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Estimation of radiocesium dietary intake from time series data of radiocesium concentrations in sewer sludge

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ABSTRACT

After the Fukushima accident, it became important to determine the quantity of radionuclide ingested by inhabitants. The most common methods currently used to obtain such data are the "market basket" (MB) and "duplicate" (DP) methods. However, it is difficult to conduct monitorings using these methods with sufficient frequency as they are high cost and time-consuming. The present study proposes a new method to estimate the ingestion of radionuclides, based on the time-dependent concentrations of radiocesium in sewer sludge, which addresses the uncertainties of the two common methods. The newly proposed method, which we designate as SL, consists of three steps: (1) the separation of wet weather and dry weather data, (2) determining the mass balance of the wastewater treatment plant (WWTP), and (3) developing a reverse biokinetic model to relate the amount of radionuclides ingested to the amounts contained in the sewer sludge. We tested the new method using the time-dependent radiocesium concentrations in sewer sludge from the WWTP in Fukushima City. The results from the SL method agreed to those from the MB while overestimated those from DP method. The trend lines for all three methods, however, are in good agreement. Sensitivity analyses of SL method indicate further studies on uncertainties of sensitive parameters are deemed necessary to improve the accuracy of the method.

1. Introduction

The Fukushima Daiichi Nuclear Power Station (FDNPS) accident released numerous radionuclides into the environment and deposited them over a wide area surrounding the FDNPS. After the accident, public concern was raised about radiation exposure, including the dietary intake of short-half-life nuclides such as ^{131}I and long-half-life nuclides such as ^{137}Cs (Beresford et al., 2016). The internal dose resulting from the dietary intake pathway was found to account for about half of the total internal dose, according to a study conducted in the fifth month after the accident (Koizumi et al., 2012). This indicates that the dietary intake of radionuclides after a radiological emergency provides important information for assessing the internal exposure of the general public. However, the availability of such information is very limited, since the method is relatively complex compared with methods for determining the external exposure and the internal exposure from the inhalation pathway (Murakami and Oki, 2014).

Currently, there are two methods for determining the dietary intake of radionuclides. The first method, termed "market basket" (MB), is performed by measuring radionuclide activity in each type of foodstuff that is collected from local markets. The total dietary intake is then calculated based on the composition of nutritional data. This method was employed after the Chernobyl accident (Strand et al., 1999; Toader and Vasilache, 1996) as well as after the FDNPS accident (Koizumi et al., 2012; Tsutsumi et al., 2013). It is suitable for use when the radionuclide content in each food commodity is low (Hisamatsu et al., 1987). However, radionuclide loss during food processing and preparation is a large contributor to the uncertainty of this method. Radionuclide removal due to cooking varies over the range of 7–90%, according to the type of foodstuff (Hachinohe et al., 2015a; Hachinohe et al., 2015b) and in the range of 10–90%, depending upon the cooking method (Nabeshi et al., 2016; Perepellyatnikova et al., 1996).

To address the uncertainty in the MB method, one can apply the duplicate (DP) method, which samples the ready-to-eat daily meals from volunteers' homes instead of collecting samples of foodstuffs, and it determines the total dietary intake by direct measurements on the samples. The DP method also was widely used after both the Chernobyl accident (Cooper et al., 1992; Toader and Vasilache, 1996; Shiraishi et al., 2006) and the FDNPS accident (Harada et al., 2013). This method is more direct and eliminates the uncertainty caused by the cooking process. However, for determining the average dietary intake for a large population, the uncertainty of the DP method becomes high, because

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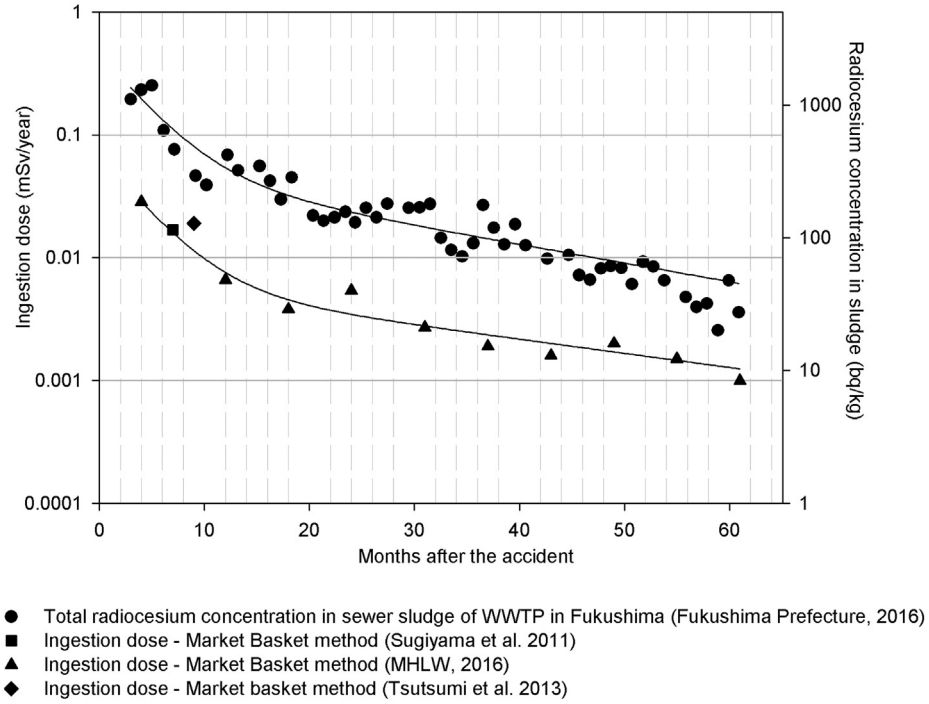

Fig. 1. Dietary intake of radiocesium (Sugiyama et al., 2011; MHLW, 2016; Tsutsumi et al., 2013) versus concentration in sewer sludge (Fukushima Prefecture, 2016). The solid line represents a linear regression. The value of Correlation coefficient (r) = 0.93 indicates a strong correlation between these two parameters.

the daily meal varies widely from one individual to another (Ohtsuka et al., 2013). In addition, the sampling activity cannot be done with sufficient frequency without causing public anxiety.

After the FDNPS accident, some of the radiocesium deposited on the surface of the urban area was redistributed into the sanitary sewer, as confirmed by the detection of radiocesium in sewer sludge in Fukushima Prefecture (Pratama et al., 2014). Moreover, most of the sewers in Fukushima Prefecture are parts of a separate system that only receives wastewater from households. In our previous study (Pratama et al., 2018), we found a strong correlation between the radiocesium concentration in sewer sludge and the dietary intake of radiocesium provided by the Ministry of Health, Labour and Welfare, as shown in Fig. 1. This suggests that some fraction of the radiocesium contained in the sewer sludge originates from the discharge of excreta. Studies by Roth et al. (2001) and Anderson et al. (2013), which determined the internal exposure by using radionuclide concentrations in data from urine and feces, suggest that if the portion of radiocesium in sewer sludge that originates from excreta can be quantified, it should be possible to develop a new method for estimating the time-dependent dietary intake of radiocesium.

There would be several benefits if this method could be successfully developed. As the sewer system serves > 90% of the population, this method can be expected to provide a more representative collective ingestion dose of radiocesium from a large population. Also, as the public has not so much interest in the facility, WWTP is typically an "out of sight" facility. Thus the sampling activity can be done with sufficient frequency without causing public anxiety. Finally, the sampling activity itself would be relatively simpler than the other two methods since it only requires surveyors to collect samples from one point. Hence, it will significantly reduce the cost.

In the present study, we developed a specific new method for estimating the dietary intake of radionuclides based on the time-dependent radiocesium concentration in sewer sludge (SL). The method includes a mass balance analysis of radiocesium in WWTP and an inverse biokinetic model of radiocesium in the human body. We compared the results from the SL method to those obtained using the DP and MB methods. Finally, we performed sensitivity and uncertainty analyses to determine the limitations of our method.

2. Methodology

In the present work, we have used the time dependence of the radiocesium (^{134}Cs and ^{137}Cs) concentration in sewer sludge data from the wastewater treatment plant (WWTP) in Fukushima City as a case study. We collected the daily data on the radiocesium concentration in dewatered sludge (the output of the WWTP) for the period from May 26, 2011, to May 30, 2016. We calculated the dietary intake of radiocesium from these data in three steps:

- Separation of wet weather and dry weather data
- Determination of the WWTP mass balance
- Utilization of a reverse biokinetic model

2.1. Separation of wet weather and dry weather data

Our previous study found that, in addition to human excretion, rain-derived inflow and infiltration (RDII) also contributes to the amount of radiocesium in sewer sludge (Pratama et al., 2018). RDII is the increased portion of wastewater flow into a sewer system that occurs during and after a rainfall or snowmelt event (EPA, 2008). Consequently, radiocesium on the ground surface, which is washed off by rainfall or snowmelt, also enters the sewer system. As a result, radiocesium from human excretion and that from RDII flow are mixed, affecting the accuracy of the estimation. To minimize the contribution of the radiocesium flux from RDII, we eliminated the radiocesium concentration data for rainy weather days (rainfall depth > 1 mm) from the main data. However, the RDII effect on the inflow rate could also be seen for several days following a rainy day. Thus, we also removed the data taken on dry weather days that meet the following criteria:

- Data taken before the fourth day after the last rainy day. The concentration of radiocesium in sewer sludge returns to the pre-rainfall level on the fourth day after the last rainy day. Several exceptions were applied to this rule, particularly during extreme weather