Temporal variations and future estimations of ^{90}Sr and ^{137}Cs in atmospheric depositions after the Fukushima Daiichi Nuclear Power Plant accident with 63 years of continuous observations

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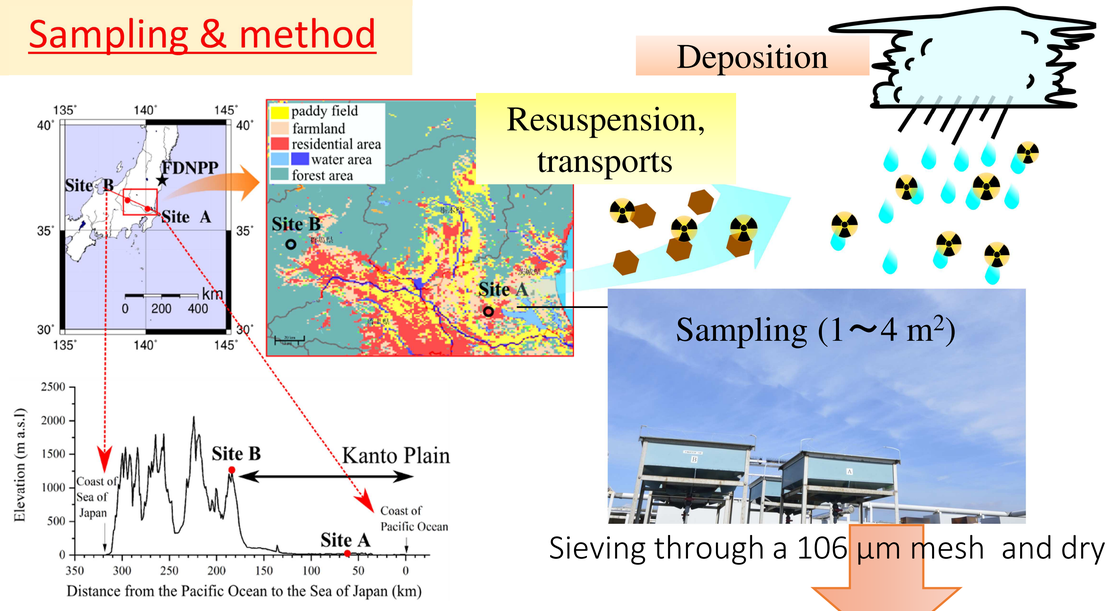
Introduction

- We have measured the artificial radionuclides, such as ^{90}Sr and ^{137}Cs, in atmospheric depositions since 1957 in the Kanto areas around Tokyo, Japan (site A) and since 2007 at a top of the mountain in the corner of the Kanto plain (site B). As the result, we clarified the variations in ^{90}Sr and ^{137}Cs, which were emitted from atmospheric nuclear tests and nuclear power plant accidents, and their environmental processes due to their diffusion, deposition, and resuspension.
- In this study, we show our long-term observation results of ^{90}Sr and ^{137}Cs in monthly atmospheric deposition samples and estimate the current environmental processes and decay periods of ^{90}Sr and ^{137}Cs with measurements of ^{134}Cs and stable elements and isotopes (Na, Mg, Al, K, Ca, Ti, Mn, Fe, Ni, Cu, Zn, Sr, Ba, ^{9}Be, ^{133}Cs, ^{232}Th, and ^{238}U).

Sampling & method



Measuring $^{134\&137}Cs$ by Ge semiconductor detectors (coaxial type from ORTEC EG&G and Eurisys)

Measuring stable elements (Na, Mg, Al, K, Ca, Ti, Mn, Fe, Ni, Cu, Zn, Sr, and Ba) and isotopes (^{9}Be, ^{133}Cs, ^{232}Th, and ^{238}U) by inductively coupled plasma atomic emission spectrometry (CIROS-120, Rigaku Corp., Japan, or Vista-PRO, Varian Inc., USA) and inductively coupled plasma mass spectrometry (Agilent7500c or Agilent8000, Agilent, Ltd., USA)

Measuring ^{90}Sr by alpha/beta counting system (Tennelec LB5100, Mirion Technologies, USA)

Figure 1. Location of observation sites and procedures of sampling and analysis.

Conclusion

- Activity levels in atmospheric depositions at site A (Bq m^{-2})…
  - ^{90}Sr… Atmospheric nuclear test > FDNPP > Chernobyl >> Just before the FDNPP ≒ latest (2018)
  - ^{137}Cs… FDNPP > Atmospheric nuclear test > Chernobyl > latest (2018) >> Just before the FDNPP
  - Activity levels of ^{90}Sr returned to the preaccident level. On the other hand, those of ^{137}Cs is still ~400 times higher than the preaccident levels. These values were same level as those of 1983.
  - Seasonal variations of ^{90}Sr at sites A and B showed the similar trend to the preaccident period.
- Resuspension process…
  - Site A… Mineral dusts from the neighboring surface and the remote area hosted ^{90}Sr and ^{137}Cs.
  - Site B… Forest ecosystem dominated ^{90}Sr cycle, but the environmental process of ^{137}Cs cycle could not be clarified.
- Future estimation…
  - The present environmental half-live of ^{137}Cs at sites A and B were estimated as 4.7 and 5.9 years, respectively.
  - Approximately 42 and 48 years are required to reduce the atmospheric ^{137}Cs deposition rate from 2011.

Result

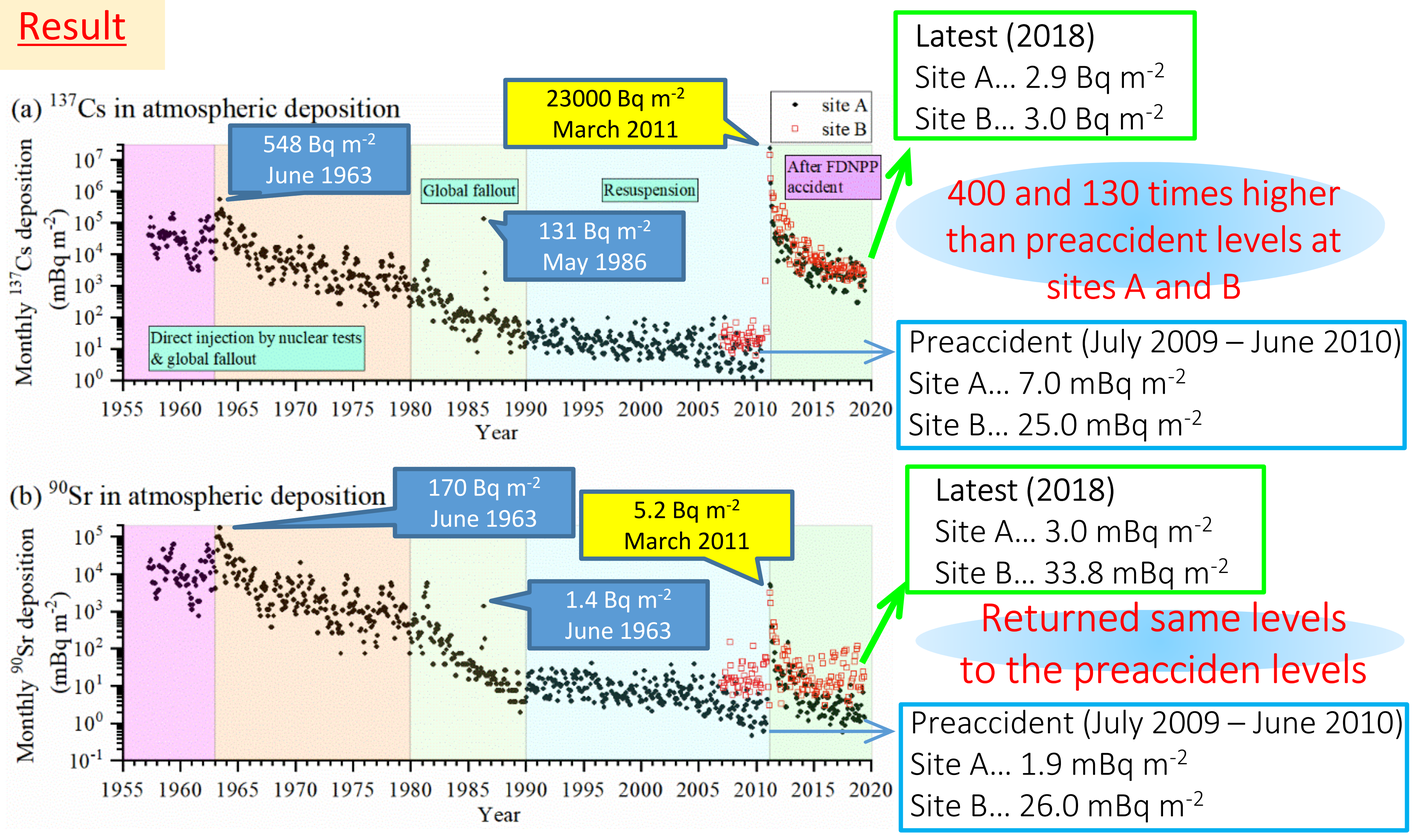


Figure 2. Observation results of (a)^{137}Cs and (b)^{90}Sr in atmospheric depositions (mBq m^{-2}) from 1957 to 2019 at site A (closed black circles) and that after 2007 at site B (open red squares).

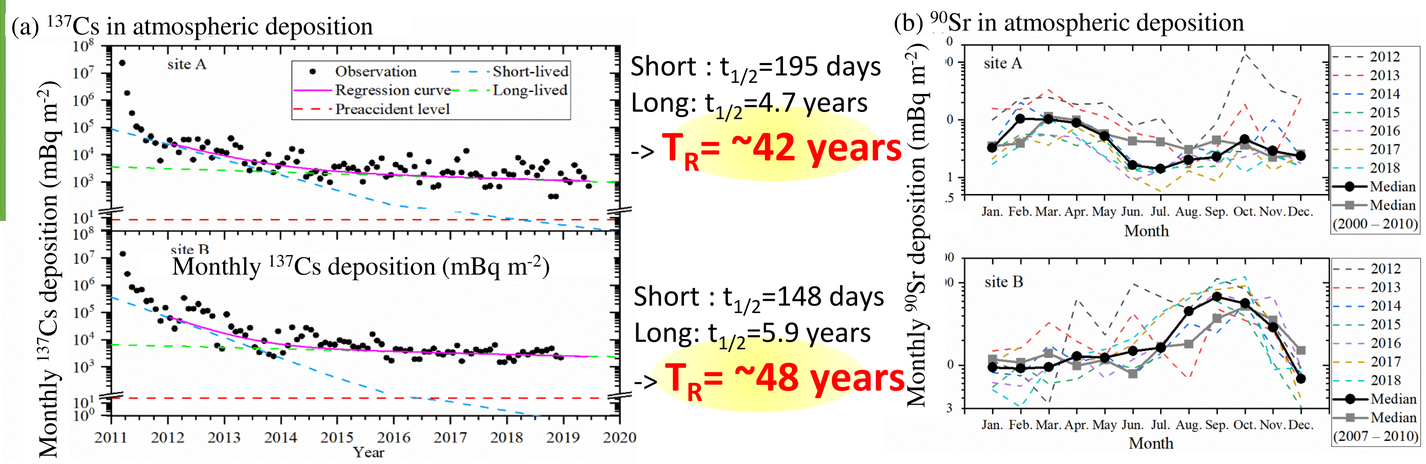


Figure 3. (a)Time series of ^{137}Cs and (b) seasonal changes of ^{90}Sr atmospheric deposition after the FDNPP accident. We adopted a multiple exponential function (short and long lived components) after the accident (2012–2018; the resuspension phase).

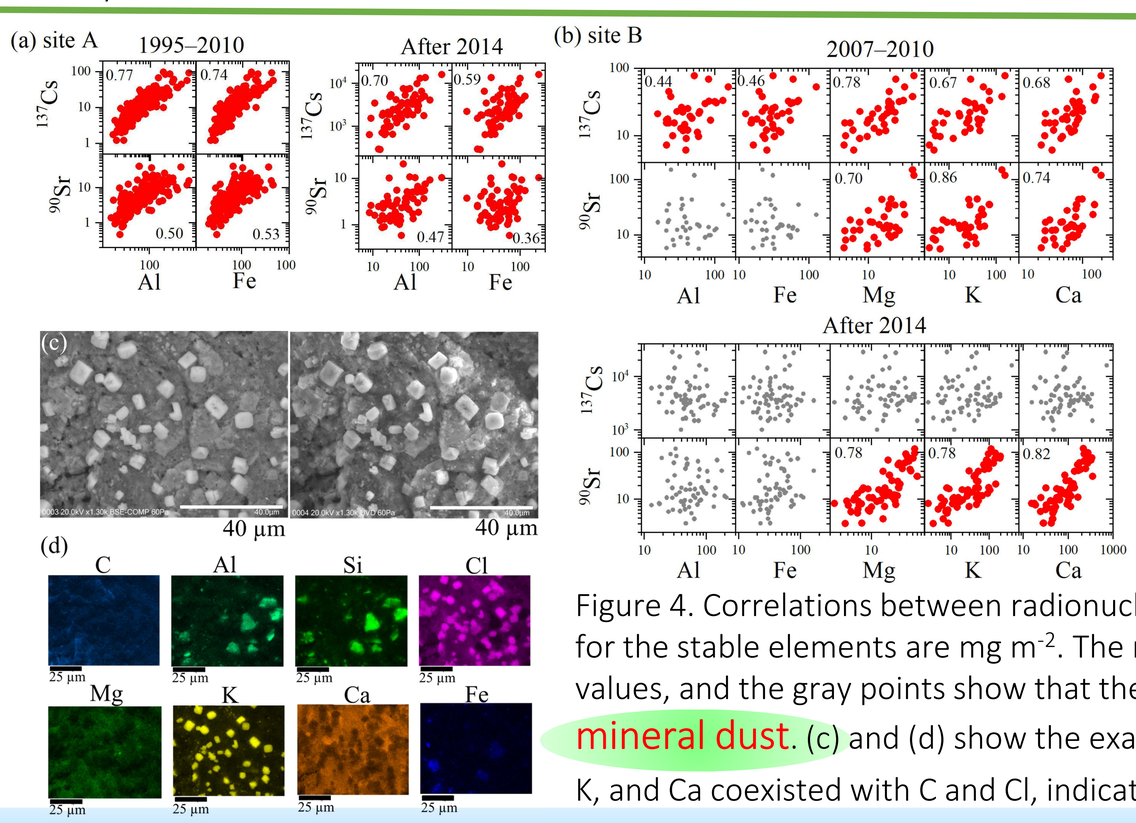


Figure 4. Correlations between radionuclides and stable elements at sites (a) A and (b) B. The units for ^{90}Sr and ^{137}Cs are mBq m^{-2}, and those for the stable elements are mg m^{-2}. The red points reveal that the correlations are significant ($p<0.05$) based on the correlation coefficient values, and the gray points show that the correlations are not significant ($p\geq0.05$). Aluminum and Fe are recognized as the tracers of the mineral dust. (c) and (d) show the example of the electron microscopic analysis of deposition samples (site B, October 2016). Magnesium , K, and Ca coexisted with C and Cl, indicating that they were salt and organics materials and the leaching from the leaves contributed.

Reference: Kinase et al. Sci Rep 10, 21627 (2020). https://doi.org/10.1038/s41598-020-78312-3