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Inventory estimation of ^{137}Cs in radioactive wastes generated from contaminated water treatment system in Fukushima Daiichi Nuclear Power Station

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ABSTRACT

Based on the analysis of data from contaminated water, the concentration of ^{137}Cs in radioactive waste such as used cesium adsorption vessels and sludge generated from the Cesium Adsorption Device and Second Cesium Adsorption Device and from the Decontamination Device, which were operating or suspended as a part of the contaminated water treatment system at the Fukushima Daiichi nuclear power station was calculated. An estimate is made of the total amount of ^{137}Cs recovered by decontamination from 28 June, 2011 to 12 August, 2014.

KEYWORDS

Fukushima Daiichi Nuclear Power Station, Contaminated water treatment, Radioactive waste, ^{137}Cs inventory estimation

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1. Introduction

In the process of decommissioning the Fukushima Daiichi nuclear power station[1], contaminated water accumulated in reactor buildings is decontaminated using water treatment systems such as a Cesium Adsorption Device and Second Cesium Adsorption Device and a Decontamination Device. A large amount of radioactive waste such as used cesium adsorption vessels and sludge was generated by these systems. A quantitative inventory of the radioactive waste is needed to progress in the timely processing and disposal of this waste. However, directly analyzing the waste is difficult because its radioactivity is extremely significant for humans to handle, and the contaminated water treatment devices are not equipped with sampling devices because of their urgent installation after the accident. Therefore, the waste inventory is estimated by analyzing contaminated water upstream and downstream of the water treatment devices. Because many analytical data regarding ^{137}Cs have now become available, the present study contributes to the effort of making a quantitative inventory of the waste by estimating the concentration of ^{137}Cs in the waste and the total amount of ^{137}Cs removed from contaminated water from 28 June, 2011 to 12 August, 2014.

2. Method to Determine ^{137}Cs Concentration in Waste and Decontamination Amount

2.1 Data

The data used consisted of the results of weekly analyses of radioactivity of contaminated water upstream and downstream of the water treatment devices [1,2] and weekly reports of the contaminated water treatment system [3]. These data detailed the amount of contaminated water that was treated and the amount of waste generated. The waste took the form of used adsorption vessels and sludge. The data were provided by the Tokyo electric power company.

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2.2 Adsorption vessels

For a given time period, usually one week, we determined the amount A_{ves} (Bq) of ^{137}Cs removed from the contaminated water by the Cesium Adsorption Device. The result for A_{ves} is expressed as $A_{ves} = (C_{in} - C_{out})V$, here C_{in} and C_{out} (Bq/m^3) are the ^{137}Cs concentrations in the contaminated water upstream and downstream of the Cesium Adsorption Device, respectively, and V is the volume in m^3 of water treated during the given time period. The total amount ΣA_{ves} of ^{137}Cs removed by the Cesium Adsorption Device is obtained by summing over A_{ves}. The concentration C_{ves} (Bq/vessel) of ^{137}Cs in each vessel is calculated by summing A_{ves} over the period that the vessel served in the device. The average concentration C_{vesav} of ^{137}Cs in a vessel over a given time period is calculated by dividing ΣA_{ves} by the number n of the adsorption vessels used over the given time period. The values for ΣA_{ves} and C_{vesav} for the Second Cesium Adsorption Device are calculated in the same way.

2.3 Sludge

A precipitate was generated by a Decontamination Device and was stored together with the contaminated water in a concrete pit (the precipitate could not be separated from the water). Hereinafter, the mixture of precipitate and contaminated water is called "sludge." Because precipitate and solution were both present in the sludge, the amount A_{slu} of ^{137}Cs equals the sum of the amount A_{pre} of ^{137}Cs in the precipitate and that in the solution, A_{sol}. The amount A_{pre} generated for a given water treatment period is calculated by $A_{pre} = (C_{din} - C_{dout})V$, where C_{din} and C_{dout} are the ^{137}Cs concentration in the contaminated solution upstream and downstream of the Decontamination Device, respectively, and V is the volume in m^3 of water treated. The amount A_{sol} is calculated by $A_{sol} = C_{dout}V_{slu}$, where V_{slu} is the volume of sludge generated. The sum ΣA_{slu} is obtained by summing over all A_{pre} and A_{sol}. The concentration C_{slu} of ^{137}Cs in the sludge is obtained by dividing ΣA_{slu} by the total volume ΣV_{slu} of sludge. However, because A_{pre} became much, much larger than A_{sol} due to the efficient transfer of ^{137}Cs from the contaminated water into the precipitate, A_{sol} can be neglected for calculating ΣA_{slu} and C_{slu}.

3. Determination of ^{137}Cs Concentration in Waste and Decontamination Amount

3.1 Adsorption vessels from Cesium Adsorption Device

Figure 1 plots the concentrations C_{in} and C_{out} of ^{137}Cs in the contaminated water upstream and downstream, respectively, of the Cesium Adsorption Device. These results are obtained from sampling data acquired from 17 June, 2011 to 12 August, 2014. The concentration of ^{137}Cs in the contaminated water decreased over this period; that is, the contaminated water was effectively decontaminated. The decontamination factor DF ($=C_{in}/C_{out}$) exceeded 10^4 over most of the operating period, and the Cesium Adsorption Device worked well. Although the downstream concentration C_{out} was reduced by six orders of magnitude with respect to the initial concentration after the accident, the upstream concentration C_{in} was only reduced by two orders of magnitude. Because the Cesium Adsorption Device had a good decontamination factor and because water free of ^{137}Cs was introduced in the reactor buildings, a lower concentration of cesium was expected in the accumulated water. However, the concentration was not reduced below 10^{10} Bq/m^3. This fact implies that from somewhere in the reactor buildings, ^{137}Cs is being supplied to some degree to the accumulated water.

The concentrations C_{in} and C_{out} are used to calculate the amount A_{ves} of ^{137}Cs removed. The concentration C_{ves} from all adsorption vessels used by the Cesium Adsorption Device is calculated from the weekly values for A_{ves} and C_{ves} and is plotted in Fig. 2 together with numerous adsorption vessels used each week. When more than one vessel is used in a given week, the concentration C_{ves} is calculated by taking the mean value for that week. During the initial period after the accident, many adsorption vessels were used per week, and the concentration was more than ten times its value for later periods. The concentration C_{ves} decreased with time due to the progress of decontamination, as shown in Fig. 1. During the period studied, 514 vessels were used.

Figure 3 plots the sum ΣA_{ves} for the same period. Although ΣA_{ves} sharply increased in the initial period after the accident, the increase after October 2011 was much less. This result is also attributed to the progression of ^{137}Cs decontamination of contaminated water. Based on these results, the amount ΣA_{ves} is calculated to be 1.7×10^{17} [Bq], which accounts for 99.6% of the total amount of ^{137}Cs input in the Cesium Adsorption Device. In addition, the concentration C_{vesav} is 3.3×10^{14} Bq/vessel during this period.

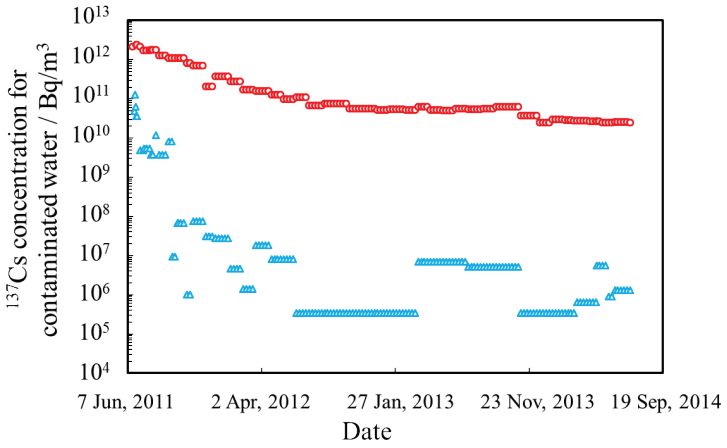

Fig. 1. Evolution of ^{137}Cs concentration in contaminated water upstream and downstream of Cesium Adsorption Device. Red circles (○) are C_{in} and blue triangles (△) are C_{out}.

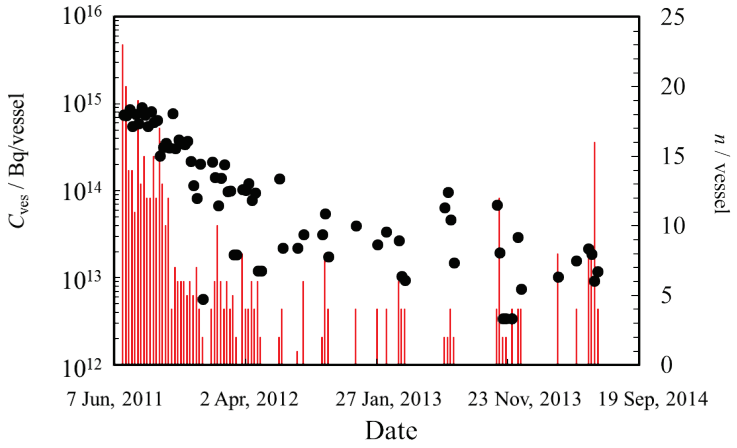

Fig. 2. ^{137}Cs concentration in used adsorption vessel (points) and numerous vessels (bars) used in Cesium Adsorption Device.

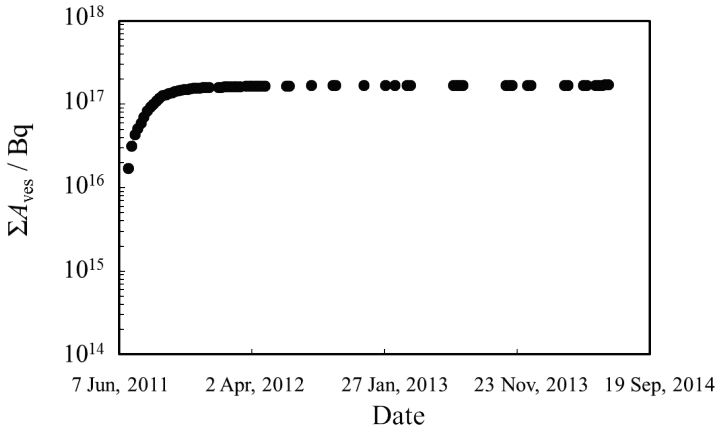

Fig. 3. Evolution of amount ΣA_{ves} in Cesium Adsorption Device.

3.2 Adsorption vessel from Second Cesium Adsorption Device

Figure 4 shows the concentrations C_{in} and C_{out} in the Second Cesium Adsorption Device plotted from 19 August, 2011 to 12 August, 2014. This C_{in} is slightly different from that of Fig.1 because water source of the Cesium Adsorption Device is different from that of the Second Cesium Adsorption Device [1]. The concentrations C_{in} and C_{out} are used for calculating the amount A_{ves} of ^{137}Cs removed by the Second Cesium Adsorption Device. The Second Cesium Adsorption Device also had a high decontamination factor (approximately 10^5). As is the case for the results shown in Fig. 1, the concentration C_{in} did not decrease below 10^{10} Bq/m^3.

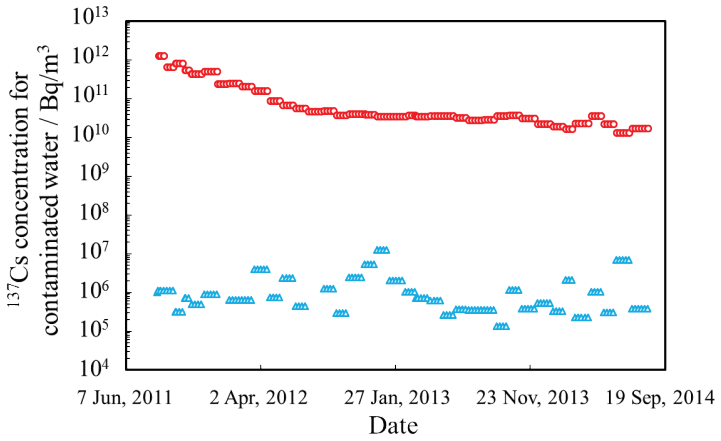

Fig. 4. Evolution of ^{137}Cs concentration in contaminated water upstream and downstream of Second Cesium Adsorption Device. Red circles (○) are C_{in} and blue triangles (△) are C_{out}.

The concentration C_{ves} for all adsorption vessels used in the Second Cesium Adsorption Device were calculated and are plotted in Fig. 5 for each week together with numerous adsorption vessels used each week. The trend of C_{ves} is similar to that observed for the Cesium Adsorption Device. However, 110 vessels were changed out during this period, which is less than that for the Cesium Adsorption Device.

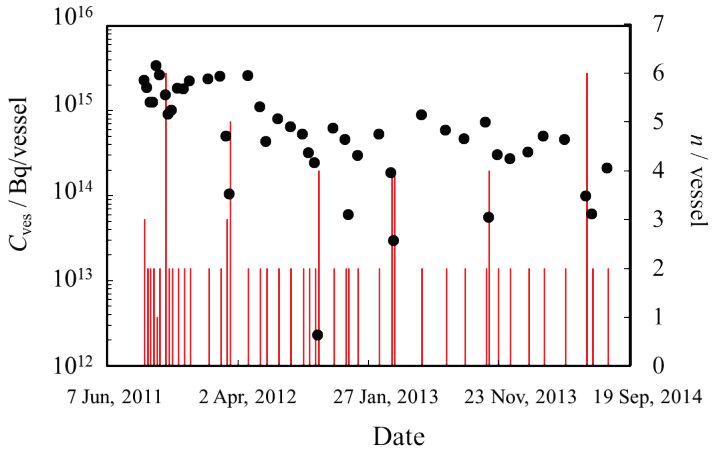

Fig. 5. ^{137}Cs concentration in used adsorption vessels (points) and numerous vessels (bars) for Second Cesium Adsorption Device.

The amount ΣA_{ves} is plotted for the same period in Fig. 6. The trend is also similar to that observed for the Cesium Adsorption Device; the rapid increase in ΣA_{ves} changes to a slow increase after January 2012. Based on these results, the amount ΣA_{ves} for the Second Cesium Adsorption Device is calculated to be 9.0×10^{16} Bq, which accounts for over 99.9% of the total amount of ^{137}Cs input amount into the

Second Cesium Adsorption Device. In addition, the concentration C_{vesav} was 8.1×10^{14} Bq/vessel during this period.

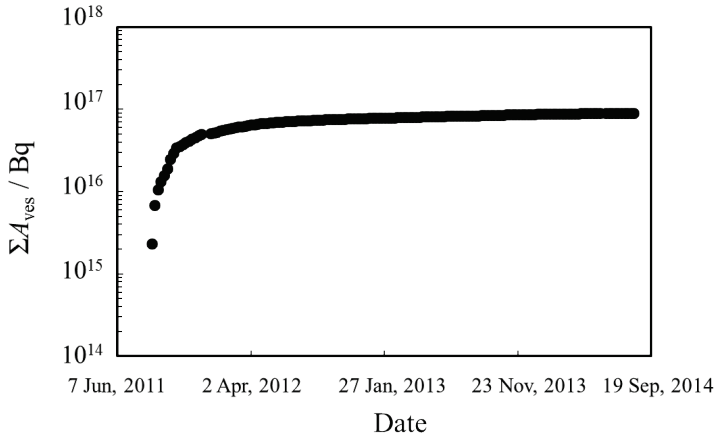

Fig. 6. Evolution of amount ΣA_{ves} in Second Cesium Adsorption Device.

3.3 Sludge from Decontamination Device

The concentrations C_{din} and C_{dout} in the Decontamination Device are plotted in Fig. 7 from 22 June, 2011 to 13 September, 2011. The concentration C_{din} was rather less than C_{in} for the Cesium Adsorption Device and Second Cesium Adsorption Device because the Decontamination Device was introduced downstream of the Cesium Adsorption Device[1]. The decontamination factor for the Decontamination Device is also high, i.e., approximately 10^4.

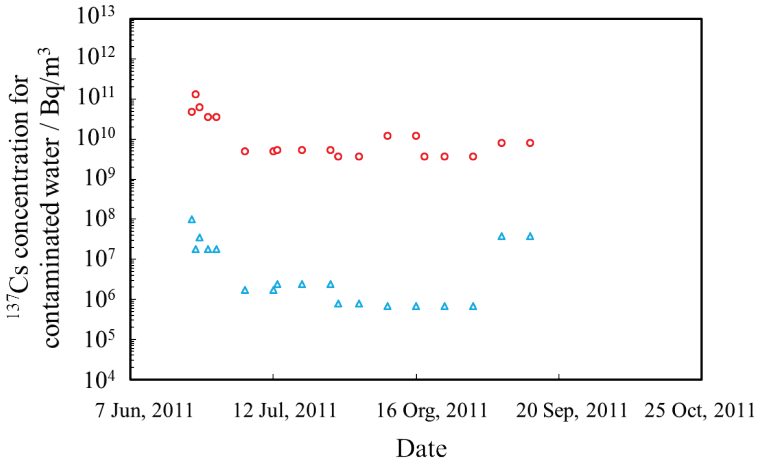

Fig. 7. Evolution of ^{137}Cs concentration in contaminated water upstream and downstream of the Decontamination Device. Red circles (○) are C_{din} and blue triangles (△) are C_{dout}.

The amount ΣA_{slu} is plotted in Fig. 8 for the same period as for Fig. 7. The total is 8.0×10^{14} Bq. This value accounts for more than 99.9% of the total amount of ^{137}Cs input. The Decontamination Device was not used since 16 September, 2011, and the amount ΣA_{slu} was fixed. When in operation, the Decontamination Device treated water downstream of the Cesium Adsorption Device. Consequently, ΣA_{slu} is two orders of magnitude less than ΣA_{ves} (Fig. 3). The treated volume ΣV_{slu} was 579 m^3 during this period, which gives $C_{slu} = 1.4 \times 10^{12}$ Bq/m^3.

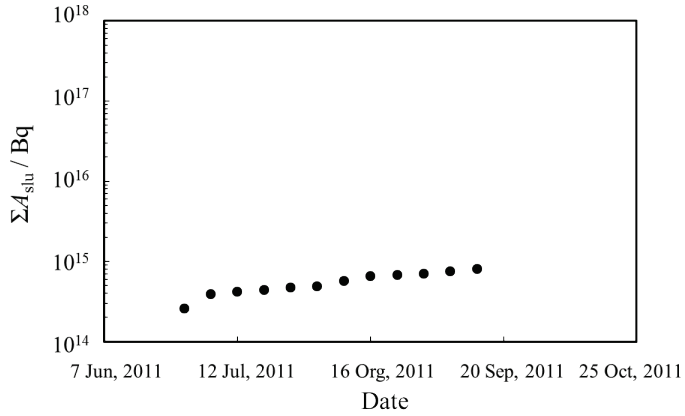

Fig. 8. Evolution of ΣA_{slu} in Decontamination Device.

3.4 Total amount of ^{137}Cs removed

Based on the results of the sums of ^{137}Cs removed by the three devices, the total amount of ^{137}Cs removed by the overall water treatment system between 28 June, 2011 and 12 August, 2014 is estimated to be 2.6×10^{17} Bq, which corresponds to approximately 37% of the ^{137}Cs contained in the fuels of reactors 1–3 of the Fukushima Daiichi nuclear power station, that is, 7.0×10^{17} Bq(100 %) [4]. However, the amount of ^{137}Cs released in the atmosphere and ocean is estimated to be 1.0×10^{16} Bq (approximately 1%) [5] and 3.6×10^{15} Bq (approximately 1%) [6], respectively, and that remaining in the accumulated water is estimated to be 9.1×10^{15} Bq (approximately 1%) [2]. Thus, 4.2×10^{17} Bq (approximately 60%) of the ^{137}Cs remains somewhere in the reactor building is estimated.

4. Conclusion

In this study, as a part of the inventory estimation, we estimate the concentration of ^{137}Cs in radioactive waste such as used cesium adsorption vessels and sludge and the total amount of ^{137}Cs removed by those devices between 28 June, 2011 and 12 August, 2014. In the future, an estimate of the inventory of other radioactive nuclides in that waste will also be attempted.

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