Strontium-90 contamination, analysis and difficulties related to the Fukushima nuclear accident


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Sr-90 has a significant human health risk due to its long biological and physical half-life and accumulation in bone tissue wherein high-energy beta particles emitted by its daughter nuclide, Y-90, can cause bone marrow damage.
Features of radiostrontium isotopes

Strontium is an alkali earth metal showing biological and chemical similarity to calcium. It has four stable natural occurring isotopes ($^{84}$Sr, $^{86}$Sr, $^{87}$Sr, $^{88}$Sr) and manifold artificially produced radioactive isotopes.

In radioecologically view $^{89}$Sr and $^{90}$Sr have importance and they are called together as radiostrontium isotopes.

<table>
<thead>
<tr>
<th>Radiostrontium Isotopes</th>
<th>$^{89}$Sr</th>
<th>$^{90}$Sr</th>
</tr>
</thead>
<tbody>
<tr>
<td>Parents isotopes</td>
<td>$^{89}$Kr ($^{89}$Rb)</td>
<td>$^{90}$Kr ($^{90}$Rb)</td>
</tr>
<tr>
<td>Daughter isotopes</td>
<td>$^{89}$Y, stable</td>
<td>$^{90}$Y ($^{90}$Zr, stable)</td>
</tr>
<tr>
<td>Half-life</td>
<td>50.5 d</td>
<td>28.8 y</td>
</tr>
<tr>
<td>Decay Mode</td>
<td>$\beta^-$</td>
<td>$\beta^-$</td>
</tr>
<tr>
<td>Max. (mean) Beta Energy keV</td>
<td>1495(583)</td>
<td>546 (196)</td>
</tr>
<tr>
<td>Cumulative Fission Yield (%)</td>
<td>4.73</td>
<td>5.77</td>
</tr>
</tbody>
</table>

(Thermal neutron fission of $^{235}$U)
Source of Radiostrontium isotopes

Radioactive radiostrontium isotopes are produced by neutron fission of heavy mass weight U or Pu isotopes.

Source of environmental contamination:

- Nuclear weapon testing  
  (worldwide contamination)
- Operation and accident of nuclear facilities  
  (local contamination)
- Operation of reprocessing plants  
  (local contamination)

Sources of pictures: http://www.batguano.com/nuclear/39.jpg  
http://www.abovetopsecret.com/forum/thread674830/pg1
Presence of Radiostrontium isotopes in the Environment

• The appearance of anthropogenic radionuclides in the global environment began with the development of nuclear weapons in 1945.

• The northern hemisphere has a higher $^{90}$Sr deposition ratio since many more tests were completed there in comparison to the southern hemisphere. From the late 1990s onwards, the stratospheric fallout contribution became negligible and instead re-suspension has played an important role in deposition.
Radiostrontium background in Japan

Background results of Sr-90 at Okuma Town, Fukushima Prefecture, Japan

16th nuclear explosion test carried out by the People’s Republic of China

- Wet and dry fallout (MBq km-2) Average: 0.03 (except the data of 1974)
- Tap water (mBq L-1) Average: 2.1
- Soil 0-5 cm (Bq kg-1) Average: 3.7
Present source of radiostrontium isotopes in Japan

The radiostrontium isotopes inventory of the Fukushima units affected by the accident and Chernobyl Nuclear Power Plant (CNPP)

<table>
<thead>
<tr>
<th>Units</th>
<th>89Sr PBq</th>
<th>90Sr PBq</th>
<th>89Sr/90Sr</th>
</tr>
</thead>
<tbody>
<tr>
<td>Reactor Core Unit 1</td>
<td>1360</td>
<td>150</td>
<td>9.07</td>
</tr>
<tr>
<td>Reactor Core Unit 2</td>
<td>2210</td>
<td>191</td>
<td>11.57</td>
</tr>
<tr>
<td>Reactor Core Unit 3</td>
<td>2350</td>
<td>181</td>
<td>12.98</td>
</tr>
<tr>
<td>Total</td>
<td>5920</td>
<td>522</td>
<td>11.21</td>
</tr>
<tr>
<td>Spent Fule Pool Unit 1</td>
<td>1.57</td>
<td>114</td>
<td>0.01</td>
</tr>
<tr>
<td>Spent Fule Pool Unit 2</td>
<td>27.9</td>
<td>306</td>
<td>0.09</td>
</tr>
<tr>
<td>Spent Fule Pool Unit 3</td>
<td>15.3</td>
<td>269</td>
<td>0.06</td>
</tr>
<tr>
<td>Spent Fule Pool Unit 4</td>
<td>670</td>
<td>619</td>
<td>1.08</td>
</tr>
<tr>
<td>Total</td>
<td>715</td>
<td>1310</td>
<td>0.31</td>
</tr>
<tr>
<td>CNPP</td>
<td>2300</td>
<td>200</td>
<td>11.5</td>
</tr>
</tbody>
</table>

The total Fukushima inventory was almost two and half times higher than Chernobyl’s and the 89Sr/90Sr ratio was very similar at around 12. In the Spent Fule Pools, where there were no further nuclear fission chain reactions, the production of fission products ceased and the amount of shorter half-life radionuclides such as 89Sr, decreased by radioactive decay, resulting gradually decreasing 89Sr/90Sr ratio. The analysis of the 89Sr/90Sr ratio can reveal the main source of radiostrontium contamination.

The published data on 89Sr contamination is strongly limited, but the available results (not presented here) show a range for the 89Sr/90Sr ratio of between 10.3 and 14.9, suggesting Reactor Core origins instead of the Spent Fuel Pools.
The expression of stagnant water means that the contaminated water collected in the reactor buildings, turbine buildings and various underground tunnels and stored water in tanks at FDNPP. Leakage is present with this water, but significant amount has not been released into the environment, and many countermeasures had and have been taken to protect the environment.
Effect of the Fukushima accident

Wet and dry fallout

Max: 358

Chernobyl accident

Max: 6

Fukushima accident

Max: 470
Detailed fallout results sampled in Japan

Monthly dry and wet fallout at Fukushima, Tokyo and Akita in the same time intervals in different years
Sr-90 contamination in Japanese soil after the Fukushima accident

Activity concentration of $^{90}$Sr in Soil samples (Bq/kg) vs. Distance from the FDNPP.
Dose estimation

• Accomplishing a reliable dose estimation using the available radiostrontium data might be questionable but comparison is possible based on the UNSCEAR estimation for nuclear weapon tests and for the Chernobyl accident.

• In the case of the nuclear weapon tests, the maximum estimated average annual effective dose for the northern hemisphere was 125 µSv year⁻¹ (1963). In this case, the contribution of radiostrontium isotopes via ingestion and inhalation was about 10 µSv year⁻¹.  
  
  UNSCEAR 2000 ANNEX C

• For the Chernobyl accident, the average annual effective dose received by 84% of the population (1,573,383) of the contaminated area was below 1 mSv year⁻¹ between 1986 and 1995. The estimated contribution of radiostrontium isotopes was 5-10%, meaning a maximum of 100 µSv year⁻¹.  
  
  UNSCEAR 2000 ANNEX J

• Considering uncertainties and applying overestimation, the maximum range of the average annual effective dose originating from radiostrontium isotopes released by the Fukushima accident and excluding the restricted area is tens of µSv year⁻¹.

• In relation to the effective dose originating from natural sources in Japan (2.1 mSv year⁻¹) and the estimated effective dose from external radioactive sources dispersed via the Fukushima accident among the evacuated residents (1-6 mSv, max 19 mSv), the effective dose burden from radiostrontium isotopes is probably negligible.
Difficulties of Sr-90 Analysis in soil samples originated from Fukushima

Soil sample preparation

- DRYING
- HOMOGENIZATION
- SIEVING
- ASHING (400-600°C)

Wet chemical destruction

- OPEN
- Closed (Microwave or electric oven)

Chemical separation of Sr

- precipitation/co-precipitation,
- liquid–liquid extraction,
- ion-exchange chromatography,
- EXTRACTION CHROMATOGRAPHY

Beta particle detection

- Gas ionization detector
- Semiconductor detector
- Solid scintillation detector
- LIQUID SCINTILLATION DETECTOR

BOLD, UPPERCASE LETTER: applied procedure at NIRS
Fukushima soil samples are rich in Al$_2$O$_3$ and Fe$_2$O$_3$ comparing to samples from Chernobyl in which SiO$_2$ abundance is dominant.

Mineral composition and concentration of the soil sample can influence the Sr recovery on the extraction chromatography (Sr resin).

**Sr Recovery in the NIRS laboratory**

- Open wet chemical decomposition $\sim$70% (50-80%)
- Extraction chromatography $\sim$90% (80-99%)
- Total: $\sim$65%

Sr recovery on the Sr-resin is very good. Elevated amount of Al$_2$O$_3$ and Fe$_2$O$_3$ has no disturbing effect.

Mineral composition was measured with XRF spectrometer.
Problem with the radiocesium isotopes

Main features of radiocesium isotopes

<table>
<thead>
<tr>
<th>Radiostrontium Isotopes</th>
<th>Cs-134</th>
<th>Cs-137</th>
</tr>
</thead>
<tbody>
<tr>
<td>Parents isotopes</td>
<td>Neutron capture (Cs-133)</td>
<td>Xe-137</td>
</tr>
<tr>
<td></td>
<td>Fission product</td>
<td></td>
</tr>
<tr>
<td>Daughter isotopes</td>
<td>Ba-134</td>
<td>Ba-137</td>
</tr>
<tr>
<td>Half-life</td>
<td>2.07 y</td>
<td>30.2 y</td>
</tr>
<tr>
<td>Decay Mode</td>
<td>β</td>
<td>β</td>
</tr>
<tr>
<td>Max. Beta Energy keV</td>
<td>650</td>
<td>510</td>
</tr>
</tbody>
</table>

Gamma rays have discrete energy spectrum
Element and isotope discrimination is possible
Beta particles have continuous kinetic energy spectrum
Element and isotope discrimination is NOT possible, Highly efficient element separation is demanded

Presence of radiocesium isotopes in the measurand can interfere with radiostrontium isotopes causing false radiostrontium result
Cs separation efficiency test on Sr resin

Typical Cs-137/Sr-90 activity ratio in Fukushima soil samples is about 10,000
(Cs 137: ~200 kBq/kg, Sr-90: ~20 Bq/kg)

Considering 10% uncertainty (2 Bq/kg) originated from Cs-137

99.999 (five 9)%!

Cs separation efficiency is required

Has the Sr resin capability to fulfill this requirement?
Result of Cs separation efficiency test on Sr resin

Using standard solution (Cs and Sr) 99.999% (five 9) purification is possible.

In case of Fukushima soil sample the maximum purification efficiency is 99.99%

If the Cs-137/Sr-90 activity ratio is 10,000 Reliable Sr-90 measurement is questionable (Cs-137 and Sr-90 activity could be similar in the measurand)

Depending on the Cs-137/Sr-90 activity ratio in the soil sample Sr result measured with LSC can be affected by radiocesium isotopes contamination

Contamination check with gamma spectroscopy Detection limit should be in the range of tens of mBqS

Recovery was measured with ICP-MS
Results of Sr-90 measurement

<table>
<thead>
<tr>
<th>No.</th>
<th>Sampling point</th>
<th>Date of sampling</th>
<th>Distance From FDNPP</th>
<th>Note</th>
<th>Dose rate µSv/h</th>
<th>Cs-134 kBq/kg</th>
<th>Cs-137 kBq/kg</th>
<th>Sr-90 Bq/kg</th>
<th>Cs-137/Sr-90</th>
</tr>
</thead>
<tbody>
<tr>
<td>1.</td>
<td>Minamitsushima</td>
<td>2013.05.16</td>
<td>&lt;30km</td>
<td>grass field</td>
<td>6.5</td>
<td>77.78±0.1</td>
<td>154.3±0.14</td>
<td>8.9±0.8</td>
<td>15,000</td>
</tr>
<tr>
<td>2.</td>
<td>Chojabara</td>
<td>2013.05.16</td>
<td>&lt;3km</td>
<td>forest</td>
<td>21.9</td>
<td>86.43±0.18</td>
<td>170.2±0.26</td>
<td>20±1.3</td>
<td>8,500</td>
</tr>
<tr>
<td>3.</td>
<td>Mankai-bashi</td>
<td>2013.05.16</td>
<td>&lt;10km</td>
<td>grass field</td>
<td>0.6</td>
<td>0.301±0.004</td>
<td>0.603±0.005</td>
<td>&lt;6.8</td>
<td>100</td>
</tr>
<tr>
<td>4.</td>
<td>Murohara hashi</td>
<td>2013.05.16</td>
<td>&lt;20km</td>
<td>river bank</td>
<td>7.4</td>
<td>37.71±0.11</td>
<td>77.59±0.16</td>
<td>23.4±1.5</td>
<td>4,000</td>
</tr>
</tbody>
</table>

Open chemical decomposition
Sr resin used in the extraction chromatography
Beta particle detection; Tricarb 3100 liquid scintillation detector system

The Sr-90 measurement results using the above discussed methods are presented here. The number of samples is low for correct conclusion but significant positive correlation with radiocesium isotopes cannot be confirmed as the highest Sr-90 value is related to moderated radiocesium contamination. However, the lowest Sr-90 value is related to slightly contaminated soil sample. Significant negative correlation with the distance from the FDNPP also cannot be confirmed as it was discussed in Slide 12.
Thank you very much for your kind attention!

Please do not hesitate to contact me if you have questions: norbert@fml.nirs.go.jp