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Research article

Cesium and strontium loads into a combined sewer system from rainwater runoff

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ABSTRACT

In this study, combined sewage samples were taken with time in several rain events and sanitary sewage samples were taken with time in dry weather to calculate Cs and Sr loads to sewers from rainwater runoff. Cs and Sr in rainwater were present as particulate forms at first flush and the particulate Cs and Sr were mainly bound with inorganic suspended solids such as clay minerals in combined sewage samples. In addition, multiple linear regression analysis showed Cs and Sr loads from rainwater runoff could be estimated by the total amount of rainfall and antecedent dry weather days. The variation of the Sr load from rainwater to sewers was more sensitive to total amount of rainfall and antecedent dry weather days than that of the Cs load.

1. Introduction

Radionuclides were released into the environment during the nuclear accident at the Fukushima Daiichi Nuclear Power Plant (F1NPP) in March 2011. Deposition of such radionuclides as ^{134}Cs, ^{137}Cs, and ^{90}Sr was found on soil surfaces in eastern Japan (Matsunaga et al., 2013; Steinhauser et al., 2013), including Iwate Prefecture, which is more than 150 km north of the F1NPP. Radiocesium contamination higher than 60 Bq/m^2 was observed in some areas in Iwate prefecture on October 13, 2011 by the Ministry of Education, Culture, Sports, Science and Economy (2011). Some of the deposited radionuclides entered sewers with rainwater runoff, and then were transferred to the sewage sludge by the sewage treatment (Ishikawa et al., 2013; Tsushima et al., 2013). It has also been reported that the radiocesium concentrations in sewage sludge collected by combined sewer systems were higher than those in sewage sludge collected by separate sewer systems (Iwate Prefecture, 2015).

Rainwater runoff containing contaminants causes water pollution; various studies have been made about contaminants in rainwater runoff including evaluation of the contribution of different types of runoffs to biochemical oxygen demand (BOD), suspended solids (SS), and heavy metals in combined sewer systems (Gromaire et al., 2001), characterization of polycyclic aromatic hydrocarbons in stormwater runoff (Hwang and Foster, 2006), and runoff characteristics of pollutant loads associated with catchment area characteristics (Kim et al., 2001). Furthermore, since first flush has relatively high pollutant concentrations, there have been investigations of first flush phenomena. First flush load of SS depends on the maximum rainfall intensity, maximum inflow, rainfall duration and the antecedent dry period (Gupta and Saul, 1996). First flush effects of metal elements vary with rain intensity and types of surfaces such as parking lot and roof surfaces (Li et al., 2012). Heavy metals could be sorbed by SS because there were strong correlations between heavy metals and total SS in urban runoff (Djukić et al., 2016). Radionuclides, especially radiocesium, were spread through the air by the F1NPP accident, and they are known to be sorbed strongly by clay minerals in soil (Cornel, 1993; Ishikawa et al., 2008) and thus be retained on the soil surface for a long time (Koarashi et al., 2012). Therefore, when surface soil is carried into a sewer by rain, the radionuclides sorbed by the surface soil can be carried into the sewer as well. However, there has not been much research on Cs and Sr runoff flowing into sewers.

The objective of this study was to investigate stable Cs and Sr (^{133}Cs, ^{88}Sr) loads of sewers associated with rainwater runoff in

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order to know the behaviors of Cs and Sr deposited on surface ground as a first step to estimate radiocesium and radiostrontium loads to sewers in rain events. Combined sewage samples were taken with time in several rain events and dry weather, and then Cs and Sr loads of sewers from rainwater runoff were calculated. The ratios between dissolved and particulate forms of Cs or Sr in sewage were also observed.

2. Materials and methods

2.1. Sample collection

Sample collection was carried out at the Morioka high rate filtration plant (MHRFP). The plant serves the 6.24 km^2 Nakagawara catchment area situated in Morioka City, Iwate Prefecture. Half of the area is covered by a separate sewer system and the remaining area is covered by a combined sewer system. The area covered by the combined sewer system is an old commercial area and a residential area with a population density of about 7000 persons/km^2. Fig. 1 shows the sewer system in the catchment area. In dry weather, all of the sewage generated in the catchment area flows into the Tonan sewage treatment plant via the combined sewer system. In rainy weather, all of the sewage also flows into the Tonan sewage treatment plant when the flow rate is under a certain value. At a flow rate greater than the preset value, for example due to a heavy rain, the excess sewage flows into the MHRFP. Such excess sewage is generally discharged into rivers directly, which causes water pollution of the rivers. The MHRFP, which has been in operation since April 2013, treats the excess sewage to reduce the pollutant load to the Kitakami River, Yana River, and Nakatsu River. Its maximum sewage treatment capacity is 197,000 m^3/day.

Two types of samples were collected at the MHRFP: sanitary sewage in dry weather (hereafter, simply sanitary sewage), and combined sewage during rainy weather (combined sewage). The sanitary sewage was collected every hour from 9:15 on June 4 to 8:15 on June 5, 2014. Before this collection period it had not rained since May 27, 2014, which meant the collected samples did not contain rainwater.

The combined sewage contained sanitary sewage and rainwater during rainy weather. The combined sewage was collected seven times from June 2013 to October 2014 at the sampling point (the black circle in Fig. 1). Sewage sampling was started when sewage started to flow into the MHRFP when the flow rate of sewage exceeded the preset value because of the rainy weather. The sewage samples of 1 L each were taken every 10 or 20 min by an automatic water sampler set at the sampling point. Sample collection was finished when the inflow of sewage to the MHRFP was stopped or 24 samples had been collected which is the maximum number of sampling bottles that could be set in the automatic water sampler. Table 1 summarizes the sampling date, the sampling time, number of samples, and rain characteristics such as total amount of rainfall (R_{tot}), duration of rainfall (T_c), maximum rainfall intensity (I_{max}), and antecedent dry weather days before a 0.5 mm/h rainfall ($T_{0.5mm}$) and a 1 mm/h rainfall (T_{1mm}) for each rain event. The rainfall data were recorded as the total rainfall amount for each hour, i.e., 0:00–1.:00, 1:00–2:00, etc.

2.2. Analysis

Concentrations of SS and volatile suspended solids (VSS) in each sample were determined by a glass fiber filter paper (GS-25, ADVANTEC, Japan) method (Japan Sewage Works Association, 2012). VSS determination was done for only the samples collected in 2014 (5 rain events). Inorganic SS was calculated as the difference between SS and VSS. The analysis was done in triplicate.

Total Cs and Sr concentrations in each sample were determined. The samples were prepared based on the standard method for examination of wastewater (Japan Sewage Works Association, 2012). Five milliliters of concentrated HNO_3 was added to 100 mL of each sample in a glass beaker. The solution mixture was digested and evaporated on a hot plate at about 120 °C until the solution volume was reduced to around 15 mL. Then, 5 mL of concentrated HNO_3 and 5 mL of HCl were added to the solution. The beaker was covered with a watch glass and the solution mixture was digested on the hot plate at about 120 °C for 30 min. After the digested solution was diluted in a measuring flask to a total volume of 100 mL, the diluted solution was filtered through a 0.45 μm pore size membrane filter (Cellulose mixed ester, ADVANTEC, Japan). Cs and Sr concentrations in the filtrate were determined using an ICP atomic spectrometer (ICPE-9000, Shimadzu, Japan) and an ICP mass spectrometer (iCAP Qc, Thermo Scientific, USA). FAMIC-C-09, “fermented sewage sludge” (Food and Agricultural Materials Inspection Center) and BCR 176R “Incineration ash” (European Commission, Community Bureau of Reference) were used as reference materials for measurement validation.

Multiple linear regression analysis was done using the statistics software JPM 11 (SAS). A multiple regression equation was created with each element load from rainwater runoff (L_{run} [mg/event])) for

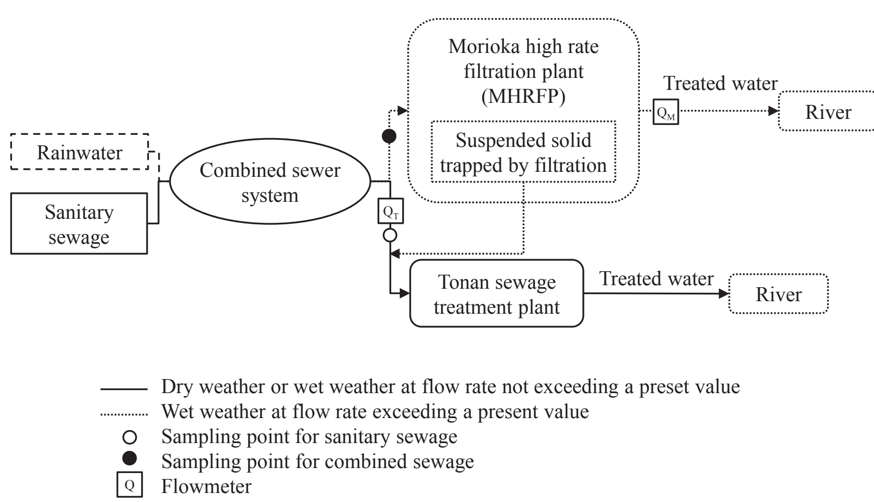

Fig. 1. Schematic drawing of sewer system in the area investigated in this study.