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TRANSFER OF RADIONUCLIDES TO ANIMALS - AN HISTORICAL PERSPECTIVE OF WORK DONE IN THE UNITED STATES

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INTRODUCTION

Good afternoon ladies and gentlemen. It is my pleasure to meet with you today at this most historic setting as a participant in this important workshop. I was honored to be asked to contribute both as a speaker and as a member of the program committee.

I would like to start by telling an interesting story about the first recorded tracer study in man (1). It is fitting because the story takes place here in England and the main character is none other than George Hevesy who was the recipient of a Nobel prize in 1943 for his pioneering work in the application of radionuclides as biological tracers. He was awarded the Atoms for Peace Award in 1959.

"George Hevesy, a young Hungarian physicist working in England, was the first scientist to use an isotopic tracer in biology. In 1923 he immersed the roots of bean plants in solutions containing a radioactive isotope of lead. He traced the ascent of the lead into the stem and leaves by simply measuring their radioactivity. According to Dr. Hevesy, his first tracer experiment was designed for a less exalted purpose. Since he was suffering from the endemic affliction of young scientists of that period, impecuniousness, he was lodging in a very modest boarding house. He observed that roast beef was invariably followed by hash on the weekly menu. He had strong suspicions that even the meager leavings of the roast beef from the plates of the boarders greeted them again the following day disguised as hash. Therefore he once sprinkled a trace of radioactivity on his leavings from a meal and the next day he surreptitiously carried off a sample of hash to the laboratory. His instruments confirmed his suspicions: the hash was radioactive." The connection of this story to the workshop topic may not be too far fetched if one thinks of the uptake of the tracer by a live cow rather than by the roast beef.

Hevesy, a Hungarian who was a truly international scientist, worked in Manchester, England with Rutherford from 1912 to 1920. For the following 6 years he worked with Neils Bohr in Copenhagen. After he received the Nobel prize he was offered, and accepted, Swedish citizenship.

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As part of the preparation for this paper I had a literature search done by the Information Resources Organization at ORNL. The search parameters excluded man and laboratory animals. Included were the search terms farm animals, radionuclides, metabolism etc. However, many of the retrieved references dealt with investigations of physiological or metabolic parameters using radionuclides that are not of interest to this workshop. Some of these, for example, were related to the use of radionuclides in nuclear medicine. I was also impressed with the large fraction of the usable references that came from laboratories outside of the United States.

Included in the listing are translations of work done in the Soviet Union. It was also clear that most of the contributions came from only several laboratories and that much of the work was done quite a few years ago. It remains to be seen what the impact of the Chernobyl nuclear power station accident will be on these studies.

One lesson learned from preparing this paper is that the amount of effort devoted to this area of research over the years has been surprisingly small. This is especially true if one compares the total effort with research utilizing radionuclides to study metabolism in laboratory animals.

The search produced several hundred references containing much information on radionuclides and animals. Careful review and verification of the listing need to be completed. This report will not be a comprehensive review of the listing. The purpose of this report is to cover the subject from a historical perspective.

I was also struck by the large number of non-classified internal reports as compared to the number of open literature publications for work done in the U.S. The reason is not clear. Perhaps there were limited numbers of professional journals in which to publish these kinds of data or perhaps the sponsors of the work were more inclined to have the material appear in internal reports rather than open literature publications. Many of the very early studies appeared in the Journal of Dairy Science. For reference, the Radiation Research and Health Physics journals were started in 1954 and 1958, respectively.

In the mid 1960's, there were five veterinarians assigned to the Division of Biology and Medicine within the U.S. Atomic Energy Commission (AEC). Four were on assignment from the military establishment and one was a civilian. There are none at present and, in fact, there is only one person on staff in the U.S. Department of Energy (progeny organization of the AEC). AEC-supported animal studies in the early 1960's included more than animal metabolism studies e.g.,

agricultural research. Also, in the mid 1960's there was a major change in program emphasis within the AEC. Basic cell biology studies were being developed at the expense of work done in animals. In the latter category, laboratory research had a higher priority than did work with farm animals. Another major change in program emphasis is happening now with the increasing emphasis on work on the Human Genome Program.

WHY IS THIS WORKSHOP OF INTEREST?

Nuclear fuel cycle

Concern for radioactive releases from the nuclear fuel cycle has provided the incentive for many studies of interest to this workshop. There were early concerns about the uptake of radionuclides by human beings eating fish caught in the Columbia river near the Hanford site. In some cases the information on the uptake of materials such as ^{65}Zn by fish and then man could be verified by whole body counting techniques. Large reactor operations and fuel reprocessing operations took place at the Hanford site.

Studies were also undertaken at other DOE facilities such as Savannah River, Rocky Flats, Idaho Falls, and Oak Ridge.

The development of commercial nuclear power also required data and models that predicted uptake of radionuclides by people living in the vicinity of the facility. The interests were mainly in fission products and, to a lesser extent, neutron activation products. These models will not be discussed in this presentation.

In more recent years the emphasis has shifted to the possibility of contamination of the general public from radioactive sources associated with disposal of nuclear or mixed (chemical and nuclear) wastes.

Chernobyl

The accident at the nuclear power station at Chernobyl in the Soviet Union created a large and almost instant demand for information. Governments around the world were extremely concerned for the health and safety of its citizens both from direct effects of atmospheric fallout and from the ingestion of plant and animal products containing radioactive materials.

There was also an economic factor in these considerations and, in retrospect, some decisions were made under the pressures of the time that are now viewed as being unsound. There was an urgent need for information of the kind being discussed at this workshop. In some cases, generic information was used to advantage; in others, lessons were learned too late to be of help or by trial and error. For

example, it came as a surprise to many that the levels of cesium in the milk of goats and sheep were higher than those of cows from the same general location. Also a surprise to some was the high level of contamination in reindeer. These situations were predictable because of information collected decades ago from animal studies designed to understand the uptake, distribution, retention and loss of radionuclides originating in nuclear-weapons fallout (2). Neither should it have been surprising to find high levels of contamination in mushrooms or in animals, including man, that eat them (3).

A recent report from the Oak Ridge National Laboratory (4) indicates the large uncertainties that exist when one attempts to estimate the amount of radiocesium from Chernobyl that would find its way to man. Many parameters are required for the many models used to move the material from source, through the environment, including animals that are eaten as food, to man. The data indicate that the amount actually detected in people was between one and two orders of magnitude lower than that predicted by the models. Much is known about cesium because of its presence in nuclear fallout and its ease of measurement in man. One can only wonder how good the data are for other radionuclides.

SPACE APPLICATIONS OF RADIOISOTOPES

Radioisotope Thermoelectric Generators (RTG)

Both the U.S. and the Soviet Union use radioisotope thermoelectric generators (RTG) as power sources for space systems. The fuel currently used by the U.S. is mainly ^{238}Pu with about 20% ^{239}Pu by activity. Shortly after launch in April 1964, a U.S. satellite, SNAP 9A, re-entered the atmosphere and burned up at high altitude over the Mozambique channel. The radioactive inventory, 6.3×10^{14} Bq (17 kCi) of ^{238}Pu , reentered primarily through the southern hemisphere. Subsequently, ^{238}Pu was measured for many years in air samples and on the ground. This large input of ^{238}Pu greatly perturbed the global $^{238}\text{Pu}/^{239}\text{Pu}$ ratios resulting from nuclear weapons testing. In addition, 4.8×10^{11} Bq (13 Ci) of ^{239}Pu from SNAP 9A were part of the source term.

Other accidents have occurred with no significant releases of radioactivity. However, one U.S. manned mission to the moon, Apollo 13, experienced a failure mode that resulted in the RTG being returned to Earth. The 1.7×10^{15} Bq (44.5 kCi) ^{238}Pu inventory attached to the lunar landing vehicle ended up near the Tonga Trench, as planned, in the Pacific Ocean. The power source also contained 1.2×10^{12} Bq (33 Ci) ^{239}Pu (5).

There were plans to launch several large RTG's in 1986, just after the U.S. space shuttle Challenger accident. These missions, Ulysses and Galileo, were scheduled

to use three RTGs containing almost 1.5×10^{16} Bq (400 kCi) ^{238}Pu ; two RTG's containing a total of 1×10^{16} Bq (265 kCi) aboard Galileo and a single RTG on Ulysses. These missions are being critically re-evaluated at present.

Cosmos nuclear power reactors

A 14 May, 1988 article in the Knoxville News-Sentinel (Tennessee) reported that the satellite Kosmos 1900 had been in a steadily decaying orbit for a month and, if no orbital corrections were made, the satellite would enter Earth's atmosphere (6). Kosmos 1900's orbit covers the Earth between 65 degrees south and north latitude excluding Antarctica in the south and a swath in the north bounded by the inhabited areas of Alaska, Greenland, Siberia and Scandinavia. The concern expressed in the article was that the reentering satellite would "shower the Earth with radioactive debris". Kosmos 1900 is a RORSAT, an English-language designation which stands for Radar Ocean Reconnaissance Satellite. The Soviet Union launched Kosmos 1900 on 12 December, 1987 and lost radio contact with the craft in April. They expect the satellite to reenter in August or September. An 8 August 1988 article in the Knoxville News-Sentinel points out that the U.S. Federal officials have begun contingency planning in case Kosmos 1900 reenters and impacts on U.S. territory (7). The Federal Emergency Management Agency is coordinating the planning.

Two other satellites in the Kosmos RORSAT series have fallen from orbit. Kosmos 954, powered by 50 kg (110 pounds) of Uranium, reentered the atmosphere in January 1978 and spread radioactive contamination across parts of Canada. On 23 January, 1983, Kosmos 1402 dropped from orbit over the Indian Ocean. The News-Sentinel article states that no radioactive debris is known to have fallen to Earth. Of interest is a report that appeared in Health Physics in 1984 (8) concerning the detection of uranium isotopes in the U.S. confirming the destruction of Kosmos 1402. Apparently the article meant earth in the sense of land rather than the planet Earth.

Current predictions are that the amount of radioactive materials used in space applications will continue to increase. A recent article in Time magazine points out that 7 countries (9) now have the capability to launch satellites. According to the article, Israel will soon become the eighth.* Most if not all these countries have the capability to use radionuclides as power sources in these satellites.

* Israel successfully launched the satellite shortly after the workshop.

EARLY WORK IN THE U.S.

Hanford, Washington

It is clear that much of the early work in the U.S. of interest to this workshop was done at the Hanford site in the state of Washington. A leader in this effort was Dr. Leo K. Bustad, a veterinarian, who had the foresight to recognize the importance and need for these kind of studies. Bustad also was a prime advocate for the use of the "mini pig" in animal studies using radionuclides (10). This allowed the researcher to use an animal larger than the dog or non-human primate in mass and one that had many characteristics, including body mass, in common with human subjects.

There is an old German saying that goes as follows:

dogs look up to man;
cats look down on him;
the pig looks him right in the eye.

The earliest research interests at Hanford involved radioisotopes of iodine. These materials, notably ^{131}I , were released from the facility at Hanford and were of interest because of its presence in nuclear weapons fallout. An example of early work published by this group was on the chronic administration of ^{131}I to sheep that appeared in 1957 (11). Another example is the 1963 paper on the thyroid uptake of ^{131}I by sheep following various routes of administration (12). In 1965, Shannon et al., reported on the public health aspects of Cs-137 in ruminants (13). Weeks et al., reported on the uptake of plutonium by pigs in 1956 (14).

There was also a major interest at Hanford in the early years in using a miniature goat for metabolic studies (15). Dr. Bustad was also heavily involved in these efforts. Another key contributor to work at Hanford was Dr. R. O. McClellan, also a veterinarian, became the director of the Inhalation Toxicology Research Institute in Albuquerque, New Mexico.

Strontium-90 was also studied in a variety of animals because it is a high-yield product of nuclear fission (16). It is also clear that some of the long term studies at Hanford were never completed because of guidance from the sponsoring agency. An example is the complete data analysis of the ^{90}Sr swine studies.

University of Tennessee - Atomic Energy Commission/Agricultural Research Laboratory (UT-AEC/ARL), Oak Ridge, TN

In 1948 the U.S. Atomic Energy Commission (AEC) and the University of Tennessee (UT) established at Oak Ridge, TN, a cooperative research program to study the effects of radiation from nuclear weapons fallout on a herd of Hereford cattle

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accidentally contaminated during the testing of the first atomic bomb detonation at Alamogordo, New Mexico in 1945. This cooperative program led to the establishment of the UT-AEC Agricultural Research Laboratory. The research program was rapidly expanded to include radioisotope studies and radiation effects of agricultural products. From its inception the laboratory has contributed to the understanding of the transport of radionuclides into mans food chain and to the biological effects of external radiation on plants and animals.

As an example, the following list illustrates the diverse research endeavors undertaken at that site:

- studies on the effects of irradiation of the central nervous system
- reproduction
- unilateral vs bilateral irradiation of swine
- late effects of burros surviving mixed radiations from atomic bombs
- radionuclide metabolism in beef cattle, sheep, swine and poultry
- embryo morphology and radiosensitivity
- mutations from irradiated seeds

Among the radionuclides studied were ^{32}P , ^{45}Ca , ^{65}Zn , ^{75}Se , ^{89}Sr , ^{90}Sr , ^{131}I , ^{134}Cs , ^{137}Cs , and ^{144}Ce - ^{144}Pr .

An example of an early publication from the Oak Ridge group is the 1953 publication by Hood and Comar on the metabolism of ^{137}Cs by mice and farm animals (17). Another, on ^{18}F in domestic animals, was reported in 1955 (18). Mraz and coworkers published information on Cs and K metabolism in sheen (19) and swine (20). Data on the uptake and excretion of Cs and K in lactating dairy cows was reported by Cragle in 1961 (21).

In 1973 the laboratory became known as the Comparative Animal Research Laboratory (CARL) and its primary concern was a program designed to estimate the risk to man from effluents of energy producing systems. Extrapolation of data from animal systems to man was of prime interest to researchers at CARL.

Niobium-95 metabolism in newborn and weanling swine and sheep was of interest to the Oak Ridge group. Some of these studies were reported by Mraz and Eisele in 1977 (22).

In more recent years some work was done with transuranic radionuclides, notably americium (23). The facilities are currently part of the Oak Ridge

Associated Universities (ORAU). The ORAU mission no longer includes research using animals and radionuclides.

Cornell University, Ithaca, New York

One of the key investigators at the Oak Ridge facility was Dr. Cyril Comar who contributed much to our understanding of the metabolism of radionuclides in animals. Comar left Oak Ridge to work at the New York State Veterinary College (Department of Physical Biology) at Cornell University where he and his colleagues continued work on the same subject. However, the Cornell program was expanded considerably beyond "animal metabolism" investigations.

The Cornell group under Comar remained active for many years. Wasserman et al., reported on the metabolic behaviour of Cs-137 and Ba-137m in lactating goats (24). Another key individual in this activity was Dr. Fred Lengemann who recently retired from Cornell. In 1968 he and his coworkers reported on predictions of Cs-137 intake from milk by human populations following a short term deposition event (25)

Colorado State University (CSU), Fort Collins, Colorado

Some important studies have been done in the Department of Animal Sciences at Colorado State University by Dr. G. Ward and colleagues. A report on the effects of feeding practices on Cs-137 levels in milk appeared in 1965 (26). In 1969, the CSU group reported on the transfer of fallout Cs-137 from feed to meat of cattle (27). This group is somewhat unique in that they are still very active as witnessed by their participation in this meeting and recent publications (28,29).

INTERSPECIES CORRELATIONS

We can learn much from interspecies correlations. I think a great deal. For example, Figure 1 shows the relation between heat production and body weight for animals ranging in mass from the mouse to the elephant. Although heat production based on body surface predicts a function with a slope of $2/3$, the actual slope is $3/4$ (30). Figure 2 gives the relation between the log of water loss and the log of body weight for animals ranging in size from the mouse to the horse. The regression coefficient, (0.8006 ± 0.0692) indicates that daily body water turnover is not proportional to body weight; if it were the value would be unity (31). Note that the desert kangaroo rat (*Dipodomys*) does not fit the relation as it has developed special mechanisms for living in a very xeric environment. Figure 3 relates log body water and log body solids for the 7 species. This relation shows that the ratio of water to solids is relatively constant for the 7 species and indicates that the degree of tissue hydration is quite normal in the Kangaroo rat. This emphasizes the fact that the unusual ability of this species to

exist in the desert involves special adaptations of the kidneys for conservation of water and not specialized water depots (31).

I thought I would share an interesting observation about how some bench scientists have been confused by observed differences in the metabolism of materials by animals. Cesium has been widely studied and much is known about the metabolism of this radionuclide in a large variety of animals and in man. However, when interspecies relations were being developed for cesium it became clear that although the data for many species conformed to allometric relations based on metabolic rate or body weight, the values for some animals clearly deviated from "expected" values. One such example is given in Figure 4 which shows the log-log relation of biological half-time (for the longest retention component) to body weight for 12 species of animals given radiocesium (32). The data analysis showed that two separate power functions could describe the data. It became clear that the monogastric mammals constituted one group and the ruminants (multi-chambered mammals) constituted still another group. Both sets of data can be fitted by power functions. The ruminants, often consumed by man as food, will reach equilibrium with environmental cesium at lower levels than those levels predicted from the body weight relation for mono-gastric mammals. Incidentally, it is not clear why the authors included the hen in these data as they refer to "mammalian species."

Figure 1. Interspecies correlation between log of heat production and log of body weight for 12 species.

Figure 2. Interspecific correlation between log daily water loss and log body weight for 7 species.

Figure 3. Interspecific correlation between log body water content and log body solids content for 7 species.

Figure 4. Interspecies correlation between log biological half-life of cesium and log body weight.

ACCELERATING TURNOVER OF INCORPORATED RADIONUCLIDES

A word needs to be said about accelerating the turnover of radionuclides from animals following incorporation from the environment. Considerable work has been done in laboratory animals and some work has been done in human subjects; there is in existence literature concerning this subject (33). Data also exist on the effects of dietary compositions on the transfer coefficients (percent intake per liter of milk) for radionuclides in farm animals (34).

Ferric ferrocyanide. (FFC)

Prussian blue, ferric ferrocyanide, has proved very effective in accelerating the loss of radionuclides of cesium following incorporation in the body. This material is available for use by human subjects in some countries and is kept available in nuclear power stations for use following internal contamination. FFC is also effective in reducing the body content of rubidium, another group I element.

FFC was used to reduce the body burdens of animals after the Chernobyl accident. Some work was also undertaken to study the effect of FFC on altering the human body content of radiocesium originating from Chernobyl (35). Some of the people exposed to ^{137}Cs in Goiania, Brazil, were treated with FFC (36).

Various materials have been used to decrease the uptake of radionuclides by animals and others have been used to accelerate the turnover of the radionuclide after incorporation into the body. Some of these materials, shown in Table 1, were summarized for radiocesium almost twenty years ago (37).

SUMMARY

This report presents a historical perspective of work done in the U.S. on the metabolism of radionuclides by animals. Also covered are the important concepts of interspecies metabolic correlations and means of accelerating turnover of internally deposited radionuclides. Although a large amount of data exist for a large number of radionuclides and animal species there are many parameters of interest and additional data is needed. Also, much of the data are in internal reports that are not easily accessible. In-depth reviews of work done in the U.S. are not available.

If I were asked for recommendations following this workshop, I would propose the formation of a committee to comprehensively review the available data on the metabolism of radionuclides by animals and, if necessary, to plan work that could be conducted by multinational teams of scientists. The results of the planned experiments would remove the remaining uncertainties associated with the uptake, retention and loss of radionuclides by animals that are of economic importance, primarily as food sources, to man. The data would also provide the necessary input to other international efforts related to the calculation of radiation dose per unit intake for members of the general public. These kinds of joint international studies are common in the physical sciences. More are needed in the biological sciences. In addition, this is truly a global problem requiring international collaboration and cooperation. The sharing of the burden would help ensure the collection of the necessary data in a timely and cost effective manner.

TABLE 1. Materials tested as possible antidotes for internal radioactive cesium contamination in animals and man

Materials	References
Desoxycorticosterone	Dorfman and Potts, 1947
Parathyroid extract, cortisone and	Mraz, LeNoir, Pinajian and
dinestrol diacetate	Patrick, 1956
Dietary Na and K	Mraz, LeNoir, Pinajian and
	Patrick, 1957
	Wasserman, Comar and Tapper, 1963
Oat hulls, alfalfa, soybean oil meal	Mraz and Patrick, 1957a
Bentonite, vermiculite, amberlite,	Mraz and Patrick, 1957b
alfalfa and beet pulp	
Dietary Cs	Ogawa, Machida, Suzuki and Shibate,
	1958
	Furchner and Richmond, 1962
Desiccated thyroid, Na and K	Mraz and Johnson, 1958
Citric acid, oxalic acid, Na citrate and	Ryabova, 1958
Ca gluconate	
Levulinic acid oxime and α -ketoglutaric	Takamiya, 1960
acid oxime	
Diamox	Richmond and Furchner, 1961a
Acetazolamide	Sastry and Bush, 1962
Dietary K	Richmond and Furchner, 1961b
	Wasserman and Comar, 1961
	McNeill, Green and Rapoport, 1961
EDTA	Geller, 1963
Diuretics	Rosoff, Cohn and Spencer, 1963
Temperature	Furchner and Richmond, 1963
	Furchner, Richmond and Drake, 1965
Ferric cyanoferrate (II)	Nigrovic, 1963
Chlorothiazide	Harris and McNeill, 1964
Prussian blue	Nigrovic, 1965
	Nigrovic, Bohne and Madshus, 1966
	Madshus, Stromme, Bohne and
	Nigrovic, 1966
Ferric ferrocyanide	Richmond and Bunde, 1966
Chlorothiazide, acetazolamide and	Sastry and Bush, 1964
chloride	

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