Chapter 5

Incorporation and Accumulation of Strontium-90 in the Hard Tissue of Animals and Their Relationship with Strontium-90 Pollution in the Environment

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Abstract Substantial amounts of radionuclides including strontium-90 (^{90}Sr) were released by the Fukushima Daiichi Nuclear Power Plant (FNPP) accident. In the present study, we describe and discuss the presence of ^{90}Sr in the ex-evacuation zone of the FNPP accident and its relationship with ^{90}Sr activity concentration in the hard tissue of animals. We found that the activity concentration of ^{90}Sr in the hard tissue exhibited a positive correlation with ^{90}Sr pollution in their corresponding terrestrial and marine environments. Hard tissues, such as the teeth, bones, and otoliths, of animals and fishes could serve as useful tools in assessing ^{90}Sr pollution in the environment during the period of the formation of those tissues.

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5.1 Introduction

Strontium-90 (^{90}Sr) has been released to the environment from various nuclear disasters in the past, such as nuclear weapon tests [1], the Chernobyl Nuclear Power Plant accident [2, 3], release of contaminated radionuclides from nuclear fuel processing plants at Sellafield and Mayak facilities [4, 5], and sea disposal operations by the former Soviet Union [6]. Strontium-90 was released also by the Fukushima Daiichi Nuclear Power Plant (FNPP) accident in 2011 and remained in the terrestrial and marine environments due to its long physical half-life of 28.8 years. Its long biological half-life and bone-seeking property may have adverse effects on the bone marrow, and special attention should be paid to the behavior of ^{90}Sr. Strontium-90 easily transfers to terrestrial and marine biota because it is rather soluble [7, 8]. However, the relationship between ^{90}Sr in the environment and in animal body is still unclear. In this chapter, we describe and discuss the presence of ^{90}Sr in the environment and its migration into the teeth and bones of cattle abandoned in the ex-evacuation zone of the FNPP accident. We also discuss ^{90}Sr activity concentration in the otolith of marine fishes around FNPP. Through the discussion, we suggest that ^{90}Sr activity concentration in the hard tissue of animal body might reflect the extent of ^{90}Sr pollution in the environment.

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5.2 ^{90}Sr Pollution by the FNPP Accident

5.2.1 ^{90}Sr in the Environment

The Ministry of Education, Culture, Sports, Science and Technology, Japan (MEXT), reported that 0.1–6 kBq/m^2 of ^{90}Sr and 0.3–17 kBq/m^2 of ^{89}Sr were detected in soil within a 20-km radius from FNPP (the ex-evacuation zone) [9]. Strontium-90 is known to have been present in the Japanese environment before the FNPP accident, which probably stemmed from the atmospheric nuclear weapon tests conducted during the 1950s–1980s. However, the existence of ^{89}Sr in soil of the ex-evacuation zone indicates that the ^{90}Sr pollution was caused by the FNPP accident, because ^{89}Sr has a relatively short half-life of 50.5 days and more than 50 years have passed since the atmospheric nuclear weapon tests were discontinued.

It was reported that the amount of ^{90}Sr released by the FNPP accident into the atmosphere is smaller than those of volatile radionuclides (noble gases, iodine, tellurium, and cesium). The amount of released ^{90}Sr is estimated to be three orders of magnitude smaller than that of cesium-137 (^{137}Cs) [3]. The nuclear fuel of FNPP did not exceed the temperature of 2,700 K that is necessary for the volatilization of refractory elements, such as ^{90}Sr and actinides [10]. Schwantes et al. estimated that most of radioactive Sr retained inside the reactors [11]. These reports suggest that the area polluted by ^{90}Sr might be smaller than that by ^{137}Cs attributed to the FNPP accident.

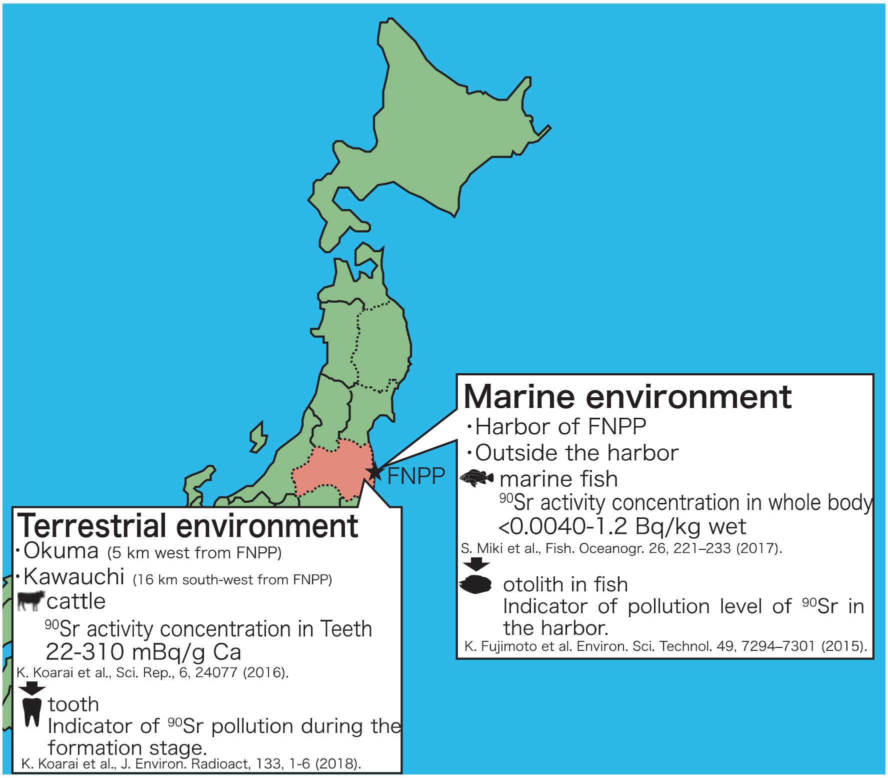
It was reported that ^{90}Sr activity concentration in soil and vegetation samples is one to four orders of magnitude lower than that of ^{137}Cs [12]. Sahoo et al. showed that ^{90}Sr activity concentration in soil from the ex-evacuation zone of the FNPP accident ranged from 3 to 23 Bq/kg, whereas ^{137}Cs activity concentration ranged from 0.7 to 110 kBq/kg [13], which is consistent with the previous reports [3, 10]. The ratio of ^{90}Sr/^{137}Cs in soil widely varies depending on the area examined.

The FNPP accident caused ^{90}Sr pollution not only in the terrestrial environment but also in the marine environment. Since the most of ^{90}Sr was presumed to remain in the reactor of FNPP, atmospheric release of ^{90}Sr was speculated to be minor. The total amount of ^{90}Sr released into the sea was estimated in the range from 0.09–0.9 PBq [14] to 1–6.5 PBq [15], and ^{90}Sr activity concentration in surface seawater near the harbor of FNPP was 0.2–400 kBq/m^3 [15]. The activity concentration of ^{90}Sr was an order lower than that of ^{137}Cs from April 2011 to February 2012, except for December 2011 when ^{90}Sr and ^{137}Cs activity concentrations were equivalent due to the discharge of ^{90}Sr-contaminated wastewater [16]. In the Pacific Ocean 15 km east from FNPP, ^{90}Sr activity concentration was two orders lower than that in seawater near FNPP. Owing to diffusion in offshore regions of Fukushima Prefecture, the pollution is believed to be limited to the sea around FNPP. These suggest that the ^{90}Sr pollution of the terrestrial and marine environments are limited to the vicinity of FNPP.

5.2.2 ^{90}Sr Activity Concentration in the Hard Tissue of Abandoned Cattle After the FNPP Accident

We previously reported activity concentration and specific activity of ^{90}Sr in the teeth of cattle stayed in the ex-evacuation zone (Fig. 5.1) of the FNPP accident [17]. Because of chemical similarity between Sr and calcium (Ca), the teeth incorporate Sr during their formation period (calcification period) and retain it until they fall out or are worn down. As the amount of ^{90}Sr accumulated in the teeth is assumed to reflect the amount of ^{90}Sr incorporated into the body during the formation of the teeth, we hypothesized that the assessment of ^{90}Sr activity concentration in the teeth might provide useful information about the degree of ^{90}Sr contamination in the environment.

Figure 5.2 shows the relationship between ^{90}Sr activity concentration in hard tissues (teeth and mandibular bones) of cattle and in soil from the ex-evacuation zone (Okuma Town and Kawauchi Village) after the FNPP accident. Activity concentrations of ^{90}Sr in the teeth were 150–830 mBq/g Ca (ratio of ^{90}Sr radioactivity to Ca weight in the tooth) (average, 470 mBq/g Ca) for Okuma Town and 100–310 mBq/g



Terrestrial environment
- Okuma (5 km west from FNPP)
- Kawauchi (16 km south-west from FNPP)

cattle

^{90}Sr activity concentration in Teeth
22-310 mBq/g Ca

K. Koarai et al., Sci. Rep., 6, 24077 (2016).

tooth
Indicator of ^{90}Sr pollution during the formation stage.

K. Koarai et al., J. Environ. Radioact, 133, 1-6 (2018).

Marine environment
- Harbor of FNPP
- Outside the harbor

marine fish

^{90}Sr activity concentration in whole body
<0.0040-1.2 Bq/kg wet

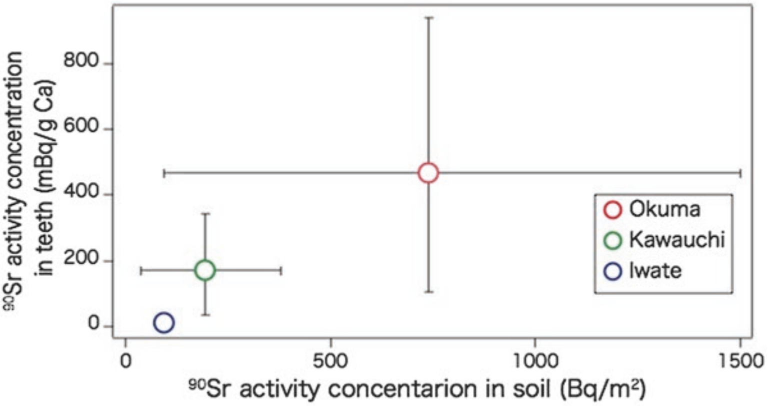
S. Miki et al., Fish. Oceanogr. 26, 221–233 (2017).

otolith in fish
Indicator of pollution level of ^{90}Sr in the harbor.

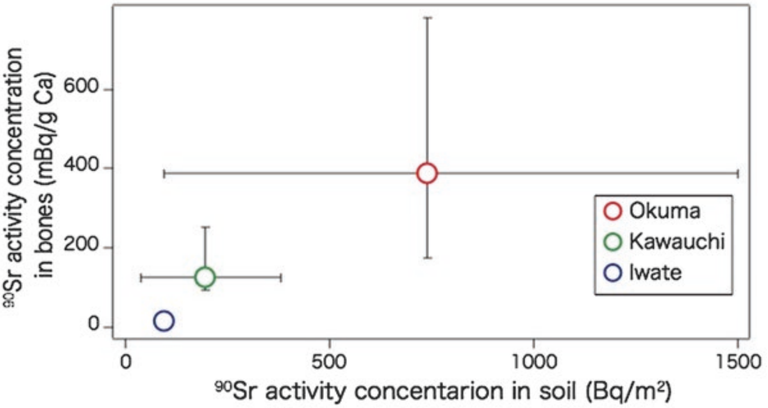
K. Fujimoto et al. Environ. Sci. Technol. 49, 7294–7301 (2015).

Fig. 5.1 ^{90}Sr pollution in animals after the FNPP accident

Ca (average, 170 mBq/g Ca) for Kawauchi Village [17]. The deposited amount of ^{90}Sr in soil was 94–1500 Bq/m^2 (average, 740 Bq/m^2) for Okuma Town and 39–380 Bq/m^2 (average, 200 Bq/m^2) for Kawauchi Village [9]. We selected Iwate Prefecture as the control area because it is approximately 250 km north from FNPP and is considered free from FNPP-related ^{90}Sr pollution. As shown in Fig. 5.2a, ^{90}Sr activity concentration in cattle teeth was significantly correlated with that in soil; the higher the ^{90}Sr activity concentration in soil, the higher the activity concentration in cattle teeth. A similar correlation was also found between ^{90}Sr activity concentration in soil and that in the mandibular bone (Fig. 5.2b). Metabolic turnover rates (modeling and remodeling) of the mandibular bone in the cattle are not well-



(a) Soil vs Tooth



(b) Soil vs Bone

Fig. 5.2 Relationship between ^{90}Sr activity concentration in hard tissues and that in soil. (**a**) Relationship between teeth and soils. (**b**) Relationship between bones and soils. Data of ^{90}Sr activity concentration in the teeth were obtained from our previous report [17]. Data of ^{90}Sr activity concentration in soil were obtained from Japanese government reports [9, 18]. Each symbol indicates the average value with error bars showing the maximum and minimum values known. However, a major part of the original bone that formed before the FNNP accident was believed to be replaced by new bone formed after the accident, because, at the time the cattle were killed, 16–25 months had already passed since the accident. Thus, ^{90}Sr activity concentrations in hard tissues (both in the teeth and bones) reflect environmental ^{90}Sr levels during the formation of the tissues.

5.2.3 ^{90}Sr Pollution in Fish

Miki et al. reported ^{90}Sr activity concentrations in marine fish before and after the FNPP accident (Fig. 5.1) [19]. Activity concentrations higher than the background level before the FNPP accident were detected in 4 of 26 specimens collected just outside of the ex-evacuation zone, whereas ^{90}Sr activity concentrations in all samples collected offshore the neighborhood of Fukushima Prefecture were under the detection limit. The report pointed out that ^{90}Sr was detected only in marine fish living in the ocean near FNPP. Higher ^{137}Cs activity concentration was found in the whole body of teleost fish; the activity ratio of ^{137}Cs to ^{90}Sr was 5–190 times higher than that before the accident.

Regarding ^{90}Sr activity concentration in marine biota, the otolith of fish is a hard tissue composed of $CaCO_3$ and might be a unique index of ^{90}Sr pollution. In the process of otolith calcification, Sr is incorporated with Ca and is not metabolized once it is formed. It is reported that the β-ray count rate from otoliths of Japanese rockfish correlates with the activity concentration of radioactive Cs and ^{90}Sr in the whole body of fishes in the main harbor of FNPP. On the other hand, no β-rays were detected from fish collected outside of the main harbor of FNPP [20].

The otolith could reflect ^{90}Sr pollution in the living area of the fish. However, the use of otoliths as a biomonitor may not be feasible for detecting low levels of pollution, such as that outside FNPP harbor, because the amount of ^{90}Sr would be too small in the otolith. If more sensitive determination methods, such as mass-spectrometric quantification [21], were developed, ^{90}Sr in otoliths could be determined even with low radioactivity of the nuclide.

5.2.4 Migration of ^{90}Sr from the Environment to Hard Tissues of Animals

Strontium in soil exists in two forms; one is an insoluble solid form, and the other is the soluble form (exchangeable fraction) that can be dissolved in water or other aqueous solutions such as ammonium acetate [22, 23]. Considering the migration of ^{90}Sr from soil to hard tissues of animals, ^{90}Sr in the latter form is believed to be

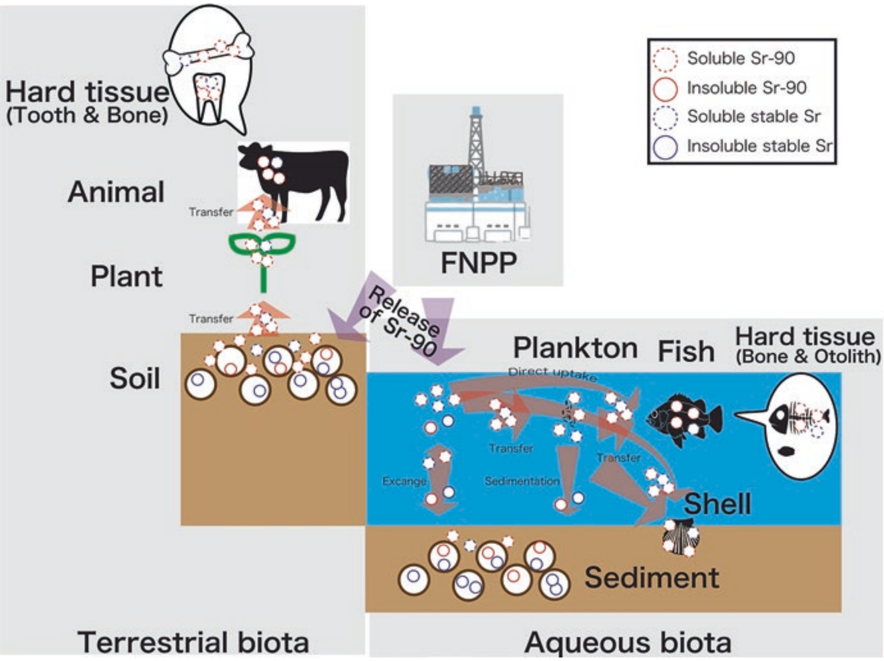


Fig. 5.3 Diagram of ^{90}Sr transfer in terrestrial and aqueous biota

important because Sr isotopes in the exchangeable fraction would be transferable in terrestrial biota. According to our analysis of soil from the ex-evacuation zone [23], only 1–9% of Sr in soil was in the exchangeable fraction, whereas 37–84% of ^{90}Sr in soil was in the exchangeable fraction. Consequently, the specific activity of ^{90}Sr (^{90}Sr/total Sr ≈ stable Sr) in the solid form was low (23–33 Bq/g Sr in Kawauchi Village and 11–36 Bq/g Sr in Okuma Town), and specific activity in the exchangeable fraction was high (200–330 Bq/g Sr in Kawauchi Village and 130–1200 Bq/g Sr in Okuma Town). As both ^{90}Sr and stable Sr behave similarly in the biota, the specific activity of ^{90}Sr would basically be identical in the migration route of ^{90}Sr from soil to plants and plants to animals (Fig. 5.3). In fact, specific activities of ^{90}Sr in cattle teeth (260–640 Bq/g Sr in Kawauchi Village and 380–1400 Bq/g Sr in Okuma Town) were in a similar level as those in the exchangeable fraction in soil.

After the FNPP accident, cattle in the ex-evacuation zone were released to the polluted field and allowed to graze on contaminated grasses. Once it enters the body, Sr is known to be incorporated in hard tissues during their formation period and accumulate there. The main chemical form of Sr in the teeth and bones is hydroxyapatite, $Ca_{10}(PO_4)_6(OH)_2$, in which any fraction of Ca may be replaced by Sr with little change in the apatite structure. ^{90}Sr incorporated in the hard tissue may exist as stable $Sr_XCa_{(10\text{-}X)}(PO_4)_6(OH)_2$ in the apatite crystal. Thus, ^{90}Sr activity concentration in the hard tissue of animals reflects the environmental pollution level during the formation of the tissue. Soluble form of Sr may migrate through the food chain and get directly incorporated into fish body along with the absorption of Ca^{2+}. ^{90}Sr would be accumulated in the hard tissues of fish, such as the bones, teeth, and otoliths. Therefore, the hard tissue of fishes may also provide useful information about ^{90}Sr pollution in the marine biota, as previously reported [20].

5.3 Summary and Perspectives for the Future Study

We described several aspects of ^{90}Sr pollution in the environment after the FNPP accident and ^{90}Sr activity concentration in the hard tissue of animal body. This article reviewed ^{90}Sr pollution in soil and seawater, hard tissues of abandoned cattle, and contaminated marine fishes around FNPP after the accident. Although the degree of environmental pollution has gradually been declining for the last several years, continuous research is necessary for clarifying ^{90}Sr mobility in the environment affected by the FNPP accident. Hard tissues such as the teeth, bones, and otoliths have the potential to reflect ^{90}Sr pollution of the environment during their formation period. It should be emphasized that ^{90}Sr specific activity in the teeth during their formation period and that in the exchangeable fraction in soil is at the same level [23]. This means that ^{90}Sr specific activity in the teeth may serve as a useful tool for understanding the migration route of ^{90}Sr in terrestrial biota. Furthermore, as the amount of ^{90}Sr in the teeth may be proportional to the amount of ^{90}Sr incorporated into the body during teeth formation, ^{90}Sr in the teeth provides a useful index for the individual assessment of internal exposure to radiation. Estimation of individual exposure dose is essential for understanding the biological effect of radiation, especially animals affected by the FNPP accident. In this sense, the examination of radioactive nuclides including ^{90}Sr in hard tissues provides useful information about the exposure dose to radiation. Nevertheless, further studies are required to elucidate the relationship between ^{90}Sr in hard tissues and internal exposure dose to the nuclide.

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