CHAPTER I

INTRODUCTION

1.1 The Problem

At the present time, fallout represents the major source of the hazardous long-lived radionuclides of concern to the general population. These radioactive materials created as the result of nuclear detonations, besides nuclear weapons testing, manufacturing and processing of nuclear fuel, reactor operation and accidents use of nuclear power sources in space vehicles and disposal of radioactive waste, are also sources of man-made environmental contamination.

Since the mid 1950's, problem of exposure to radiation have been stimulated as a result of the expanding use of nuclear energy and the occurrence of the fallout from nuclear testing programs of several nations. Recognition of the damaging effects of tissue by ionizing radiation has created a general awareness that relatively brief exposure to high levels of radioactivity may produce acute illness. Of special significance in connection with normal peacetime pursuits is the fact that prolonged contact with much lower levels may generate less immediately recognizable but certainly bodily impairments. The concern was diminished after the Partial Test Ban Treaty of 1963, eventhough two countries, France and the People's Republic of China, who have not signed the treaty are currently testing nuclear weapons in the atmosphere from time to time. Up to November 1976, The Republic of China has already conducted her 21st nuclear test. As reported in 1976 that the radioactivity were distributed at many areas in Fuguchima, northern of Japan, also Cs-137 was found to be 113 picocurie per litre of milk in Tokyo which resulted from fallout of nuclear test in China.

Fission products debris or fallout is classified as Stratospheric, Tropospheric and local. Most of the radioactive debris produced during 1961-1962 resulted from high yield weapons and was injected into the stratospheric. This together with previous stratospheric fallout, constituted the main source of world-wide contamination since over 90 % of all fission products produced by nuclear weapons test have been from weapons with yield than one magaton. Tropospheric fallout took place within a month after production in an irregular band at about the same latitude as the detonation, it was of lesser importance since it was produced by weapons of less than 1 megaton yield, which represented less than 10% of those tested. Local fallout was essentially restricted to the site of the explosion, and has not contributed significantly to the general contamination of the biosphere.

The approximated yields released from weapons test conducted in the atmosphere by all nations have been as follows: (a) 1945-1958-air burst; 38 megatons fission yield out of a total of 69 megatons surface burst; 54 fission yield out of 105; (b) 1961-1962 air burst; 101 fission yield out of 337. This has resulted in an estimated inventory of Sr-90, as of January 1964 and corrected for decay, as follows: total atmosphere 4.3 megacuries; world wide deposition 9.0 megacuries; Northern hemisphere (0-80°N) - 31 mCi/km^2; southern hemisphere (0-50°N) - 7.7 mCi/km^2 (U.N. Sci. Comm. Effects of Att. Radiation, Rept.)

The result of Cs-137 production by the end of the year 1958 was about 14 megacuries. It was estimated that 9 megacuries of this reached the stratosphere. During the second test period (September 1961 - December 1962), 15 megacuries of Cs-137 was beleived to have been injected into the stratosphere (HASL-142, 1963). Sr-90 deposition has also been shown to be roughly proportional to the amount of precipitation.

The main part of Cs-137 fallout comes down with rain. Along with the same latitude the level of Cs-137 has been observed to be almost directly proportional to the amount of rainfall. 80 to 90 percent of the Cs-137 fallout is water soluble.

In regard to radioactive fallout the main objectives are to understand the relationships between amount and patterns of release and the harm that may be caused to the human population. It is real need to monitor environment, to make prediction as to what will happen from known and anticipated injection of contamination and to know how to reduce the exposure to man.

In attempt to arrive at an evaluation of the possible dangers associated with relatively low level of fallout, resulting from bomb explosion; both those tests which have already occurred and those which will take place in the future, to human health. Therefore it is of the greatest importance to obtain observations which can be used to evaluate the danger. One of the important steps is the environmental monitoring including the determination of specific radionuclides in food, water and air.

The environmental radioactivity surveillance has been routinely conducted by the Office of Atomic Energy for Peace (OAEP) and some other laboratory. The results, up to present, show no significant contamination of fallout. (Swateganit, et., al. Bhodigen, et., al.). In addition, the determination of gross beta activity, Cs-137 and Sr-90 in water around OAEP which a Thai Research Reactor-1 (TRR-1) is situated and is beleived to pollute radioactivity into the environment has been carried out. The recent report indicated also no contamination. (Karasuddhi, et., al; Mhaitong, 1976) Furthermore, the investigation of some fission products in rice samples collected from various part of the country denotes no contamination as well. (Pumpuang, 1976). Eversince there is no sources of harmful radiation contamination inside of the country, contamination outside of the country must be brought into the consideration.

The amounts of imported food and food products particularly milk and diary products were increased greatly in the past in Thailand. Though this country, since 1970 has changed her status from the country of importer to exporter in can-foods industries. Still many foreign brands and various kinds of can-foods are distributed widely in supermarkets and local grocery stores.

As mentioned earlier, some countries still continue conducting atmospheric nuclear test, this results in fallout contamination in world wide region. It was also reported that noticable high radiation level were found in a few items of food such as marine fish and milk. These foods were actually not allowed to be consumed in those countries. There is a possibility that those contaminated foods would be transferred into canned foods and exported to other countries. Consequently, it is the real need for consumer's countries to be aware of and set up their own regulations for quality assurance.

Regarding this reason and the radiation surveillance of long-lived radionuclides particularly fission products has not yet been reported in Thailand at present. Beside, the information obtained will be of great useful as a basic knowledge for the authorized governmental agencies to be able to set up a maximum permissible limit for radiation level and concentration of fission products in these food-stuffs. It is, therefore the purpose of the study to investigate the radiation level and some long lived radionuclides in imported foods and food products.

1.2 Objectives of the study

1.2.1 To measure the radiation level of long-lived radionuclides in imported foods and food-products.

1.2.2 To investigate the activity concentration of long-lived fission products in those foods.

1.2.3 To provide the informations used for guidance in the stipulation of radiation hazards in imported food and food-products within the country.

1.2.4 To stimulate the authorized governmental agencies for radiation legalization.

1.3 Scope of the investigation

1.3.1 Food samples were purchased monthly from different supermarkets or grocery stores.

1.3.2 Each sample was blended and searched for gamma spectrum of long-lived radionuclides using a gamma spectrometer.

1.3.3 Samples were oven-dried and ashed respectively for the analysis of gross beta activity using low background anti-coincidence G-M counter.

1.3.4 Sample were wet-ashed and investigated for strontium-90 by solvent extraction technique using tri-n-butyl phosphate as an extractant.

1.4 The benefit obtaining from the study.

1.4.1 The information obtaining will be brought into the consideration whether those foods and food-products had been radiation-contaminated.

1.4.2 The information obtaining can be used to assess the radiation safety limit for imported food and food products.

1.4.3 This information will be a great help for Thailand's economic as well as the health and welfare of her people.

1.5 Literature search of previous work.

1.5.1 The occurrence, pathway and uptake of Sr-90 and Cs-137 in the environment.

Sr-90 and Cs-137 resulted from process in atomic bomb explosion or reactor operation were accomplished with other 200 or so fission products. The fallout emitted gamma photons of various energies, with an average of the order of 0.9 MeV. Sr-90 was produced at a level equivalent to about 1 mCi of Sr-90 per square mile of the earth sarface for every 2 megaton of fission energy and radiocesium was produced at about 50 percent higher yield (Hardy, 1967). Both were considered to represent the most possible hazard to man; their potential importance were determined by: (a) production yield (b) half-life (c) extent to which they entered parts of plants, consumed by animals (d) extent to which it entered human food of animal origin (e) extent to which they entered into man (f) degree and pattern of retention in man and (g) radio sensitivity of tissue of retention (Comar, 1965).

There were two important ways inwhich plants may become contaminated by fission products from the atmosphere.

(a) Indirect contamination which occured when radioactive materials entered the soil and passed into the plant through the roots as soil nutrients did.

(b) Direct contamination, inwhich passage through the soil was by passed.

Direct contamination reflected events of the recent past; it varied with the rate at which fallout settled and was designated as "rate dependent". Indirect contamination was designated as "accumulative dependent". (Frere, et., al. 1963).

Radionuclides in/on plants may reach man directly by his consumption of foods of plant origin or indirectly by his consumption of animal products. The grazing animals effectively collected contamination from plant materials and concentrated it in animal products. Man could also receive contamination though his consumption of fisheries's product. (Russell)

In biological systems, the behavior of Sr-90 was similar to and was governed by the behavior of Ca, which was essential constituent of animals and humans. Sr-90 then followed it through the food chain, finally to man; the end of the food chain and concentrated in bones, teeth and milk. There was a remarkable constancy of calcium in many tissue and fluid (eg. blood and milk). (Wasserman, et., al. 1961). Then Sr-90 to Ca ratio in the body tissues or secretion was directly related to that ratio that existed in the diet. Fortunately, Sr-90 a heavior element was discriminated against. (Comar, et., al. 1956).

The absorption of Sr-90 and Ca from soil was affected by the clay content, humus content, pH, moisture level, concentration of electrolytes and especially the calcium content. (Comar, 1965). Conditions that led to shallow root development tended to increase Sr-90 absorption. The addition of lime to soils of low Ca-content could reduce the Sr-90 uptake by a factor of up to three. The acquired Sr-90 were obtained from milk, milk products, vegetables, cereals and fish (AEC, 1956).

Cs-137 decays with a half-life of 30.5 years. After reaching the ground along with the rain, Cs-137 was very effective bound by the soil or absorbed by plants through their outer surface. Cs-137 was further transported from plants either directly or in the process of being eaten by animals into man. A part of Cs-137 that had reached water courses and seas was carried through various food chain into fish and further into man (Davis, 1963). **Resembling to potasium, cesium** accumulated in soft tissue mostly in muscle. The compounds of this element were more soluble than the corresponding compound of Sr and Ca. (Glasstone, 1962). The amount of internal exposure to Cs-137 was determined by the quantity of this isotope in the diet. If the major mechanism for its incorporation into the diet was through the root systems of plants, then the dose would be more or less proportional to the total amount of Cs-137 accumulated on the ground. If directly deposited on the leaves of plant, the internal dose would be more nearly proportional to the rate of descent of delayed fallout. The potasium content of plants could effect on the Cs-137 level such a way that when the potasium content increased, the Cs-137 content decreased. (Hasanen, 1972). In the sea, due to its high electrolyte and low Cs-137 content, the Cs-137 level in the fish was an order lower than in the fresh-water courses. Also the flesh of aquatic organism concentrated Cs-137 by a factor of 5 to 20 over that of the surrounding water. Man obtained most of his Cs-137 from milk, meat and grain products **(Russell,** 1966)

1.5.2 Environmental studies

The first introduction of man-made radionuclides into a marine environment was at Bikini Atoll in 1946. Two 2 kiloton devices were detonated, the following year through 1958, Bikini and Eniwetok became the Pacific Proving Ground. During that time nuclear and thermonuclear devices with a total yield of many megatons were detonated at the atolls. Dunning (1957) and Glasstone (1962) reported that Rongelap Atoll was heavily contaminated with radioactive fallout in 1954. Held (1954) studied the land-dwelling hermit crab, Coenobita sp. and coconut crab, Birgus Latro and stated that they were sensitive index of biologically available fission-products in the environment; 84 % Cs-137, 10 % Sr-90 + Y - 90 and 1 % Cs-144-Pr-144 were in muscle tissue, radioactivity in the skeleton was found to be due almost entirely to radiostrontium and the Y-90 daughter of Sr-90. The level of Sr-90 and Cs-137 were also found to remain virtually constant at 4500 pCi of Sr-90 per gm. of skeleton and 450 pCi of Cs-137 per gm. of muscle in Coenobiota sp. at Eniwetok over a period of two years.

In the year 1958, Setler, et. al. revealed that Sr-90 and Cs-137 when released into the environment concentrated in bottom mud, soils and biological materials. Welander, et. al. (1967) studied of Birgus sp. at Rongelap Atoll and showed that the crabs contained more than 100 pCi Sr-90 per gm. of skeleton and 100 pCi of Cs-137 per gm. of muscle over a period of 10 years, 1954-1963. Chakravarti and Eisler (1958) reported that there was a linear relationship between Sr-90 activity and gross beta activity of the Liver of the coconut crab (Birgus Latra sp.) collected at

Rongelap Atoll. Walker, et. al. (1959) found that the principal radionuclides in plant tissues grown in greenhouse pot cultures using soil from Rongelop Atoll, which received fallout contamination in 1954 were to be Cs-137. Palumbo (1961) reported that Sr-90 and Cs-137 were both soluble and were the ones taken up in highest amount by the land plants and animals.

In 1963 Hardy, Jr., et. al. reported that the Cs-137 and Sr-90 body burdens of people living on Rongelap Island were high compared to most other populations of the world. This was that the natives consumed food that were contaminated with long-lived fission products radioactivity resulting from a fallout incursion in 1954. Several native food items were consumed by one of the authors under controlled conditions. The result of metabolic balance study was 50 % of ingested Cs-137 in the Rongelap food had been excreted in urine after 85 days, where as 14 % had been eliminated in the feces during the same time. In cantrast, most of Sr-90 was unabsorbed, 50 % had been excreated in feces at 10 days, where as only 2 $\frac{1}{2}$ % had been excreated in urine. The retention of Cs-137 showed a biological half life of 24 days Sr-90 retention as a function of time was best described as a series of exponentials and approach a value of 25 % after 140 days.

During 1946 to 1957, Morphy and Campbell analyzed Sr-90 and Cs-137 content of New York milk and the data showed the two large peaks at the end of the two periods of heavy atmospheric nuclear testing of the U.S. and the U.S.S.R. . Gustafson, et. al. (1964) reported that milk was found to be the most important single dietary contributor of Cs-137, after the resumption of nuclear testing in 1961, the fivefold increased in the chicago dietary level between 1961 and late 1963 had been matched by a similar increased in whole body burden with a time lag of 6-7 months. In 1968 Phelps and his team reported that many various biological and environmental samples were shown to be contaminated with world wide fallout. The only radionuclides in food items detected were Cs-137 and K-40. The lowest level of Cs-137 was found to be 12.7 pCi/228 gm. of shrimp, 34 pCi/gm. of milk and 24.2 pCi/gm. of beef.

Investigation of the diets of the Finnish Lapps had revealed that fish was the second important sources of radiocesium values especially in Pike. Analyses of white-fish, trout and pike seemed to have activities risen during the year 1962. (the period of 1961-1962 was the time of 2nd nuclear testing). Since 1968, the Cs-137 in Lapp had remained almost constant, and this was due to the atmospheric nuclear testing by the People's Republic of China in the latter half of the 1960's. (Hasanen, 1972).

Gamarekian and staffs disclosed in 1959 that the white bread sold in a New York City supermarket had a radioactive Sr-90 content over the maximum permissible limit.

Another Atomic Energy Commission (AEC) report showed that in 1957, the Sr-90 level in plants grown in several parts of the United States had exceeded the permissible limit for foods. Plumbo (1957) indicated the abrupt rise in Sr-90 content of milk in the Mandan, North Dakota area. Whole cereal grains were particularly likely to show a high Sr-90 content in years in which an especially heavily contaminated rain occurred while the grain was maturing. Hasanen (1972)informed that the high level of Cs-137 in the Finland Lapp obtained from the Reindeer and Caribou breeders, the large part of their diets. These animals lived on various lichen and mosses which contained between ten to a hundred times more Cs-137 than other terrestrial plants. For example, for Lapland males a daily intake of 5000 pCi of Cs-137 was reported, of which meat contributed about 4330 pCi; this compares to a total diet intake in the United States of about 30 pCi. Booth and Samuels reported in 1968 the fallout Sr-90 level in Canadian milk, wheat, whole diet and human bone. Peak values occurred in 1963. In wheat, the levels correlated with the rate of fallout deposition during the season of growth, contrary to milk, uptake from the soil was much more important. Also the level in milk obtained from higher natural calcium content soil was much lower. Milk surveillance on March 1974 showed Sr-90 monthly average 0-17 pCi/l. (picocurie per liter) in the United States, and the highest 12 month average was 16 pCi/l (Minn.) representing 8.0 % of the Federal Radiation Council Radiation Protection Guide. Cs-137 monthly average ranged from 0-59 pCi/l and the highest 12-month average was 48 pCi/l (South florida) representing 1.3 % of the value derivedfrom the recommendation of the Federal Radiation Council Reports . . (Hainberger, et. al. 1974).

1.5.3 Toxicity of fallout

Wherever fallout occurred there was a chance that radioactive material would enter the body through the digestive tract (due to the consumption of food and water contaminated with fission products),through the lungs (by breathing air containing fallout particle), or through wounds or abrasions. (USAEC, 1962). It should be noted that even a very small quantity of radioactive material present in the body can produce considerable injury.

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Both Sr-90 and Cs-137 presented a potential long-term hazard. The special interest in the delayed fallout arose from the fact that it may occur in significant amounts in many parts of the globe remote from the point of the nuclear detonation, as well as in close by areas. The most serious pathological change in the body was due to the effects of internal radiation. Once they entered the body they became internal source and stayed there for a long period of time. Regarding many experiments, the biological half life of Cs-137 is 70-140 days. Because of the penetrating properties of 0.667 MeV., γ-ray and 1.176 MeV., β energy emission, the radiation was distributed more or less uniform to all part of the body. In spite of gamma ray emmission, the biological half life of Cs-137 is so relatively short for the same amount of delayed fallout, the residual Cs-137 is less of a general biological hazard than the residual Sr-90. (Glasstone, 1962).

Although Sr-90 emits beta particles of fairly low energy of 0.54 and 2.27 MeV., sufficient amount of this isotope can produce damage because once it gets into skeleton it will stay there for a long time the result from damaging quantities of Sr-90 may be anaemia, bone necrosis, cancer and possibly leukemia. (USAEC, 1962)

Cs-137 however constitutes a relatively more important genetic hazard because of it fairly long range gamma ray and is held in the reproductive organs. (Glasstone, 1962).

After the World War II, Grahn observed the Japanese survivors and found that Leukemia was at 2 to 3 fold greater frequency on the average than in unexposed Japanese. The same result reported by Warren, (1945) that Leukemia (irradiation of bone marrow) deaths were consistently higher in radiologists than among other medical specialist. Also in 1959, Coaster pointed out that the average life span of the radiologist was 60.5 years as compared with 65.7 years for other medical groups.

Polikovpor and Ivanor (1961, 1962), Ivancr (1965) and Polikovpor (1966) reported on the effect of Sr-90-

Y-90 at low concentration in sea water on the development of eggs of Blacks sea fishes. Reduced hatching of the larvae and early mortality were seen at concentration of 10^{-7}Ci/l (Curie per liter) and above, and the number of abnormalities increased significantly and with remarkable consistancy at concentrations of 10^{-10} Ci/l and above. White and Angelovic (1966) reported that eggs and Larvae of nummichogs (Fundulus heteroclitus) showed a general retardation in the rate of development when chronically exposured to low level of Cs-137. Ivanor (1967) reported decreasing in the mitotic activity when exposed to Sr-90 from 10^{-10} Ci/l to 10^{-5} Ci/l of the cell and the percentage of chromosome aberrations increased when the concentration exceeded 10^{-9} Ci/l.