A study on the run-off of dissolved ^{137}Cs from contaminated slopes to an irrigation canal after the Fukushima Dai-ichi Nuclear Power Plant accident

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After the large earthquake and the tsunami disaster hit north-east Japan on March 11, 2011, a variety of radionuclides were released from the Fukushima Dai-ichi Nuclear Power Plant (FDNPP) accident. Among the nuclides, ^{137}Cs has been regarded to have persistent effects on the environment from its relatively long half-life (30.1 y). In the aquatic environment, there are two forms of radiocesium: a 'particulate' form that is strongly combined with suspended sediment, and a 'dissolved' form. Preceding studies reported that there was a tendency of radiocesium in river water to redistribute mostly in the particulate fraction, which was suggested to be a consequence of strong adsorption of radiocesium by clay minerals in soil or sediment in rivers. Nevertheless, dissolved radiocesium may not be ignorable, because it can be imbibed by plants more efficiently than that in particulate form, thereby accumulating in the ecosystem and circulating biologically. Although dissolved radiocesium has been still constantly flowing out from catchments even in a small but certain amount under base-flow condition, the major sources of it have not been completely confirmed.

We focused on the possible risk of remaining ^{137}Cs in the environment around paddy fields. Severe contamination of radiocesium in paddy fields has been removed by the decontamination project. However, slopes of agricultural canals and bunds of paddy fields have not been subject to the decontamination. Since soil can strongly adsorb radiocesium as was mentioned above, litter covering the slopes probably plays a substantial role in supplying dissolved ^{137}Cs.

The objectives of this study were to (1) clarify the trend in the discharge and the redistribution of ^{137}Cs from contaminated slopes along an agricultural canal by directly collecting the overland flow, and (2) analyze the possible mechanisms of the discharge of dissolved ^{137}Cs by investigating and partitioning the forms of ^{137}Cs in the litter samples from the slopes.

The sample of the overland flow was filtered through glass filters and 0.45 μm membrane filters. ^{137}Cs in the filtrate was defined as a dissolved fraction, while in the residue as a particulate fraction. ^{137}Cs in the litter samples were partitioned by employing the sequential extraction method. ^{137}Cs extracted by deionized water (H_2O) was defined as a water-soluble fraction, by ammonium acetate (CH_3COONH_4) as an ion-exchangeable fraction, by alkalis ($Na_2P_4O_7$, NaOH) as a fraction that associated with humic and fluvic acid, and hydrogen peroxide (H_2O_2) as a fraction that associate with other organic matters, such as lignin.

The results showed that dissolved ^{137}Cs constantly and stably ran out of the catchments, while a large proportion of particulate ^{137}Cs ran out exclusively under storm conditions. The annual discharges of dissolved radiocesium ranged between 0.063 –4.7 Bq m^{-2} y^{-1}, and K_d values (ratio of particulate to dissolved radioactivity concentrations) under base-flow condition were in the order of 104 L kg^{-1}, which were less than the geometric mean of the K_ds for forest streams and major rivers reported in the preceding studies.

Storages of currently water-soluble ^{137}Cs in the litter of the slopes were estimated to be 2.2 –32 Bq m^{-2}, and the surface run-off rates of precipitation were 1.8 –8.3%. From these observed values, the discharges of dissolved ^{137}Cs from the catchments were estimated to be 0.040 –2.0 Bq m^{-2} y^{-1}, which roughly corresponded to the observed values. Because there were also ^{137}Cs in the ion-exchangeable form or forms associating with the humic substances, they have potentially become a water-soluble form after

each runoff event to keep the concentration of dissolved ^{137}Cs within the observed range for years. This evidence and discussion suggest that water-soluble ^{137}Cs in the litter layer on the catchments might be one of the major sources of dissolved ^{137}Cs that has been discharged from the contaminated paddy environment.

Keywords: Fukushima Dai-ichi Nuclear Power Plant accident, Radiocesium, Paddy fields, Sequential extraction

