Observations of atmospheric radionuclides from the Fukushima nuclear accident in Tsukuba, Japan

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TALK OUTLINE

- Aim of the research
- Extent of the pollution by the Fukushima accident
- Observations and experimental at the MRI
- Gamma emitters in the atmosphere at the accident
- Plume transport reconstruction by the modeling
- Radio-Sr in the atmosphere at the accident
- Temporal changes in radio-Cs concentration in the atmosphere
- Long-term monitoring of radioactive fallout ($^{90}$Sr and $^{137}$Cs); comparison with the Fukushima fallout
- Summaries
POINTS TO BE CLARIFIED ACCORDING TO OBSERVATION AND MODEL

1. Point source
2. Emission inventory
3. Temporal variation

Learn quantitative relations

• Transportation and diffusion
• Wet deposition
• Dry deposition

Transportation and deposition result

Health and environmental impacts
Result of aircraft monitoring by the Ministry of Education, Culture, Sports, Science and Technology

(Total of cumulative Cs-134,137 pollution of the surface (kBq/m²) in the range that the survey completed up to fall, 2011)

November 11, 2011  Announcement
### Emission Estimate by Nuclear and Industrial Safety Agency (Oct. 20, 2011)

<table>
<thead>
<tr>
<th>Nuclide</th>
<th>Half life</th>
<th>Emission (PBq)</th>
<th>Emission (PBq)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Kr-85</td>
<td>10.72y</td>
<td></td>
<td>33</td>
</tr>
<tr>
<td>Xe-133</td>
<td>5.25d</td>
<td>11000</td>
<td>6500</td>
</tr>
<tr>
<td>Te-129m</td>
<td>33.6d</td>
<td>3.3</td>
<td>240</td>
</tr>
<tr>
<td>Te-132</td>
<td>3.26d</td>
<td>88</td>
<td>~1150</td>
</tr>
<tr>
<td>I-131</td>
<td>8.04d</td>
<td>160</td>
<td>~1760</td>
</tr>
<tr>
<td>I-133</td>
<td>20.8h</td>
<td>42</td>
<td>910</td>
</tr>
<tr>
<td>Cs-134</td>
<td>2.06y</td>
<td>18</td>
<td>~47</td>
</tr>
<tr>
<td>Cs-136</td>
<td>13.1d</td>
<td></td>
<td>36</td>
</tr>
<tr>
<td>Cs-137</td>
<td>30.0y</td>
<td>15</td>
<td>85</td>
</tr>
<tr>
<td>Sr-89</td>
<td>50.5 d</td>
<td>2</td>
<td>~115</td>
</tr>
<tr>
<td>Sr-90</td>
<td>29.12y</td>
<td>0.14</td>
<td>~10</td>
</tr>
<tr>
<td>Pu-239</td>
<td>24065y</td>
<td>0.00000032</td>
<td>0.015</td>
</tr>
<tr>
<td>Pu-240</td>
<td>6537y</td>
<td>0.00000032</td>
<td>0.013</td>
</tr>
</tbody>
</table>

**Comparison:** Emission by the Chernobyl accident (Chernobyl forum)
SAMPLING, SAMPLE PREPARATIONS AND GAMMA MEASUREMENT

HV filter sampling

Total = Wet and dry deposition sampling

Until March 2011

4 m² deposition sampler

Stocked samples

For Sr analysis

For γ-measurement

Hydraulic press machine

Rotary evaporator

For γ-measurement
### Radiochemical Separation of Radio-Sr

<table>
<thead>
<tr>
<th>Step</th>
<th>Processes</th>
</tr>
</thead>
</table>
| 1    | • Sample dissolution in nitric acid  
      • Adding stable Sr and Ca carriers |
| 2    | • Hydroxide precipitation (Bulk recovery of alkaline earths)  
      • Oxalate precipitation |
| 3    | • Fuming nitric acid precipitation  
      • Barium chromate precipitation |
| 4    | • Hydroxide precipitation (Recovery of Sr)  
      • Iron hydroxide precipitation |
| 5    | • Fixation as Sr carbonate  
      • Low BG β counting |

**Yield of Sr**: about 60~90%
GAMMA-RAY SPECTRUM OF A HV FILTER SAMPLE

A sample collected at the observation field of MRI, Tsukuba

Igarashi et al., ICAS2011
RADIOACTIVITY IN AEROSOL SAMPLES AT THE MRI IN MARCH 2011

In logarithmic scale

Date


Activity concentration (Bq/m³)

99Mo
132Te
131I
129Te
129mTe
133I
134Cs
137Cs
136Cs

ND levels

Igarashi et al., ICAS2011
TRANSPORT OF RADIOACTIVE PLUME TO TSUKUBA (MAR./15 AND MAR./20)

Non volatile I-131 [Bq/m$^3$] 2011/03/15 06JST

Cs-137 [Bq/m$^3$] 2011/03/20 12JST

Observed I-131 at the MRI: 70Bq/m$^3$

Observed Cs-137 at the MRI: 40Bq/m$^3$

Kajino et al., MSJ2011Fall
β-measurement of $^{89}\text{Sr}$ and $^{90}\text{Sr}(^{90}\text{Y})$

Radio-equilibrium with Sr-90+Y-90

- $^{89}\text{Sr} : ^{90}\text{Sr} = 2 : 0.14$
  (based on NISA estimation)
- Measured from 2011/Dec/7

Igarashi et al., 2012JpGU

Radio-eqlbrm. $^{90}\text{Sr}$ and $^{90}\text{Y}$

- Maximum β-energy
  - $^{90}\text{Sr}(28.8\text{y})$ 0.546MeV
  - $^{90}\text{Y}(2.67\text{d})$ 2.240MeV
  - $^{89}\text{Sr}(50.52\text{d})$ 1.497MeV
DETERMINATION OF $^{89}$Sr AND $^{90}$Sr
(MAR. 15, 2011 FILTER SAMPLE)

$A_{Total} = A_{Sr-90} + A_{Y-90} (1 - e^{-\lambda_{90}t}) + A_{Sr-89} e^{-\lambda_{89}t}$

NISA estimate
$^{89}$Sr : $^{90}$Sr = 2 : 0.14
March 15th meas. $^{89}$Sr : $^{90}$Sr = 10 : 0.14

Igarashi et al., 2012JpGU
Max. on March 15
1.5 ± 0.27 mBq/m³

Igarashi et al., 2012 JpGU
• $^{137}\text{Cs}/^{90}\text{Sr}$ ratio in the weapon fallout during the 1960-70
  ⇒ About 1.6 (Krey et al., 1970)
• Chernobyl accident in 1986 ⇒ 95.7 (Aoyama et al., 1991)

Igarashi et al., 2012 JpGU
TEMPORAL CHANGE IN ATMOS. RADIO-Cs CONCENTRATION AT THE MRI, TSUKUBA


Maximum concentration of 38 Bq/m³ of $^{137}$Cs during 20-21 March, 2011

In July 2012 average concentration of $^{137}$Cs was 23 μBq/m³

$^{137}$Cs concentration prior to March, 2011 was ~ 1 μBq/m³
DECREASE IN ATMOSPHERIC RADIO-CS CONCENTRATIONS


- $y = 0.268 \times e^{-1.40x}$, $R = 0.267$
- $y = 0.478 \times e^{-1.79x}$, $R = 0.343$

**Atoms. Activity Conc. (mBq/m³)**
- $1^{34}$Cs
- $1^{37}$Cs

**T$_{1/2}$**
- $1^{37}$Cs: 0.495 year
- $1^{34}$Cs: 0.387 year
LONG-TERM DEPOSITION RECORD AT THE MRI, TSUKUBA WITH THE FUKUSHIMA FALLOUT

Analysis of the total deposition sample collected monthly

Radioactivity deposition (mBq/m²/month)

Year


Koenji, Tokyo

Tsukuba

Chernobyl Accident

Fukushima Accident

Fukushima accident atmospheric fallout

Nuclear tests by former USSR, USA, etc.

Nuclear tests by China

Igarashi et al., 2012JpGU
# Atmospheric Deposition After the Fukushima Accident at the MRI, Tsukuba

**Unit:** Bq/m² /month

<table>
<thead>
<tr>
<th>Month</th>
<th>$^{90}$Sr</th>
<th>Errors</th>
<th>$^{137}$Cs</th>
<th>Errors</th>
<th>Ratio</th>
</tr>
</thead>
<tbody>
<tr>
<td>Mar/2011</td>
<td>4.36 ±0.09</td>
<td>2.308E+04</td>
<td>±924</td>
<td></td>
<td>5292</td>
</tr>
<tr>
<td>Apr/2011</td>
<td>4.00 ±0.07</td>
<td>1.776E+03</td>
<td>±1.3</td>
<td></td>
<td>444</td>
</tr>
<tr>
<td>May/2011</td>
<td>0.33 ±0.03</td>
<td>330 ±0.3</td>
<td>989</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Jun/2011</td>
<td>0.13 ±0.02</td>
<td>104 ±0.1</td>
<td>804</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Jul/2011</td>
<td>0.05 ±0.01</td>
<td>82.0 ±0.1</td>
<td>1808</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Aug/2011</td>
<td>0.07 ±0.01</td>
<td>31.9 ±0.1</td>
<td>435</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Sep/2011</td>
<td></td>
<td>45.9 ±0.1</td>
<td>1</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Oct/2011</td>
<td></td>
<td>25.8 ±0.1</td>
<td>0</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Nov/2011</td>
<td></td>
<td>5.9 ±0.0</td>
<td>0</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Dec/2011</td>
<td></td>
<td>20.3 ±0.1</td>
<td>0</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Jan/2012</td>
<td></td>
<td>32.6 ±0.1</td>
<td>0</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

Igarashi et al., 2012 JpGU
We have continued observations of atmospheric concentrations of radioactive Sr and Cs, etc. and their depositions at the MRI, Tsukuba before and after the Fukushima nuclear accident.

The plume transport to the MRI from the Fukushima accident was captured and reconstructed by the transport modeling.

By tracking the temporal change of total $\beta$-activity of Sr, which was resolved into $^{89}$Sr and $^{90}$Sr.

The relative ratio of 70 is estimated for $^{89}$Sr and $^{90}$Sr in March 2011, which is about 5 times larger than the NISA estimate.

The $^{137}$Cs/$^{90}$Sr ratio was in the range of 400 ~ 20000 in filter samples, suggesting the Cs enrichment during the transport.

The $^{137}$Cs monthly deposition at the MRI was $(23\pm0.9) \times 10^3$ Bq/m$^2$ in March 2011, which is 6 to 7 orders of magnitude larger than pre-accident level.

Equal amounts of $^{134}$Cs and $^{137}$Cs deposited, giving rise to the surface pollution of approximately 50 kBq/m$^2$ in Tsukuba, matched nearly with that by the MEXT mapping.
90Sr depositions of 4.36±0.09 Bq/m^2 in March 2011, which is less than 0.02% of the total 137Cs fallout in the month.

Level of 90Sr deposition was 3-4 orders greater than pre-accident level, which did not reach one by nuclear tests during the 1960s; impact by 90Sr will not be so large as radioactive Cs.

At the end of 2011, the radioactive fallout lowered 3-4 orders from that on the accident, yet the several-Bq/m^2-deposition is continuing. This corresponds to the level in the early 1980s when China carried out the last atmospheric nuclear test.

At the end of 2011, 137Cs concentration was at the level of tens μBq/m^3. Since the re-suspension continues over a long period of time, it is necessary to watch its future trends.

Apparent decrease in the atmos. Cs concentration occurs with a half-life of ca. 6 months since Sep. 2011, suggesting that the removal occurs relatively quickly at the 2ndary emission sources.

Identification of the major source of 2ndary emission to the atmosphere (re-suspension) is required.
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