Journal of Environmental Radioactivity 183 (2018) 1–6

Contents lists available at ScienceDirect

Journal of Environmental Radioactivity

journal homepage: www.elsevier.com/locate/jenvrad

^{90}Sr specific activity of teeth of abandoned cattle after the Fukushima accident – teeth as an indicator of environmental pollution

Kazuma Koarai[a], Yasushi Kino[a], Atsushi Takahashi[b], Toshihiko Suzuki[c,d], Yoshinaka Shimizu[c], Mirei Chiba[c], Ken Osaka[c,d], Keiichi Sasaki[c], Yusuke Urushihara[e], Tomokazu Fukuda[f], Emiko Isogai[g], Hideaki Yamashiro[h], Toshitaka Oka[a,i], Tsutomu Sekine[a,i,*], Manabu Fukumoto[j], Hisashi Shinoda[c]

[a] Department of Chemistry, Tohoku University, Japan
[b] Tohoku University Hospital, Tohoku University, Japan
[c] Graduate School of Dentistry, Tohoku University, Japan
[d] International Research Institute of Disaster Science, Tohoku University, Japan
[e] Tohoku University School of Medicine, Tohoku University, Japan
[f] Faculty of Science and Engineering, Iwate University, Japan
[g] Graduate School of Agricultural Science, Tohoku University, Japan
[h] Faculty of Agriculture, Niigata University, Japan
[i] Institute for Excellence in Higher Education, Tohoku University, Japan
[j] School of Medicine, Tokyo Medical University, Japan

ARTICLE INFO

Keywords:
^{90}Sr
^{90}Sr specific activity
Cattle teeth
Environmental pollution
Transfer
Fukushima-Daiichi Nuclear Power Plant accident

ABSTRACT

^{90}Sr specific activity in the teeth of young cattle that were abandoned in Kawauchi village and Okuma town located in the former evacuation areas of the Fukushima-Daiichi Nuclear Power Plant (FNPP) accident were measured. Additionally, specific activity in contaminated surface soils sampled from the same area was measured. (1) All cattle teeth examined were contaminated with ^{90}Sr. The specific activity, however, varied depending on the developmental stage of the teeth during the FNPP accident; teeth that had started development before the accident exhibited comparatively lower values, while teeth developed mainly after the accident showed higher values. (2) Values of ^{90}Sr-specific activity in teeth formed after the FNPP accident were higher than those of the bulk soil but similar to those in the exchangeable fraction (water and CH_3COONH_4 soluble fractions) of the soil. The findings suggest that ^{90}Sr was incorporated into the teeth during the process of development, and that ^{90}Sr in the soluble and/or leachable fractions of the soil might migrate into teeth and contribute to the amount of ^{90}Sr in the teeth. Thus, the concentration of ^{90}Sr in teeth formed after the FNPP accident might reflect the extent of ^{90}Sr pollution in the environment.

1. Introduction

The long half-life (28.8 years) and bone-seeking properties of ^{90}Sr make it an artificial radionuclide of concern among the fission products. It is well known that ^{90}Sr can persist in bone with a retention half-life of more than 10 years (Froidevaux et al., 2010, 2006). ^{90}Sr and its daughter nuclide, ^{90}Y, emit β-rays (max. energy 0.546 MeV and 2.28 MeV, respectively) that adversely affect the bone marrow. Thus, special attention is paid to ^{90}Sr in the environment long after nuclear weapon tests and/or nuclear accidents, such as the Chernobyl accident (International Atomic Energy Agency, 2010; 2008).

The Fukushima-Daiichi Nuclear Power Plant (FNPP) accident brought radioactive pollution through a substantial release of artificial radionuclides into the environment (Povinec et al., 2013; Steinhauser et al., 2014; Tagami et al., 2011; Thakur et al., 2013). In June 2011, the Japan Ministry of Education, Culture, Sports, Science, and Technology (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 the soil of areas within a 20-km radius from the FNPP (i.e., the former Fukushima evacuation zone) (MEXT, 2011). There have been other reports on ^{90}Sr contamination in the environment after the FNPP accident. Steinhauser et al. (Steinhauser, 2014; Steinhauser et al., 2014) estimated that the amounts of atmospheric release of ^{90}Sr and ^{89}Sr were ~0.02 and ~0.2 PBq, respectively. Concentrations of ^{90}Sr and ^{137}Cs in soil and vegetation samples have

* Corresponding author. Institute for Excellence in Higher Education, Tohoku University, 41 Kawauchi, Sendai, Miyagi 980-8576, Japan.
E-mail address: tsekine@m.tohoku.ac.jp (T. Sekine).

https://doi.org/10.1016/j.jenvrad.2017.12.005
Received 15 September 2017; Received in revised form 12 December 2017; Accepted 12 December 2017

been reported by Steinhauser et al. (2013), who showed that the ^{90}Sr concentration was 1–4 orders of magnitude lower than that of ^{137}Cs. Povinec et al. (2013) reported that the ^{90}Sr concentration in the western part of the northwestern Pacific Ocean was equal to or lower than that of ^{137}Cs in surface seawater between 2011 and 2012. Casacuberta et al. (2013) determined that ^{90}Sr activity in surface seawater samples ranged from 1.1 ± 0.2 to 85 ± 3 Bq m^{-3}. Miki et al. (2017) reported concentrations of ^{90}Sr in marine fish before and after the FNPP accident. Fujimoto et al. (2015) reported that ^{90}Sr in otolith could be a biomarker to show that fish had lived in the FNPP harbor. These reports pointed out that the effect of the FNPP accident on marine fish was detected only in the ocean nearby the plant. Although the scale of environmental pollution is gradually becoming clear, continuous research is necessary from the viewpoint of the importance of pollution by radioactive nuclides.

Among the various indices that can be used to understand environmental contamination levels, in this study, we focused on the activity concentration and the specific activity of ^{90}Sr in cattle teeth and in the soil. As ^{90}Sr is supposed to be incorporated in cattle teeth from contaminated soils through the consumption of water or plants, we investigated how ^{90}Sr and strontium are transferred from the soil to teeth, and which component in the soil is essential for this transfer. Strontium is one of the alkaline earth elements, and its migration behavior is thought to be associated with that of calcium. Therefore, along with strontium, we measured the calcium concentration in environmental samples to gain a deeper understanding of the migration of ^{90}Sr from the soil to teeth. We determined ^{90}Sr activity in cattle teeth and soils in cattle habitat areas after the FNPP accident. We discuss the relation between the amount of ^{90}Sr in the tooth and the developmental stage of the tooth at the time of the FNPP accident. Further, we discuss the importance of ^{90}Sr in the exchangeable/leachable fraction of the soil in understanding the presence and the amount of ^{90}Sr in the tooth.

2. Materials and methods

Detailed information about the cattle teeth collection site and date was described in our previous report (Koarai et al., 2016). The information is summarized in the following section. Tooth radioactivity data were partially taken from the same report for further discussion.

2.1. Sample collection

Cattle teeth and soil samples were collected from sites located in Kawauchi village (16 km south-west from the FNPP) and Okuma town (5 km west from the FNPP) in Fukushima Prefecture, Japan (Fig. 1). The deposition amounts of ^{90}Sr in the soil at these sites reported by the MEXT (2011) are shown in Fig. 1, together with the data generated in this study. Radioactivity in Okuma town tended to be higher than that in Kawauchi village; however, the data variation was very large, indicating that the pollution was quite localized.

Four young cattle (2 at each site) were selected for this study. The cattle were collected in the course of“Project for comprehensive exposure dose assessment for disaster-affected animals” of Tohoku University (Fukuda et al., 2016, 2015, 2013; Takahashi et al., 2015; Takino et al., 2017; Urushihara et al., 2016; Yamashiro et al., 2014, 2013). Tooth samples were collected in November and December, 2011 in Kawauchi village, and in July 2012 and March 2013 in Okuma town. The 4 cattle were 8 months old at the time of the FNPP accident. The cattle were anesthetized and euthanized following protocols approved by Tohoku University (No. 2014KDO037).

Surface-soil samples were collected in Kawauchi village (10 × 10 cm, 1–2 cm depth, cambisol, 80–160 g (dry weight), 3 batches) and in Okuma town (25 × 25 cm, 1–2 cm depth, andosol, 770–960 g (dry weight), 4 batches).

2.2. Analysis

2.2.1. Teeth

Nine teeth were dissected from the mandible (Koarai et al., 2016) and each tooth was numbered according to the chronological order of development (No. 1: first deciduous molar (DM1); No. 2: second deciduous molar (DM2); No. 3: third deciduous molar (DM3); No. 4: first molar (M1); No. 5: second molar (M2); No. 6: third molar (M3); No. 7: second premolar (P2); No. 8: third premolar (P3); No. 9: first premolar (P1)). Teeth Nos. 1–4 were fully developed before the accident and Nos. 5–9 were developed after the FNPP accident.

2.2.2. Soil samples

All soil samples were air-dried to constant weight for 3–5 days and then passed through a 2-mm-mesh sieve.

Soluble species (^{90}Sr, strontium, and calcium) were extracted from the air-dried soils by using the method reported by Krouglov et al. (1998). The air-dried soil was vigorously shaken with Milli-Q water for 30 min at room temperature, and the suspension was left overnight. The solid-to-liquid mass ratio was 10 g:100 g or 5 g:50 g. The liquid phase (F1) was separated by vacuum filtration through a 0.45-μm membrane filter (JHWP02500, 47 mm diameter; Merck Millipore). The solid residues were subsequently treated with 1 M of CH_3COONH_4 solution (100 mL) and again filtered. This filtrate (F2) was added to F1, and the mixture was used for further analysis. The sum of solutions F1 and F2 was defined as “exchangeable fraction” in this study.

To determine strontium and calcium, 60 mg of air-dried soil were digested in a mixture of 6 mL of 61% HNO_3 (EL grade, Kanto Chemical), 3 mL of 49.5% HF (for atomic absorption spectrometry, Kanto Chemical), and 4 mL of 30% H_2O_2 (for atomic absorption spectrometry, Kanto Chemical) in a pressurized microwave digester (Speedwave MWS3 +; Analytik Jena) at 200 °C for 50 min.

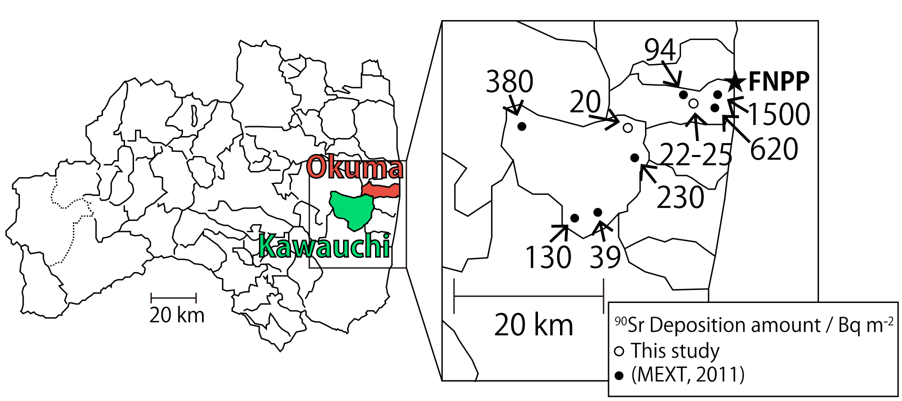

Fig. 1. Sampling sites. FNPP: The Fukushima-Daiichi Nuclear Power Plant. The site in Okuma is 5 km west of the FNPP. The site in Kawauchi is 16 km south-west of the FNPP. Okuma and Kawauchi were in the evacuation zone at the sampling period. Our results were corrected to the day of the accident (March 11, 2011). MEXT results were corrected to June 14, 2011. The maps were created with reference to open-access base maps freely available for public and academic use (source: http://maps.gsi.go.jp, from the Geographic Information Authority of Japan).