

Study on behaviour of radiocaesium and tritium at Fukushima Coast

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Radiocaesium (^{134}Cs and ^{137}Cs) released by the TEPCO Fukushima Dai-ichi Nuclear Power Plant (FNPP 1) accident that occurred in March 2011 was injected directly into the North Pacific Ocean via the atmosphere or directly discharged as contaminated water. It is considered that tritium (^3H) is also released into the atmosphere as water, then enters the ocean through precipitation or river water and enters the water circulation by evaporation from the surface of the ocean. In addition, ^3H is directly injected into the ocean as contaminated water, it is considered that it also enters the water circulation like ^3H that entered the ocean via the atmosphere. There is well known tritium source other than FNPP1 accident derived tritium in our environment. Cosmogenic tritium is big source term, too. In this presentation, radiocaesium and ^3H activity concentrations on the coast of Fukushima obtained in 2014, $^3\text{H}/^{137}\text{Cs}$ activity ratio and $^{134}\text{Cs}/^{137}\text{Cs}$ activity ratio were examined and discuss about behavior of radiocaesium and tritium.

At Tomioka, $^{134}\text{Cs}/^{137}\text{Cs}$ activity ratios ranged from 0.31 ± 0.03 to 0.35 ± 0.03 and were similar with $^{134}\text{Cs}/^{137}\text{Cs}$ activity ratios observed at the station FNPP1-T-1 where those ranged from 0.29 to 0.40. $^{134}\text{Cs}/^{137}\text{Cs}$ activity ratio off Fukushima stations ranged from 0.24 ± 0.03 to 0.37 ± 0.03 . The consistency of $^{134}\text{Cs}/^{137}\text{Cs}$ activity ratio in these regions are clearly shown. An activity ratio of 0.355 for $^{134}\text{Cs}/^{137}\text{Cs}$ activity ratio means that $^{134}\text{Cs}/^{137}\text{Cs}$ activity ratio at the time of accident, 11 March 2011, was 1.0 which is similar with $^{134}\text{Cs}/^{137}\text{Cs}$ activity ratio in the core inventory at FNPP1 at the time of the accident [19,20]. This indicates that major source of radiocaesium observed at Tomioka, off Fukushima stations and the station FNPP1-T-1 should be FNPP1 accident derived radiocaesium and originated from the same source.

^3H activity concentrations at Tomioka and Hasaki were $175 \pm 14 \text{ Bq m}^{-3}$ in June 2014 and $57 \pm 12 \text{ Bq m}^{-3}$ in August 2014, respectively. In contrast with strong gradient of ^{137}Cs activity concentrations in surface water between FNPP1-T-1 and off Fukushima stations as stated previously, ^3H activity concentrations among the off Fukushima stations including Tomioka and Hasaki which located north and south of FNPP1 showed relatively homogenous as around 60 Bq m^{-3} to 200 Bq m^{-3} during the period from May 2014 to September 2014. Only exception was observed at FNPP1-T-1 station and ^3H activity concentrations exceed 1000 Bq m^{-3} . It is also known that ^3H activity concentration in precipitation is relatively ^3H rich rather than ^3H activity concentrations in seawater due to cosmogenic ^3H of which activity concentration ranged $180 - 1000 \text{ Bq m}^{-3}$ in precipitation globally and observed ^3H activity concentrations in Tomioka river were $590 - 700 \text{ Bq m}^{-3}$ during the period from Dec. 2013 to Dec. 2015 and ^3H activity concentrations in several rivers at Fukushima region also showed similar level around several 100 Bq m^{-3} by monitoring of Fukushima Prefecture, and $690 \pm 20 \text{ Bq m}^{-3}$ on 19 January 2015 (Aoyama unpublished data). This homogenous distribution of ^3H activity concentrations in this interested region might indicate that contribution from ^3H rich water from surrounding rivers located north and south of FNPP1 site was larger rather than flux of ^3H from FNPP1 site.

On the other hand $^3\text{H}/^{137}\text{Cs}$ activity ratio at Tomioka was 2.0 ± 0.2 in June 2014, however $^3\text{H}/^{137}\text{Cs}$ activity ratio at station FNPP1-T-1 ranged from 2.2 to 15.3. $^3\text{H}/^{137}\text{Cs}$ activity ratio at off Fukushima stations ranged from 1.1 ± 0.1 to 17.1 ± 2.1 . The $^3\text{H}/^{137}\text{Cs}$ activity ratio at Tomioka is close to lower side of $^3\text{H}/^{137}\text{Cs}$ activity ratio at station FNPP1-T-1. The physical and chemical characteristics of ^3H are quite

different from those of caesium of which form in seawater is dissolved but cannot evaporate from surface of seawater. ^3H might exist as HTO and HTO can move by evaporation and precipitation through seawater surface. During the transportation processes in the ocean and also possible different source of highly contaminated water from FNPP1 site, reasons of the observed variability of $^3\text{H}/^{137}\text{Cs}$ activity ratio in this region might be complex. One possibility of this variability after released from FNPP1 site may be evaporation.

Keywords: radiocaesium, tritium, Fukushima Dai-ichi Nuclear Power Plant accident, cosmogenic tritium, river water