Why there are variations in the $^{134}Cs/^{137}Cs$ activity ratios of the Global data set of Fukushima-derived radiocesium?

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The contrasting geochemical behaviour of radiocesium in marine and freshwater environment have resulted in very different applications: as a tracer in marine system due to its conservative behaviour and as a chronometer due to the particle-reactive nature in freshwater system. One of the key applications of $^{134}Cs/^{137}Cs$ ratios (^{134}Cs $t_{1/2}$ = 2.065 years; ^{137}Cs $t_{1/2}$ = 30.07 years) in marine system is the transit time estimate of a parcel of water between two points along its pathway. The time scale involved in the advective transport from the point of origin (e.g., nuclear reprocessing plants in Sellafield) to the Arctic Ocean and advective time for transport around the perimeter of the Arctic Ocean from the Santa Ana Trough to the southern Canada Basin obtained from $^{134}Cs/^{137}Cs$ activity ratios is comparable to the values obtained from $^{3}H/^{4}He$ and chlorofluorocarbon data. Furthermore, the $^{134}Cs/^{137}Cs$ ratios in environmental samples also can be utilized to delineate the amount of ^{137}Cs derived from Fukushima accident and the amount derived from global fallout. These applications require precise value of the $^{134}Cs/^{137}Cs$ activity ratio at the source of origin.

While the $^{134}Cs/^{137}Cs$ ratios in environmental samples collected after other major nuclear accidents is well-defined (e.g., Chernobyl: 0.4), with a relatively narrow range, the range of values reported from precipitation and air samples around the globe shortly after Fukushima Dai-ichi nuclear power plant (NPP) accident vary widely, from 0.2 to 2.4. In contrast, the seawater samples collected within 20 km from the Dai-ichi NPPs indicate the $^{134}Cs/^{137}Cs$ activity ratios are extremely uniform with a value of 0.99 ± 0.03 (Buesseler *et al*., 2011). It is known the source of ^{137}Cs released from spent nuclear fuel depends on the reactor design, fuel cycle and age. From a review of the global data on the $^{134}Cs/^{137}Cs$ ratios derived from Fukushima accident, we will summarize the results and present a set of hypotheses for the variations of the activity ratios.

[1] Buesseler, K.O., M. Aoyama and M. Fukasawa (2011) Impacts of the Fukushima Nuclear Power Plants on Marine Radioactivity. *Environ. Sci. Technol.* **45**, 9931-9935