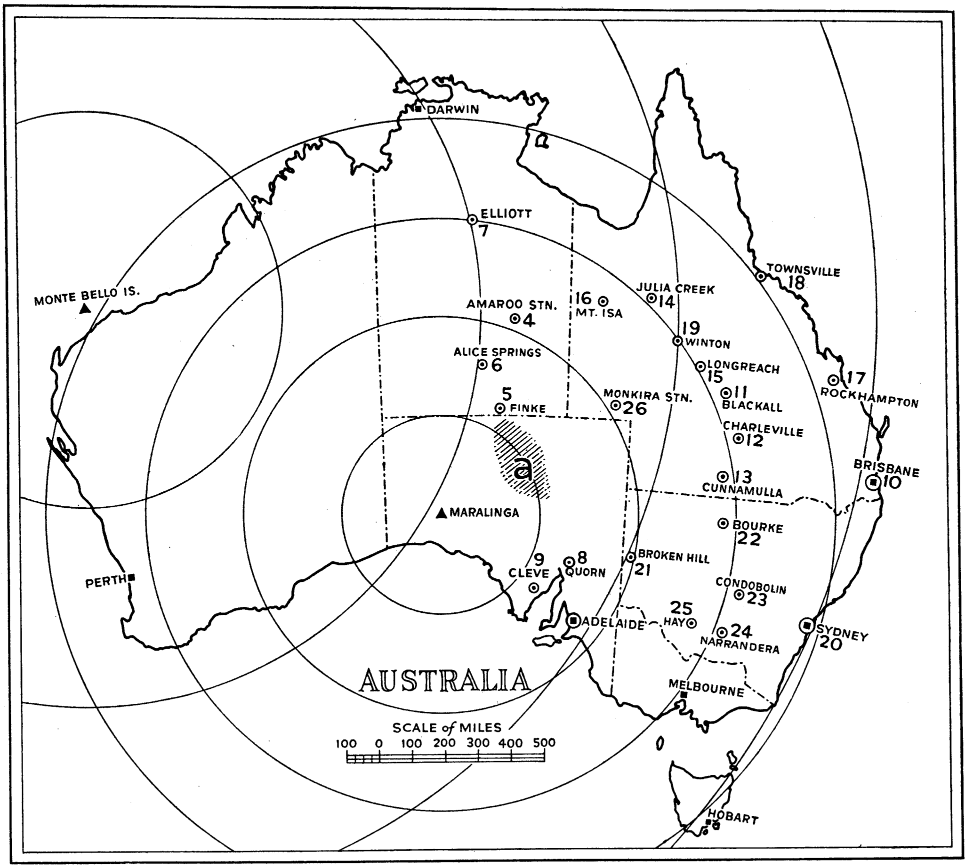

Fig. 1.—Location of areas (cf. Table 1) from which thyroid glands were collected. The circles centred on Monte Bello are *c*. 600 miles apart and those centred on Maralinga are *c*. 300 miles apart.

Later, Van Middlesworth (1956) published results of a further series of observations which focused medical attention on the danger of fall-out. Concentrations of ^{131}I estimated in glands collected at abattoirs within a radius of 200 miles from Memphis at relatively short intervals during the period extending from October 1954 to March 1956 indicated a series of peaks of concentrations of ^{131}I within the thyroids of cattle which ranged from 0·01 to 10 mμc ^{131}I/g thyroid, and which could be correlated with the American and Russian atomic explosions.

Human thyroids collected at autopsies conducted in the Memphis hospitals during this period had in them very much lower concentrations of ^{131}I—never more than 0·5 per cent. of the maximum observed in cattle*—and so doubt was shed on the suggestion that all, or even a considerable part of the ^{131}I that accumulates in the thyroids of grazing animals is absorbed through the lungs.

II. Radioactive Iodine in Thyroid Glands of Sheep and Cattle Grazing in Various Parts of Australia Subsequent to the 1956 Atomic Weapon Tests

In 1956 arrangements were made for a periodic collection of thyroid glands from sheep and cattle grazing at a number of sites distributed over northern and north-eastern Australia in areas likely to be traversed by the plumes from the Maralinga series of tests. These arrangements had, of necessity, to be made by correspondence, and as security restrictions that were imposed forbade frank disclosure of the purpose of the collections, the extent of the survey was somewhat limited. However, besides settling unequivocally some questions of a more basic scientific nature, the data demonstrated the usefulness of the ^{131}I content of the thyroid glands of grazing stock as a measure of the degree of hazard entailed in the contamination of particular areas by radioactive debris from nuclear weapons.

Collection-site locations are indicated in Figure 1. Analytical findings are summarized in Table 1. The more complete data from Adelaide and from the Alice Springs series are the basis of Figures 2, 3, 4, and 5.

Prior to mid-May all glands examined contained <5 $\mu\mu$c ^{131}I/g, which was the low limit of the sensitivity of the apparatus at our disposal.

A few days subsequent to the first Monte Bello explosion (May 16, 1956), activity due to ^{131}I could be detected in thyroid glands collected from sheep and cattle depastured on areas distributed between latitudes 24° and 30°S. within a band approximately 500 miles wide stretching from the western to the eastern seaboards.

After the second Monte Bello explosion (June 19, 1956) the concentrations of ^{131}I within the thyroids of animals in these regions increased to levels a hundredfold or more above those observed after the first explosion.† In some instances the observed concentrations at this period exceeded by approximately an order of magnitude the highest range of the ^{131}I concentrations—the Memphis figures—reported in the literature by American observers (Van Middlesworth 1956).

The concentrations of ^{131}I observed in the thyroids of grazing stock after the second Monte Bello explosion indicated that some of the areas most heavily contaminated with fall-out from the plume of this event were far distant—1500–2000

* The "standard" measure of the tidal air which in the normal course of respiration is passed through the lungs of a 70-kg man (c. 20 cu. m./day) is about one-fifth of the tidal air of a grown bullock and, as the weights of the respective thyroid glands are approximately equal, the expected concentration of ^{131}I in a human thyroid tissue would be approximately one-fifth of that found in the thyroids of cattle breathing the same air, if the ^{131}I were absorbed through the lungs and concentrated within the respective thyroids with equal efficiency.

† At Rockhampton, for instance, thyroids collected from cattle on May 29, 1956, and June 22, 1956, contained respectively 3·2 and 2·5 mμc ^{131}I, and on July 12, 1956, 440 mμc ^{131}I. The explosion responsible for the low level occurred on May 16, 1956, and for the high level on June 19, 1956.

Table 1

RADIOACTIVE IODINE CONTENT OF THYROID GLANDS OF SHEEP AND CATTLE

The procedure employed for the estimation of iodine concentration was essentially as follows: the gland was fused with KOH at 550°C, the aqueous solution of the melt was acidified with H_2SO_4 in presence of excess $NaNO_3$, and exhaustively extracted with $CHCl_3$, in which solvent, after washing with $NaHCO_3$, the iodine was titrated with $0 \cdot 01$N $Na_2S_2O_3$ in the presence of $NaHCO_3$. Observed recoveries were over 95 per cent. A dash indicates that the sample was not analysed. A 1-in. diameter well-type scintillation counter was employed to estimate the ^{131}I content of the thyroid tissue (usually of the whole gland). The γ-radiation was measured in a narrow (1 V) channel at the 0·364 MeV peak of the γ-energy spectrum of ^{131}I. Prior to each set of determinations and at intervals throughout, the instrument was adjusted to the 0·364 MeV peak and calibrated against standards provided by the X-ray and Radium Laboratory, Commonwealth Department of Health. The ^{131}I, determined in this way, was proven by γ-energy spectra and decay curves of samples periodically selected at random. There was no evidence of any radioactive substance other than ^{131}I in the glands examined. Most measurements were made at least 14 days subsequent to the origin of the radioactive fission products of which the ^{131}I was an integral part. The amount of ^{131}I found on analysis was adjusted to the day of slaughter by extrapolation according to $\tau^{\frac{1}{2}} = 8$ days. In those instances where only a part of the thyroid gland was received, the figures for total ^{131}I are low. A dash indicates that the whole gland contained <5 $\mu\mu$c ^{131}I. The two Monte Bello tests took place on 16.v.56 and 19.vi.56, and the four Maralinga tests on 27.ix.56, 4.x.56, 11.x.56, and 22.x.56 respectively

Animal	Date of Slaughter	I in Thyroid (mg I/g dry wt.)	Radioactivity of Thyroid at Time of Slaughter	
			mμc ^{131}I Total	mμc ^{131}I per Gram Tissue
Site 4: Amaroo Station, N.T.				
Cattle	9. v.56	5·2	—	—
,,	25. vii.56	5·2	312·0	21·0
,,	3.viii.56	3·5	78·0	7·8
,,	3. ix.56	4·1	8·0	0·44
,,	17. x.56	6·3	35·5	2·2
,,	20. xi.56	4·6	104·0	8·4
Site 5: Finke, N.T.				
Cattle	20. v.56	8·3	—	—
,,	2. vii.56	3·6	67·2	2·24
,,	15.viii.56	2·0	6·68	0·26
,,	22. ix.56	5·7	0·8	0·03
,,	21. x.56	5·1	19·8	1·1
,,	23. x.56	4·6	17·8	1·3
,,	21. xi.56	6·2	5·0	0·46
Site 6: Alice Springs, N.T.				
Cattle	28. vi.56	3·4	33·0	1·65
,,	28. vi.56	5·8	67·2	2·24
,,	2. vii.56	5·6	38·0	1·9
,,	5. vii.56	2·2	28·8	2·0
,,	5. vii.56	3·1	38·4	2·6
,,	9. vii.56	8·6	39·0	2·2
,,	12. vii.56	2·8	40·0	2·5
Site 6: Alice Springs, N.T. (Continued)				
Cattle	16. vii.56	—	39·6	2·2
,,	16. vii.56	3·0	42·0	1·4
,,	17. vii.56	4·0	31·2	2·2
,,	19. vii.56	3·6	19·2	1·1
,,	19. vii.56	—	32·2	1·6
,,	23. vii.56	4·5	32·0	1·3
,,	23. vii.56	—	19·6	1·1
,,	26. vii.56	2·9	32·6	1·63
,,	30. vii.56	4·9	19·8	1·6
,,	30. vii.56	—	24·4	1·0
,,	2.viii.56	3·3	15·4	0·7
,,	2.viii.56	—	15·0	0·75
,,	9.viii.56	4·1	68·0	2·7
,,	9.viii.56	3·3	17·5	1·3
,,	13.viii.56	3·9	14·5	0·5
,,	16.viii.56	3·3	12·2	0·5
,,	20.viii.56	3·9	3·8	0·2
,,	23.viii.56	3·4	6·2	0·4
,,	27.viii.56	4·7	4·8	0·16
,,	30.viii.56	5·9	5·0	0·25
,,	30.viii.56	4·1	2·6	0·1
,,	3. ix.56	3·4	2·4	0·1
,,	10. ix.56	2·6	2·4	0·08
,,	13. ix.56	8·8	1·68	0·04
,,	17. ix.56	3·4	0·9	0·03
,,	24. ix.56	4·0	0·4	0·02

TABLE 1 (*Continued*)

Animal	Date of Slaughter	I in Thyroid (mg I/g dry wt.)	Radioactivity of Thyroid at Time of Slaughter	
			mμc ¹³¹I Total	mμc ¹³¹I per Gram Tissue
Site 6: *Alice Springs, N.T.* (Continued)				
Cattle	27. ix.56	5·3	0·44	0·02
,,	1. x.56	5·9	0·5	0·035
,,	4. x.56	3·8	1·17	0·05
,,	8. x.56	3·9	2·0	0·09
,,	11. x.56	5·4	10·5	0·34
,,	15. x.56	4·3	10·0	0·42
,,	18. x.56	5·0	6·5	0·44
,,	22. x.56	7·3	8·7	0·41
,,	25. x.56	3·6	8·9	0·41
,,	29. x.56	5·2	28·3	1·0
,,	1. xi.56	4·6	40·0	1·6
,,	5. xi.56	5·5	51·0	1·7
,,	8. xi.56	4·8	51·0	1·6
,,	12. xi.56	2·8	29·0	1·2
,,	15. xi.56	3·1	41·0	1·45
,,	19. xi.56	2·6	24·0	0·95
,,	22. xi.56	4·8	10·9	0·57
,,	22. xi.56	5·3	16·3	0·81
,,	3. xii.56	6·2	8·4	0·3
,,	6. xii.56	4·8	7·0	0·29
Site 7: *Elliott, N.T.*				
Cattle	7. v.56	3·8	—	—
,,	25. vi.56	3·5	91·6	4·58
,,	6.viii.56	4·4	17·8	0·9
Site 8: *Quorn, S.A.*				
Sheep	8. v.56	7·5	—	—
,,	2. vii.56	6·1	0·44	0·11
,,	2.viii.56	2·9	1·5	0·5
,,	30.viii.56	2·2	0·5	0·07
,,	27. ix.56	4·6	0·18	0·04
,,	23. x.56	2·1	6·6	2·0
Site 9: *Cleve, S.A.*				
Sheep	29. iv.56	6·6	—	—
,,	29. vi.56	7·8	—	—
Cattle	1.viii.56	3·3	3·4	0·18
Sheep	3. ix.56	4·4	—	—
,,	11. x.56	8·2	1·0	0·18
,,	31. x.56	7·7	14·0	5·8
Site 10: *Animals Slaughtered at Brisbane Abattoirs*				
Cattle	28. v.56	6·0	2·8	0·1
,,	3. vii.56	6·4	22·5	1·2
,,	26. vii.56	4·0	31·8	1·9
,,	9.viii.56	2·6	9·2	0·4
,,	2. x.56	5·0	1·18	0·07
,,	29. x.56	4·8	7·2	0·36
,,	12. xi.56	6·4	16·2	0·65
Site 11: *Blackall, Qld.*				
Sheep	8. v.56	2·0	—	—
,,	27. vii.56	5·2	30·0	6·6
,,	19.viii.56	2·7	3·6	1·2
,,	7. ix.56	5·4	0·8	0·2
,,	31. x.56	3·6	86·0	31·0
Site 12: *Charleville, Qld.*				
Sheep	15. v.56	3·7	—	—
,,	29. vii.56	5·5	38·0	12·5
,,	23.viii.56	2·8	5·0	1·2
,,	18. ix.56	4·0	—	—
,,	22. x.56	3·5	14·4	4·8
,,	16. xi.56	1·8	23·0	4·6
Site 13: *Cunnamulla, Qld.*				
Cattle	22.viii.56	5·1	1·6	0·1
Site 14: *Julia Creek, Qld.*				
Sheep	16. v.56	4·6	—	—
,,	13. vii.56	6·0	144·0	70·0
,,	15.viii.56	2·6	7·1	1·4
,,	26. ix.56	6·3	0·41	0·09
,,	1. x.56	3·0	2·57	0·51
,,	23. xi.56	3·9	74·0	13·4
Site 15: *Longreach, Qld.*				
Sheep	3. v.56	3·0	—	—
,,	4. vii.56	1·4	131·0	44·0
,,	25. vii.56	3·0	43·0	8·6

TABLE 1 (*Continued*)

Animal	Date of Slaughter	I in Thyroid (mg I/g dry wt.)	Radioactivity of Thyroid at Time of Slaughter	
			mμc ^{131}I Total	mμc ^{131}I per Gram Tissue
Site 15: *Longreach, Qld.* (Continued)				
Sheep	7. ix.56	2·2	1·0	0·25
„	28. ix.56	1·7	—	—
„	9. xi.56	1·0	101·0	26·0
Site 16: *Mt. Isa, Qld.*				
Cattle	28. ix.56	4·5	5·8	0·38
„	7. xi.56	3·9	181·0	7·5
Site 17: *Rockhampton, Qld.*				
Cattle	29. v.56	4·8	3·2	0·15
„	22. vi.56	6·2	2·5	0·15
„	12. vii.56	2·0	440·0	17·5
„	9.viii.56	5·3	18·7	0·89
„	20. ix.56	5·9	0·6	0·03
„	30. x.56	4·8	47·0	2·2
„	9. xi.56	4·6	57·0	3·8
Site 18: *Townsville, Qld.*				
Cattle	29. vii.56	2·3	29·4	1·47
„	17.viii.56	4·2	5·2	0·26
„	18. ix.56	4·5	0·88	0·03
„	16. x.56	7·2	22·0	0·86
„	30. xi.56	6·7	88·0	5·4
Site 19: *Winton, Qld.*				
Sheep	13. v.56	1·8	—	—
„	8.viii.56	3·7	7·5	5·0
„	9.viii.56	5·1	2·9	0·7
„	10. ix.56	1·3	—	—
Site 20: *Animals Slaughtered at Sydney Abattoirs*				
Cattle	26. vi.56	5·5	—	—
„	23. vii.56	7·0	0·9	0·05
„	26. ix.56	6·4	0·2	0·005
Site 21: *Broken Hill, N.S.W.*				
Sheep	20. v.56	1·8	0·42	0·08
„	3. vii.56	4·2	—	—
Site 22: *Bourke, N.S.W.*				
Sheep	31. v.56	2·5	0·53	0·1
„	17. vi.56	3·7	0·23	0·125
„	17. vii.56	3·7	6·0	1·5
„	29.viii.56	4·9	0·27	0·09
„	12. xi.56	5·2	14·7	3·1
Site 23: *Condobolin, N.S.W.*				
Sheep	10. vii.56	4·8	1·5	0·75
„	24. ix.56	3·1	—	—
„	23. x.56	5·0	5·3	1·4
„	30. xi.56	8·2	0·27	0·12
Site 24: *Narrandera, N.S.W.*				
Sheep	31. vii.56	5·3	0·8	0·3
„	30.viii.56	4·2	0·4	0·06
„	3. x.56	3·7	—	—
„	29. x.56	5·4	3·8	1·2
„	4. xii.56	6·3	0·17	0·03
Site 25: *Hay, N.S.W.*				
Cattle	16. vii.56	3·2	0·92	0·04
Sheep	16. vii.56	—	1·01	0·17
Cattle	21. ix.56	5·1	—	—
Sheep	21. ix.56	6·6	—	—
Cattle	23. x.56	5·6	22·0	0·55
Sheep	23. x.56	—	17·5	3·8
Cattle	27. xi.56	5·0	3·4	0·2
Sheep	27. xi.56	4·3	0·50	0·12
Site 26: *Monkira Station, Qld.*				
Cattle	21. vii.56	—	20·7	1·22
„	19.viii.56	—	4·04	0·16
„	29. ix.56	3·3	4·5	0·26
„	9. x.56	3·3	830·0	37·0
„	12. xi.56	4·7	353·0	16·1

TABLE 1 (*Continued*)

Animal	Date of Slaughter	I in Thyroid (mg I/g dry wt.)	Radioactivity of Thyroid at Time of Slaughter	
			mμc ^{131}I Total	mμc ^{131}I per Gram Tissue
Site a: Animals Slaughtered at Adelaide Abattoirs				
Cattle	15. v.56	5·1	—	—
„	30. v.56	4·8	2·3	0·115
„	18. vi.56	7·1	9·6	0·34
„	18. vi.56	—	2·2	0·04
Sheep	26. vi.56	4·2	—	—
Cattle	6. vii.56	5·1	16·6	0·64
„	9. vii.56	—	20·8	1·7
Sheep	11. vii.56	—	0·8	0·26
Cattle	11. vii.56	6·1	3·75	0·23
Sheep	11. vii.56	—	0·49	0·16
„	11. vii.56	—	0·63	0·21
„	11. vii.56	—	0·60	0·4
Cattle	11. vii.56	—	9·1	0·35
Sheep	18. vii.56	—	0·79	0·13
Cattle	2.viii.56	4·9	10·4	0·3
„	2.viii.56	—	6·2	0·3
Sheep	2.viii.56	—	2·9	0·4
„	2.viii.56	—	2·4	0·5
„	8.viii.56	—	1·15	0·6
„	8.viii.56	—	0·8	0·4
Cattle	13.viii.56	4·7	1·07	0·04
„	11. ix.56	4·4	—	—
„	27. ix.56	6·3	0·46	0·02
„	23. xi.56	4·3	11·8	0·76
„	23. xi.56	7·0	11·9	0·63
„	28. xi.56	5·4	4·0	0·20
„	28. xi.56	7·0	5·4	0·27
Glenthorne, Adelaide				
Sheep	26. vi.56	—	—	—
„	28. vi.56	—	—	—
„	5. vii.56	—	—	—
„	12. vii.56	—	—	—
„	17. x.56	—	43·0	8·4
„	18. x.56	—	33·0	11·0
„	19. x.56	—	32·0	9·6
„	20. x.56	—	36·0	11·0
„	22. x.56	—	20·0	8·3
„	24. x.56	5·7	29·5	8·4
„	26. x.56	—	23·0	8·3
Glenthorne, Adelaide (Continued)				
Sheep	29. x.56	—	15·5	5·4
„	31. x.56	4·4	24·0	5·5
„	2. xi.56	—	15·0	4·9
„	2. xi.56	—	17·5	5·7
„	5. xi.56	—	12·3	3·2
„	8. xi.56	—	2·3	1·1
„	12. xi.56	—	4·1	1·3
„	15. xi.56	—	2·5	0·88
„	19. xi.56	—	4·2	0·93
„	22. xi.56	—	2·4	0·48
„	26. xi.56	—	1·15	0·43
„	29. xi.56	—	0·4	0·14
„	3. xii.56	—	0·91	0·15
„	10. xii.56	—	0·34	0·11
Roseworthy, Adelaide				
Sheep	30. x.56	—	19·8	6·6
„	30. x.56	—	12·0	5·5
„	30. x.56	—	19·4	8·0
„	30. x.56	—	11·0	5·1
„	30. x.56	—	18·0	3·9
„	5. xi.56	—	6·3	3·0
„	13. xi.56	—	5·3	1·8
„	13. xi.56	—	4·7	1·5
„	13. xi.56	—	4·0	1·1
„	13. xi.56	—	6·6	1·9
„	13. xi.56	—	5·9	1·3
„	13. xi.56	—	4·4	1·3
„	13. xi.56	—	2·8	0·9
„	13. xi.56	—	5·9	1·9
„	19. xi.56	—	3·2	0·73
„	19. xi.56	—	4·8	1·50
„	19. xi.56	—	2·2	0·85
„	19. xi.56	—	2·15	0·90
„	19. xi.56	—	2·6	0·96
„	27. xi.56	9·3	1·52	0·53
„	27. xi.56	—	0·92	0·40
„	6. xii.56	—	0·6	0·14
„	6. xii.56	—	0·6	0·12
„	6. xii.56	—	0·42	0·085
„	6. xii.56	—	0·44	0·12

miles away—from the weapon test site, cf. Table 1. The subsequent effects of this explosion could be detected in the thyroids of grazing animals depastured within a band of terrain about a thousand miles wide, stretching west to east across the Australian continent. The variability observed within this band emphasized the uneven distribution of the deposited radioactive debris.

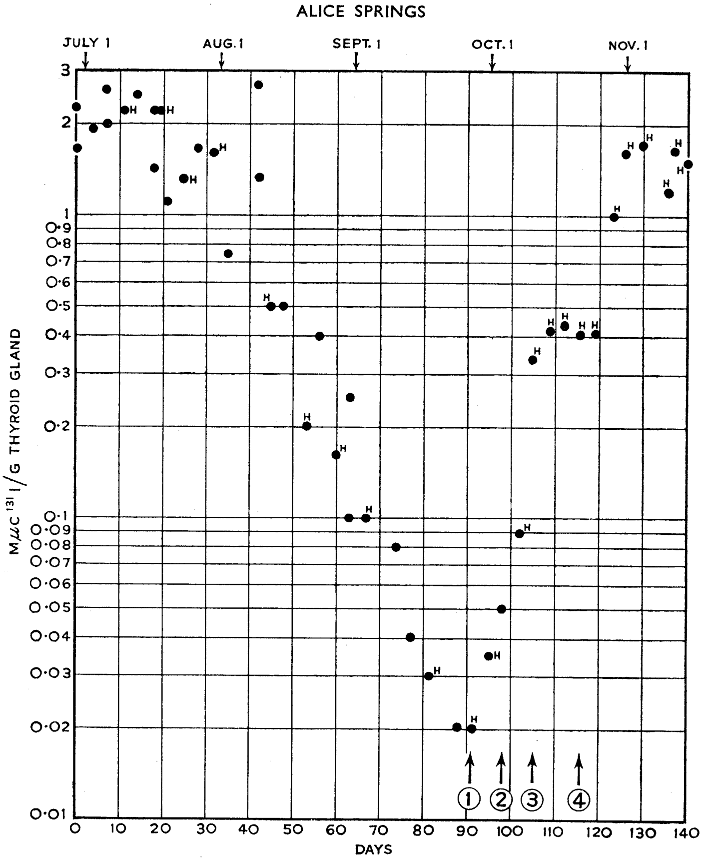

Fig. 2.—The ^{131}I concentrations of thyroid glands collected from cattle grazed in the vicinity of Alice Springs are set out in relation to the dates of the atomic explosions from which the debris containing the radioactive iodine was derived. The activity at the beginning of the observations reported here was the result of contamination originating on June 19, 1956, at Monte Bello. The accumulation in the thyroids of cattle grazed in the region of Alice Springs was about one-tenth of that observed at this period c. 1200 miles east at the same latitude, cf. Table 1. The dates of the four subsequent tests at Maralinga are indicated by serial numbers. Data marked H are from cattle grazed at Hamilton Downs Station.

Areas on the north-eastern seaboard and in central western Queensland that received the relatively heavy dressings from the Monte Bello tests are within the sector over which prevailing winds tended to carry the plumes from tests conducted later at Maralinga. In these latter areas the observations, Table 1, indicate repeated dressings from subsequent weapon tests.

The Alice Springs Series

Throughout the period extending from June 28 to mid-December 1956, thyroid glands were taken at intervals of *c.* 3 days from cattle that had grazed within the vicinity of Alice Springs. The data from this frequent sampling, set out in Figure 2, allow certain generalizations to be drawn.

Although the concentrations of ^{131}I observed in the thyroids of cattle at this site were never particularly high if compared with those found in thyroids taken from cattle grazed during this period in the same latitudes 700–1000 miles east, a noteworthy feature of the series is that after the second Monte Bello explosion, the concentrations of ^{131}I in the thyroid glands remained, within the expected limits of variance, relatively constant for a period of about 40 days before beginning to decrease exponentially.*

The accumulation and persistence of ^{131}I within the thyroids of grazing animals are influenced by a number of variables. In this instance, losses of ^{131}I due to metabolic turnover of iodine in the thyroid and to the radioactive decay of the ^{131}I deposited in the gland were made good over a period of about five half-lives of ^{131}I by additional ^{131}I assimilated from the fodder, which was itself a decaying source. The concentration of ^{131}I on the fodder plants would be determined during this interval by radioactive decay of ^{131}I to stable ^{131}Xe and, to some degree in the very early stages, by renewal from the precursor ^{131}Te. Neither wind and rain, which would remove, in part, adherent particles of radioactive debris from the fodder plants, nor the growth of the fodder plants themselves, which would, in effect, dilute the radioactive contamination of the material taken by the grazing animals, exerted any very considerable influence there during the 40 days subsequent to this precipitation. This is in distinct contrast to what happened in other circumstances in the vicinity of Adelaide (cf. Fig. 5).

The effects of contamination of the Alice Springs area by debris from the first, second, and fourth Maralinga explosions, clearly discernible in the data, express the fact that the surrounding country had been subject to at least four dressings of radioactive material from atomic explosions. The hazards of successive contaminations are cumulative.

The Adelaide Fall-out

We, in this laboratory, have been interested for some time in the radon (^{222}Rn) and thoron (^{220}Rn) content of soil atmospheres, and in the radioactivity of the air which in greater part is due to daughter products† of these emanations from the rocks and soils (Stout, Jones, and Delwiche 1957). Periodic series of estimations of radioactivity of Adelaide air that were made were not a continuous routine, but on no occasion before October 13, 1956, did chemical separations and measurements of decay characteristics of the radioactive material collected in the course of these

* The kinetics responsible for this apparent pause in the decay of the ^{131}I in the thyroids is discussed in the legend to Figure 5.

† The radioactivity of the air in Adelaide is made up mainly of ^{212}Pb, ^{214}Pb, and their degradation products, which adhere to dust particles that may be collected by drawing air through a filter paper (Stout *et al.* 1957).

studies suggest any considerable contamination from isotopes derived from explosions of nuclear weapons. However, the very large increase in the amount of radioactivity in the 24-hr catch (October 12–13) from the filtration of 20 cu. m. of air clearly indicated that the plume from the third Maralinga explosion (October 11) passed close to Adelaide and contaminated the city and surrounding country with radioactive fission products. The radioactivities of the particulate matter caught on the filter-paper disks during the week October 12–18 are set out in Table 2.

TABLE 2

ADELAIDE FALL-OUT FROM THE THIRD MARALINGA EXPLOSION

Radioactivity in tidal air (20 cu. m.)

Collection Period October 1956	Prevailing Wind	Rain (in.)	Activity on Filter Pad* (counts/100 sec)
12–13	N.-NW.	Nil	95,000
13–14	N.-NW.	Nil	11,200
14–15	NW.	0·29	1,030
15–16	SW.	Nil	3,850
16–17	SW.	0·09	350
17–18	SW.	0·36	21
19–20	SW.	Nil	62

* Collected by drawing *c.* 20 cu. m. air through a $\frac{7}{8}$-in. disk of filter paper at a steady rate over 24 hr. Radioactivity was estimated immediately after completing the collection by placing the disk 1 mm from the end window of an E.M.H.2 tube of which the counting efficiency for ^{40}K standard under similar conditions was $\leqslant 15$ per cent. The tidal air is the volume of air taken into and expelled from the lungs in the normal course of breathing. A 70 kg man breathes *c.* 20 cubic metres a day.

The greater part of the precipitation occurred on two rainless days; on the third day, October 15, about 0·3 in. of rain fell in gusty showers. The radioactivity of this rain was not determined.

This occasion provided an opportunity to study more intensively the effects of a precipitation of radioactive debris, for the Division's field station, Glenthorne, a square mile of country situated 11 miles south of Adelaide, was in the contaminated area.

The thyroid gland removed on October 17 from a sheep that had been grazing as a member of a flock depastured there contained 22 mμc ^{131}I. Prior to October 12, the thyroid glands of individuals of this flock contained <5 $\mu\mu$c ^{131}I which was the lower limit of our means of estimation. Observations started a day or so later have provided data particularly cogent to the assessment of the hazard from fall-out.

The capacity of grazing animals to concentrate radioactive isotopes from the contaminated pastures was assessed by measurements of the radioactivity in the rumen contents of sheep drawn from the flock at intervals of 2 days; the rise and fall of radioactivity in various organs and tissues was studied in these animals over the ensuing 2 months; and, later, observations were made at less frequent intervals.

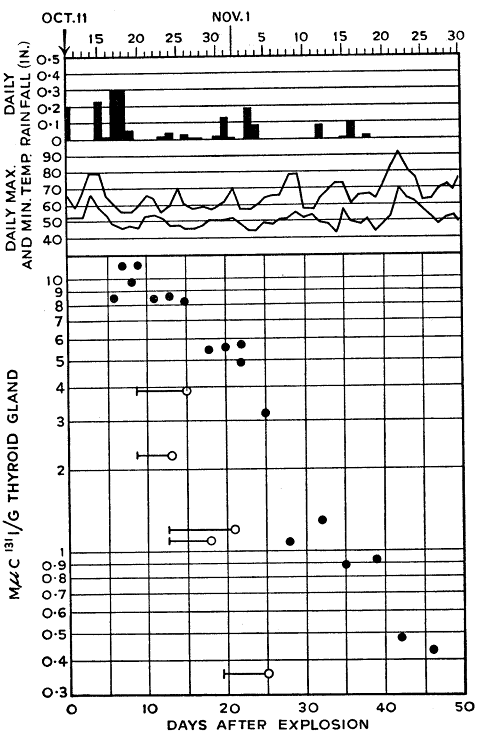

Fig. 3

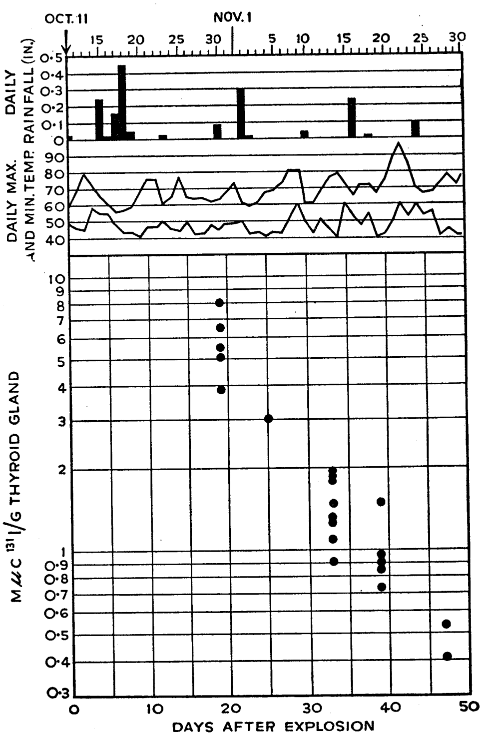

Fig. 4

Fig. 3.—The ^{131}I contents of the thyroid tissues of sheep of similar age grazed as a flock at Glenthorne, 11 miles south of Adelaide, are set out in relation to the time subsequent to the explosion on October 11, 1956, at Maralinga, *c.* 600 miles distant, from which the radioactive material that contaminated the pastures originated. The ^{131}I concentrations represented by open circles are those found in sheep transferred from pens to the contaminated pastures on October 19 (2), on October 23 (2), and October 30 (1) and allowed to graze there with the flock until slaughtered on the days indicated.

Fig. 4.—The ^{131}I contents of the thyroid tissues of sheep grazed as a flock at Roseworthy Agricultural College, 35 miles north of Adelaide, are set out in relation to the time subsequent to the explosion on October 11, 1956, at Maralinga (cf. Fig. 3). From analysis of variance of the findings on October 30, November 13, November 19, and December 6 (data of final date not shown in figure), the mean logs of the concentrations of ^{131}I (mμc/g) and their standard deviations on these dates were, respectively, $0\cdot7523\pm0\cdot0523$, $0\cdot1480\pm0\cdot0442$, $-0\cdot0189\pm0\cdot0523$, and $-0\cdot9415\pm0\cdot0585$.

Examination of rumen contents taken from animals slaughtered on October 18—the first of this series—indicated that a grazing sheep collected from the Glenthorne pastures on that day sufficient radioactive material to register close to 10^4 decompositions per second.* Radiometrical examination of this material proved

* Estimated by measuring rumen samples with an end-window (E.M.H.2) tube. The rumen contents weighed 950 g dry wt.

its identity with that of the "catch" filtered from the atmosphere in Adelaide on October 12–13. The decay of the radioactivity in the rumen contents of animals slaughtered subsequently proceeded at a rate similar to the rate of decay of the catch. The radiometrical data leave no reasonable doubt that, throughout this series of observations at least, the material from which the ^{131}I was derived was the same as the complex mixture of radioactive isotopes separated from the atmosphere.

Previously, we had obtained thyroid glands of cats, dogs, and rats that had lived in areas contaminated with fall-out, and had found very little or no ^{131}I in them at a time when the concentration of ^{131}I in the thyroids of herbivora grazing in the vicinity was high. The tentative conclusion from these observations was confirmed on October 18 *et seq.* by measurements of the radioactivity of thyroid glands from sheep drawn from a group that for some months previously had been confined in pens and fed on rations comprised essentially of chaffed cereal hay that, perchance, had been stored under conditions which would render contamination by fall-out very unlikely. These glands contained very little ^{131}I indeed—less than 0·05 per cent. of the ^{131}I found in glands from sheep of similar age that had been grazing on pastures in the immediate vicinity of the pens. The following protocol is typical of the findings:

Adelaide Fall-out: ^{131}I in Thyroids of Pen-fed and Pasture-fed Sheep

Location.—Glenthorne, the central field station of the Division of Biochemistry and General Nutrition, C.S.I.R.O., situated 11 miles south of Adelaide.

Previous History of Animals.—The animals were wether hoggets selected, at random, respectively from pen-fed and pasture-fed groups. They were closely related and of similar age (*aet.* 17 months). One, A5–071, was from a group that for some months previously had been confined to a suite of roofed pens, enclosed on three sides, and freely open to the east. During this period these wethers had been fed on dry rations (comprised essentially of cereal hay produced on the property) provided in covered food troughs, and they were watered from troughs situated under the roofed part of the pens. The other, A5–391, was one of the flock from which the pen-fed group had been drawn. This flock was grazed on mixed pastures in the immediate vicinity of the pens. It was the height of spring; the pastures were particularly lush and growing rapidly.

On the morning of October 18 (i.e. 6 days after Adelaide and its environs had been contaminated with fall-out from the third Maralinga explosion) the animals were conveyed to the laboratory, slaughtered, and the ^{131}I in their thyroids determined.

Estimation of ^{131}I.—Time 11·00–12 noon, October 18, 1956, Adelaide. Instrument calibration: discriminator bias 20 V $\equiv$ 0·364 MeV peak of ^{131}I γ-emission; channel width 1 V; E.H.T. 950 V; rise 0·03 μsec; fall 3 μsec; atten. 30 dB, at which setting background was 16·6 $\pm$ 1·8 counts/100 sec, and ^{131}I std. = 272 $\times$ 10^3 counts/100 sec/μc ^{131}I.

	Sheep No. A5–071 (pen-fed)	Sheep No. A5–391 (pasture-fed)
Weight of thyroid (g wet wt.)	2·8	2·9
Net counts/100 sec	7$\pm$3·2	9065$\pm$18
Total ^{131}I in gland (mμc)	*c.* 0·025	33·0
^{131}I/g thyroid (mμc)	$<$0·01	11·0

The decay characteristics and the energy spectrum of the γ-radiation of the gland from A5–391 were those of ^{131}I.

Thus it is probable that only a negligible proportion of the ^{131}I observed in the thyroid glands of cattle and sheep grazed on the contaminated areas considered in this report found its way into the animals via their lungs, and so, provided the age

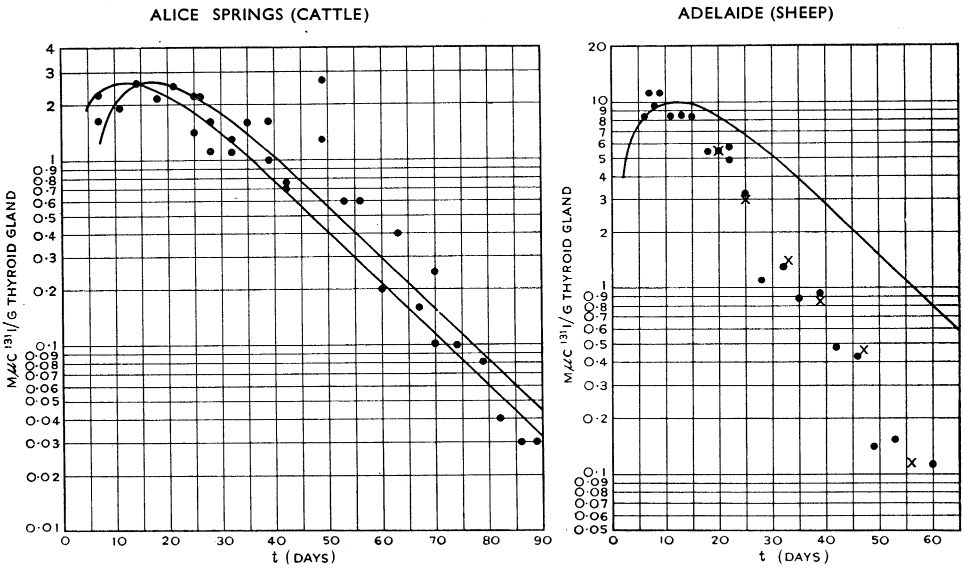

Fig. 5 (*a*) Fig. 5 (*b*)

Fig. 5.—The amount, x_0, of ^{131}I deposited at $t = 0$ on the fodder consumed in a day by a grazing animal becomes $x_0 e^{-\lambda t}$ units at time t, where λ is the ^{131}I decay constant, 0·0866; and so, if the animal's daily intake of contaminated fodder, and the efficiency of retention of ^{131}I are constant, and the biological turnover of ^{131}I is ignored, the rate of accumulation of ^{131}I in the animal's thyroid, dx/dt, at time t is proportional to

$$x_0 e^{-\lambda t} - \lambda x,$$

where

$$x = x_0 t e^{-\lambda t}.$$

This relationship expresses the extreme case in which the original concentration of fall-out on the fodder is not reduced by weathering, etc.

(*a*) Within an expected variance, the observed data of the Alice Springs series fit tolerably well the curve derived from the above relationship. To allow time for the troposphere cloud to travel from the weapon test site to the area where the observations were made, t_0 on the scale has been advanced 2 days from June 19, 1956. Zero of the second curve has been advanced (arbitrarily) a further 5 days. No rain fell in this area between July 16, 1956 (day 25) and September 2, 1956 (day 73).

(*b*) The concentrations of ^{131}I observed in the Adelaide series diminished with time much more rapidly than those implied by the curve derived from the above postulates. In this instance there were heavy falls of rain (cf. Figs. 3 and 4) and considerable growth of the pastures during the period in which the observations were made. Individual observations at Glenthorne (●) and means of observations at Roseworthy (×).

of the fall-out is known, the concentration of ^{131}I which accumulates in the thyroid gland is an expression of the rate at which the grazing animal gathers radioactive material from contaminated pastures.

The concentrations of ^{131}I in the thyroid glands of sheep grazed at Glenthorne (11 miles south of Adelaide) and at Roseworthy College (35 miles north of Adelaide) are set out respectively in Figures 3 and 4. Further details are reported in the legends to these figures.

Conditions at Glenthorne allowed an estimate of the rate of decrease of the ^{131}I concentrations of the pasture to be made by periodically transferring sheep from pens to the pastures and allowing them to graze there for intervals of 4–7 days before they were slaughtered. Conditions at Roseworthy, where one day each week a number of animals was slaughtered to provision the college, allowed an estimate to be made of the variance of the ^{131}I content of the thyroids of animals grazing as a flock under identical conditions. Similarity of the two series indicated that these pastures situated about 40 miles apart had been contaminated to an almost identical degree.

In contrast to the considerable period during which the ^{131}I concentration in glands from cattle grazed in the vicinity of Alice Springs remained nearly stationary (cf. Figs. 2, 5(*a*)) the concentration of ^{131}I in the glands of sheep subsequent to the Adelaide fall-out began almost at once to decrease exponentially at a much more rapid rate than would be implied if the intake of the radioactive fall-out remained approximately constant (cf. Fig. 5(*b*)). The pastures in the environs of Adelaide were contaminated at the height of spring when the growth of the fodder plants was itself rapid enough to, in effect, decrease materially the overall concentration of the radioactive particles that had been deposited on them. Heavy showers of rain which fell from time to time during the early stages of the observations no doubt decreased the concentration further by washing from the pastures the adherent radioactive particles of which ^{131}I was an integral part. Dilutions arising from these and possibly from other causes reduced the rate of intake of ^{131}I to an extent that was insufficient to make good radioactive decay and biological losses.

The situation observed at Alice Springs after the Monte Bello exercises (Fig. 5(*a*)), is representative of more serious conditions than those which prevailed in the environs of Adelaide after the third Maralinga explosion (Fig. 5(*b*)), for a sustained ^{131}I concentration in the thyroid under these conditions is an indication that radioactive debris remains on the pastures for a considerable period before it is in part removed to the soil from which intake is slower.

III. Discussion

The observations set out in the body of this paper emphasize the speed with which grazing animals assimilate and concentrate ^{131}I from constituents of the fission products that become deposited on pasture in areas traversed by the clouds of debris arising from atomic explosions. In these circumstances it may reasonably be assumed that a rapid accumulation of ^{131}I in the thyroids of grazing cattle indicates a rapid gathering of ^{89}Sr, ^{90}Sr, and of other bone-seeking isotopes, and a speedy launching of these radioactive substances, via milk, into human foodstuffs, thence to the skeleton where they become deposited preferentially at the sites where mineralization is proceeding. As the intensity of radiation from the relatively concentrated aggregates of radioactive material that are formed in this way is probably the prime determinant in the induction of the changes within the cells of the bone matrix and of the bone marrow that lead ultimately to osteosarcoma or to leukaemia, the hazard of neoplasm production would be influenced to an important degree by the rate at which these radioactive isotopes are ingested, for this determines the speed with which they are presented to the site of osteogenesis* and so the degree of their aggregation.

The hazard would be greatest during the periods in which the radioactive debris remains on the pastures, for then both the rate of ingestion and the ratios of ^{89}Sr, ^{90}Sr, etc. to calcium within the pastures are greatest and so the rate of transfer to human beings is at its peak.

In addition to the biological influences that tend to concentrate ^{90}Sr within human foodstuffs, particularly within milk, there are purely physical influences that tend to increase the ratio of ^{90}Sr to other radioactive constituents in the debris that moves off as a cloud from the site of test explosions. ^{90}Sr comprises only about 0·1 per cent. of the original mass 90 yield of primary fission products; and its immediate precursor, ^{90}Rb (half-life 2·7 min) only about 15 per cent. (Steinberg and Glendenin 1955; Martell 1956). The parent, ^{90}Kr (half-life 33 sec), a noble gas, is produced in high independent yield† and is sufficiently long-lived and chemically unreactive to ensure that the greater part of its daughters, ^{90}Rb and ^{90}Sr, escape complex formation with vaporized silica in the fire-ball of ground level explosions: and so, after the larger particles condense from the cooling gases and shower close to the site of the explosion, the cloud of finer debris that moves off contains a considerably higher proportion of ^{90}Sr (Martell 1956) in chemical states readily assimilable by living organisms.‡ For this reason an enrichment factor should be introduced into calculations that aim to assess the overall ingestion of ^{90}Sr by grazing animals from the ^{131}I found in their thyroid glands.

In the fission products at the time of the explosion, the atomic ratios§ of potential $^{90}Sr/^{131}I$, of potential $^{89}Sr/^{131}I$, and of potential $^{89}Sr+^{90}Sr/^{131}I$ are *c.* 1·6, 2·0, and 3·6 respectively, and these ratios increase with time according to

* As the process of osteogenesis is particularly intensified in the very young, the risk is greater in foetal and neo-natal subjects.

† Reed (1955) reported a yield of ^{90}Sr, which, based on a half-life of 28 years, corresponds to a 5·6 per cent. yield of this isotope in the fission products from ^{235}U: thus 146 c of ^{90}Sr are produced per 10^{12} kcal (1 kilo-ton T.N.T.) of energy released. The yield of ^{90}Sr from fission of ^{238}U and of ^{239}Pu would be less.

‡ Some figures have been published which, indicate the extent of the error this enrichment introduces into predictions of the quantity of ^{90}Sr based on overall radioactivity of fall-out. At Binghampton, N.Y., for instance, the amount of ^{90}Sr separated chemically from the soil was *c.* four times that predicted from radiometric measurement of fall-out collected on sticky paper. Similar observations on other areas indicated frequently an enrichment factor of *c.* 2 (Eisenbud and Harley 1956).

§ Fission of ^{235}U induced by thermal neutrons yields 4·8 per cent. of ^{89}Sr (half-life 53 days) (Reed and Turkevich 1953), 5·8 per cent. of ^{90}Sr (half-life 28 years) (Glendenin *et al.* 1956), and 2·9 per cent. of ^{131}I (half-life 8·1 days) (Coryell and Sugarman 1951). Published data relating to fast neutron-induced fission is not so complete. The ratios mentioned in the text would not be altered materially in the fission products of ^{233}U or of ^{239}Pu.

the age of the fission products and the rates of decay of the respective isotopes. Thus for a period after pastures have been contaminated by fission products of known age, an approximation of the extent of the ingestion of ^{89}Sr, ^{90}Sr, and of other bone-seeking isotopes by a grazing animal might be derived directly from the ^{131}I content of its thyroid gland, provided the efficiency of retention of ingested ^{131}I and the rate of physiological turnover of iodine by the particular species of animal employed are known. The period of usefulness of this means of assessing the intake of the constituents of fall-out that determine the primary hazard would be limited by the rate of decay of ^{131}I to approximately 3 months after the explosion from which the fission products originated.

IV. Acknowledgments

It is a pleasure to record my thanks to my colleagues, Mr. G. B. Jones and Mr. V. A. Stephen, for their assistance with the estimations and care of the instruments, and to Mr. A. Packham who discreetly arranged for the collection of the thyroids. To our many friends who so readily and without question collected and despatched the glands to this laboratory we are all deeply grateful.

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The instruments—a 1-in. diameter well-crystal scintillation counter together with amplifying, analysing, scaling, and timing units—were provided on loan for the occasion by the Atomic Weapons Research Establishment.

V. References

Coryell, C. D., and Sugarman, N., Eds. (1951).—"Radiochemical Studies: The Fission Products." Div. IV. Vol. 9. Appendix B. (McGraw-Hill Book Co.: New York.)

Eisenbud, M., and Harley, J. H. (1956).—*Science* **124**: 251.

Glendenin, L. E., Steinberg, E. P., Flynn, K. F., Hayden, R. J., and Inghram, M. G. (1956).—See Steinberg, E. P., and Glendenin, L. E. (1955).—Geneva Conference Paper 7/P/614, U.S.A.

Gunther, R. L., and Jones, H. B. (1954).—U.S. Atomic Energy Commission Document No. UCRL–2689.

Martell, E. A. (1956).—AECU–3262. (Enrico Fermi Inst. for Nuclear Studies: University of Chicago.)

Medical Research Council (1956).—"The Hazards to Man of Nuclear and Allied Radiations." (H.M.S.O.: London.)

Reed, G. W. (1955).—*Phys. Rev.* **98**: 1327.

Reed, G. W., and Turkevich, A. (1953).—*Phys. Rev.* **92**: 1473.

Steinberg, E. P., and Glendenin, L. E. (1955).—Geneva Conference Paper 7/P/614, U.S.A.

Stout, P. R., Jones, G. B., and Delwiche, C. C. (1957).—A Conference on Radioactive Isotopes in Agriculture. U.S. Atomic Energy Commission Report No. TID–7512. pp. 323–35. (U.S. Govt. Printing Office: Washington, D.C.)

Van Middlesworth, L. (1954).—*Nucleonics* **12**: 56.

Van Middlesworth, L. (1956).—*Science* **123**: 982.