Rapid isolation method for radioactive strontium using Empore™ Strontium Rad Disk

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1. Introduction

A serious accident occurred at the Fukushima Daiichi nuclear power plants (FDNPP) in March 2011, resulting in the discharge of large amounts of radionuclides into the environment. Of these nuclides, artificial 90Sr has a particularly long half-life (T1/2 = 28.6 years) and high radiotoxicity. Therefore, its measurement is a priority in environmental assessment and response. As a fission product from nuclear weapons testing, 90Sr had already been widely distributed over the Northern hemisphere before the FDNPP accident. It has been measured in atmospheric fallouts worldwide [1-4], and it was also detected in milk following the Chernobyl accident [5]. Several studies have indicated that the mobility of 90Sr in the environment is higher than that of 137Cs [6-7]. Thus, 90Sr is one of the most important radionuclides in nuclear plant accidents in terms of impact on human health. Following the FDNPP accident, many environmental samples from eastern Japan have been examined to investigate the circumstances of the accident and to assess the resultant contamination distribution. In a previous study, we used gamma-ray spectroscopy to determine the radioactivities of 131I, 134Cs, and 137Cs in such samples. Aerosol samples were collected by high-volume air samplers and the time variation of radiocesium activity concentrations was observed [8]. In contrast, only a few studies have investigated 90Sr in soils and seawater [9-10]. Consequently, data on the 90Sr activity concentration in the atmosphere and its time variation are needed to fully evaluate the internal radiation exposure and evaluate the amount of radionuclides discharged during the accident. Because 90Sr and its daughter nuclide 90Y are pure β emitters, the activity determination of 90Sr in environmental samples requires chemical separation. Classical separation methods including multi-stage operations are time consuming and produce large amounts of chemical waste[11]. In recent years, new chemical separation techniques for strontium using solid-phase extraction have been developed [12-13]. However, these new methods have only been applied to soil, milk, and surface and ground water samples. A simple and rapid solid-phase extraction method for 90Sr in aerosol samples would offer the advantage of exchange capacity, as no matrix interference (e.g., from calcium or strontium) would be present. However, such an approach has not been reported to date. In this study, we developed a new chemical separation strategy for measuring 90Sr in aerosol samples based on solid-phase extraction. The method consists of three steps: a selective Sr extraction using a Strontium Rad Disk™ (3M Inc.), an ion-exchange separation to eliminate naturally occurring 210Pb, and finally Cherenkov light measurements for the determination of 90Sr by detecting 90Y growth. The Strontium Rad Disk™ can extract Sr selectively but it also extracts Ba, Ra, and Pb simultaneously [14]. We used cation-exchange chromatography with ethylenediaminetetraacetic acid (EDTA) elution after the extraction to eliminate the naturally occurring radionuclide 210Pb, which is found in aerosol samples. However, Ba and Ra could not be eliminated. For low-background measurements, 90Sr radioactivity was determined by counting the Cherenkov light emitted from 90Y with a liquid scintillation counter [15]. Performance indicators of this technique, including Sr yields, Pb elimination rates, total operation time, and detection efficiency of Cherenkov light measurement, were determined using standard radioactive and/or stable isotope samples. Finally, the developed method was applied to aerosol samples collected in Mito City, Japan in April 2011. Thereby, we determined the 90Sr activity concentration and 90Sr/137Cs activity ratio in the atmosphere following the FDNPP accident.

2. Materials and Methods

2.1. Reagents. Empore™ Strontium Rad Disks (47 mm in diameter) were employed to extract Sr from aerosol samples. These disks contain a proprietary multi-cyclic macromolecule covalently bound to a solid silica support embedded in a stable, inert matrix of polytetrafluoroethylene fibrils. When 2-4 M HNO3 or HCl solution is passed through the disk, up to 3 mg of Sr in the solution could be selectively extracted [14]. Dowex 50W-X8 cation-exchange resin (100-200 mesh, Wako...
were collected on Pall Tissuquartz™ filters, 2500 QAT-UP collected in April 2011 at Mito City (N36.40, E140.44) in measurement of 90Y, we used a 1220 QUANTULUS™ Ultra Low efficiency for 90Sr counting was determined by measuring of the Research Center for Nuclear Physics at Osaka University. Strontium-85 was generated by proton irradiation of a Rb target using the Azimuthally Varying Field (AVF) cyclotron of at GEM40, ORTEC). The detector was connected to a multi-channel analysis system (MCA7600, Seiko EG&G). Concentrated solutions of ultrapure analytical grade HNO₃ and HCl were purchased from Kishida Inc. Diluted acid solutions were obtained by mixing the concentrated regents with Milli-Q water. All water used in this study was obtained from a Milli-Q™ water purification system. All other chemicals were of Japanese Industrial Standards special grade.

2.2. Instruments. All gamma-ray measurement was performed with a coaxial-type high-purity germanium detector (GEM40, ORTEC). The detector was connected to a multi-channel analysis system (MCA7600, Seiko EG&G).

In the experiments to develop new chemical separation methods, an LS 6500 liquid scintillation counter (Beckman Inc.) was used to measure beta-rays. For the determination of 90Sr activities in aerosol samples by the Cherenkov lights measurement of 90Y, we used a 1220 QUANTULUS™ Ultra Low Level Liquid Scintillation Spectrometer (PerkinElmer Inc.) with 20 mL Teflon™ vials (PerkinElmer Inc.) The detection efficiency for 90Sr counting was determined by measuring of the 90Sr standard solution.

An Agilent/HP 4500 ICP-MS (Agilent Technologies Inc.) was used for determining chemical yields of Sr, after the chemical separation and sequential Cherenkov light measurement.

2.3. Aerosol samples. A series of aerosol samples were collected in April 2011 at Mito City (N36.40, E140.44) in Ibaraki Prefecture of eastern Japan. In each sample, aerosols were collected on Pall Tissuquartz™ filters, 2500 QAT-UP (Sigma-Aldrich Inc.), with a Kimoto-121FT high-volume air sampler (Kimoto Texh Inc.). The size of the filter was 10 × 8 cm². The air-flow of the sampler was approximately 700 L min⁻¹. Cesium-137 activity was directly determined for each sample with the germanium semiconductor detector. A standard filter was used to determine the efficiency of the germanium semiconductor detectors. The standard filter spotted radioactive standard activity uniformly was folded into the same shape of the measurement samples [8].

We chose four filter samples from the samples collected in April for determining the 90Sr activity concentrations in the aerosols using our new method. The details of the samples are shown in Table 1. These four samples had relatively high 137Cs activities, and thus we expected that these samples would have 90Sr high activities above that of the liquid scintillation counter detection limit. We analyzed a half or quarter portion of each aerosol filter sample.

2.4. Strontium separation procedure. 2.4.1. Sample pretreatment. First, each separated filter sample was cut into pieces (approximately 5 × 2 cm²), which were then put into a 500-mL Teflon beaker. Next, 100 mL of 12 M HCl, 30 mL of 13 M HNO₃, and 1.0 mg of stable Sr carrier were added to the beaker. The acid solution was then heated on a hot plate to 150 °C for 3 h to extract Sr into the solution. After cooling to room temperature for 1 h, the solution was filtered with a 0.45-μm membrane cellulose filter (φ47, Advantec no.7) after decantation to roughly separate the solution and residues. The solution was then placed into a 300 mL-polyethylene container. The residues on the filters were washed with 30 mL of 3.25 M HCl twice followed by 30 mL Milli-Q water once. The washing solutions were also added to the polyethylene container. The mixed sample solution was weighed, and then 3 mL of the solution was removed to obtain the initial Sr concentration by ICP-MS for the chemical yield determination. The remaining solution was transferred to a 500-mL measuring flask. Milli-Q water was added into the acid to adjust the concentration to 3.25 M HCl, which is suitable for Sr extraction by the disk [14].

In this study, the extraction yield of 90Sr from each aerosol filter was obtained by comparing its activity with those of 137Cs from the same sample before and after chemical treatment. We presumed that the Sr and Cs in the aerosol sample had similar chemical extractability into the acid solution. The filter residues after extraction were dried in a desiccator at room temperature, and then formed into approximately the same geometric shape as the intact filter for gamma-ray measurement.

2.4.2 Solid-phase extraction. The complete separation procedure, including a solid-phase separation and a cation

| Table 1: Details of the four FDNPP aerosol samples and chemical analysis results |
|--------------------------------|--------|--------|--------|--------|
| Aerosol sample name | MIT4 | MIT9 | MIT12 | MIT13 |
| Air volume (m³) | 2070 | 1572 | 3085 | 2139 |
| Measurement date | 2015/7/17 | 2015/8/1 | 2015/7/18 | 2015/7/13 |
| Effective air-volume (m³) | 497 | 770 | 771 | 1069 |
| 137Cs yield | 99±2% | 99±5% | 98±3% | 98±4% |
| Stable Sr yield | 83±6% | 87±6% | 86±6% | 85±6% |
| 90Sr concentration (mBq/m³) | 1.5±0.1 | 0.79±0.6 | 0.50±0.04 | 0.055±0.005 |
| 137Cs concentration (Bq/m³) | 0.82±0.08 | 0.057±0.006 | 0.11±0.01 | 0.041±0.004 |
| 90Sr/137Cs activity ratio (×10⁷) | 1.9±0.2 | 14±1 | 4.6±0.6 | 1.3±0.2 |

Notes: 85Sr and 137Cs activity concentrations were calculated from the equation provided in the “Measurement instruments” subsection. Effective air-volume was calculated from the mass of aerosol sample analyzed. All activities were decay collected to 11 March, 2011. The confidence interval was 95%.
exchange chromatography, is illustrated in Figure 1. For solid-phase extraction, an Empore™ Strontium Rad Disk was set in a 47-mm diameter funnel and preconditioned by the addition of 10 mL of methanol, waiting 20 min, and then rinsing with 30 mL of 3.25 M HCl by vacuum filtration with a flow rate of approximately 50 mL min⁻¹. The sample acid solution in the 500-mL flask was then passed through the disk to extract Sr ions into the solid-phase. The disk was then washed with 15 mL of Milli-Q water to remove residual acids. To elute Sr ion from the disk, 10 mL of 0.02 M EDTA solution was passed through at a flow rate of 2 mL min⁻¹ [16]. Another 10 mL of this EDTA solution was passed through the disk at 10 mL min⁻¹ as the final rinse. These two solutions were combined and used for cation-exchange separation.

2.4.3 Cation-exchange separation. The cation-exchange process is illustrated in the lower-right portion of Figure 1. First, 0.25 g of Dowex 50W-X8 (100-200 mesh) ion-exchange resin was placed into an S-size Muromac™ mini-column (Φ8 × 50 mm). Then, 10 mL Milli-Q water and 10 mL of 0.01 M HCl were passed through the column consecutively to condition the ion-exchange resin. To the 20 mL EDTA sample solution, 0.75 mL of 13 M HNO₃ was added to adjust its pH, and the solution was passed through the preconditioned column. Under these conditions, both Sr and Pb ions were adsorbed onto the cation-exchange resin. Then, 10 mL of 10⁻⁴ M HCl solution was passed through the column to wash out the residual EDTA, followed by 15 mL of 2% (w/v) EDTA solution to remove Pb ions from the resin. Finally, 15 mL of 3.25 M HCl was passed through the column for Sr ion elution. The final solution was placed into a 20 mL Teflon vial to be measured by a low-background liquid scintillation counter.

2.5. Radioactivity calculation. Beta-rays from ⁹⁰Y, a daughter nuclide of ⁹⁰Sr, were measured to determine the amount of ⁹⁰Sr in the aerosol samples. The beta rays emitted from ⁹⁰Y have an endpoint energy of 2.2 MeV and can effectively emit Cherenkov light in water. The Cherenkov light counting efficiency for ⁹⁰Sr is much lower than that for ⁹⁰Y, therefore we can measure ⁹⁰Sr activity selectively. We can identify the activity of ⁹⁰Sr from the growth of ⁹⁰Y over time. Owing to the large difference between the half-lives of ⁹⁰Sr and ⁹⁰Y, radioactive equilibrium was reached within 2 weeks. The ⁹⁰Sr activity approximately coincides with the ⁹⁰Y activity after 2 weeks from ⁹⁰Sr isolation.

The ⁹⁰Sr activity concentration (Bq m⁻³) in air, A, was determined from the ⁹⁰Y activity as

\[
A = \frac{C - C_b}{\varepsilon RV}
\]

where C is the count rate after ⁹⁰Sr-⁹⁰Y radioactive equilibration, C_b is the background count rate, and \(\varepsilon\) is the counting efficiency of Cherenkov light for ⁹⁰Y. In our measurement system, the background counting rate was approximately 0.028 count per second (CPS), and the detection efficiency of Cherenkov light from ⁹⁰Y was 68.7 ± 0.1 %. \(R\) is the yield of Sr chemical separation determined by ICP-MS, and \(V\) is the collected air volume (m³) of the aerosol samples. In the ICP-MS measurement, the chemical yield was determined by measuring the amount of stable Sr carrier ions in the measurement sample after a sequential Cherenkov light measurement. A 1 mL aliquot of each measurement sample was diluted 1000 times with water and HNO₃, and adjusted to a final concentration of 5% w/v HNO₃. Four external calibration solutions were prepared with the 5% w/v HNO₃ to 0, 10, 20, 50, and 100 ng mL⁻¹. The calibration curve for the Sr was found to be linear \((R^2 ≥ 0.9999)\). In each measurement, we performed at least six analytical runs and used the averaged value. The chemical yields were determined from the ratio of the Sr mass in the measurement sample to that of the Sr added initially.

The uncertainties of the ¹³⁷Cs radioactivity concentrations were the combined uncertainties of the efficiency and counting statistics in the gamma-ray measurement. The uncertainties of the stable Sr yield resulted from errors in the ICP-MS measurement. The total uncertainties in the ⁹⁰Sr radioactivities were the combined uncertainties of ¹³⁷Cs yields, stable Sr yield, counting statistics in the ⁹⁰Sr measurement, and uncertainty in the efficiency of the Cherenkov light measurement. When reporting the results, 95% confidence intervals were used in this study.

3. Results and Discussion

3.1. Optimization of ²¹⁰Pb cation-exchange separation conditions. Cation exchange was applied after extraction by the Rad Disk method to eliminate ²¹⁰Pb, because contamination by naturally occurring ²¹⁰Pb causes considerable background signals during ⁹⁰Sr detection. Pb ions in solution are usually separated by precipitation in a chromate form at pH
investigated by measuring the gamma-rays from $^{85}$Sr (0.514 MeV) using a germanium semiconductor detector, and/or the amount of stable Sr carrier in the sample solution by ICP-MS. The Sr yield in each step of the optimized method was determined to be more than 95%, which is consistent with the disk extraction performance (Waste A) [14]. Cations other than Sr do not cause serious interference in this step. In our method, to remove any residual acid from the disk that may disrupt EDTA complex formation, the disk was washed with 10 mL water before Sr ion elution. With this washing step, the loss of Sr ions in Waste B (see Figure 1) is suppressed to within 2–3%. In the cation-exchange operation, Sr-containing EDTA eluate from the disk was mixed with 0.75 mL of 13 M HNO$_3$, and then passed through the cation-exchange resin. The Sr ion losses during passage through the column, pH conditioning, and elution of $^{210}$Pb from the column are contained in Wastes C, D, and E, respectively. We measured the activity of $^{85}$Sr in these wastes by gamma-ray counting and the amount of stable Sr in these wastes by ICP-MS. The Sr losses in these wastes are less than 1%. Hence, we conclude that no significant loss of Sr occurs in these steps.

3.3. Extraction yields of $^{89}$Sr from aerosol samples. Cesium-137 yields in acid pretreatment step were measured for alternative evaluating $^{89}$Sr leaching yields from aerosol samples. Stable Sr yields of the chemical separation were measured by ICP-MS. From the Cherenkov light counting result, the $^{137}$Cs yields and the stable Sr yields, the activities of $^{89}$Sr in the aerosol samples were determined and are summarized in Table 2.

The activities of $^{89}$Sr in the atmosphere are in the range of 0.055–1.5 mBq m$^{-3}$, which is consistent with previous measurement results [2].

For the four analyzed aerosol samples, the $^{137}$Cs is perfectly leached during acid pretreatment. Cesium-134 and -137 are transported through the atmosphere in sulfate aerosol form [18], and sulfate aerosols dissolve in HCl solution easily. Strontium is also transported through the atmosphere in sulfate aerosol form. Therefore, we conclude that the $^{89}$Sr in the filter is completely removed by the acid pretreatment, five waste solutions (A-E) were collected during chemical separation, and the chemical separation yield and Sr loss were estimated by stable Sr ion measurement using ICP-MS. The Sr yields and losses in the waste solutions are summarized in Table 2. The Sr chemical yields in all four samples are more than 80%. The sum of the Sr yields and losses in each waste are less than 1%.

### Table 2: Yields of stable Sr after chemical separation and loss of Sr in each separation step

<table>
<thead>
<tr>
<th>Sample name</th>
<th>MIT4</th>
<th>MIT9</th>
<th>MIT12</th>
<th>MIT13</th>
</tr>
</thead>
<tbody>
<tr>
<td>Waste A</td>
<td>1%</td>
<td>3%</td>
<td>1%</td>
<td>2%</td>
</tr>
<tr>
<td>Waste B</td>
<td>2%</td>
<td>3%</td>
<td>1%</td>
<td>5%</td>
</tr>
<tr>
<td>Waste C</td>
<td>1%</td>
<td>1%</td>
<td>2%</td>
<td>1%</td>
</tr>
<tr>
<td>Waste D</td>
<td>&lt;1%</td>
<td>&lt;1%</td>
<td>&lt;1%</td>
<td>&lt;1%</td>
</tr>
<tr>
<td>Waste E</td>
<td>&lt;1%</td>
<td>&lt;1%</td>
<td>&lt;1%</td>
<td>&lt;1%</td>
</tr>
</tbody>
</table>

Stable Sr yield 83±6% 87±6% 86±6% 85±6%

Notes: See Figure 1 for detailed separation procedure. The confidence interval was 95%.

Figure 2. Elution curve of $^{210}$Pb from the cation-exchange column by 2% EDTA solution at pH = 5. The elution profile was determined using 1 mL aliquots. The y-axis is the activity ratio of eluted $^{210}$Pb to initially added $^{210}$Pb.

Figure 3. Elution curve of $^{89}$Sr from the cation-exchange column by 3.25 M HCl. The elution profile was determined using 2 mL aliquots. The y-axis is the activity ratio of eluted $^{89}$Sr to initially added $^{89}$Sr.

Zhang
100% because of the margin of error in the ICP-MS measurement, and the retention of Sr ions on the disk and/or cation-exchange resin.

3.4. Backgrounds in measurement of $^{90}$Sr for aerosol samples. There are three major background interferences for $^{90}$Sr ($^{90}$Y) in the four aerosol samples in this study, i.e., $^{90}$Sr from the FDNPP accident, $^{90}$Sr from global fallout, $^{210}$Pb, and $^{226}$Ra and its daughter nuclides. Each of their effects on the $^{90}$Sr determination are discussed in details as follows.

Strontium-89 with a half-life of 50.5 days is one of the most important radionuclides from the FDNPP nuclear accident. The activity of $^{89}$Sr can be detected by Cherenkov light counting, owing to its high-energy beta-rays. However, the Day 0 counting rate in Figure 4 is approximately 0.02–0.03 CPS, which agrees well with that of a blank sample (0.028 CPS). Thus, we conclude that $^{89}$Sr activity is not detected in our measurements. The $^{89}$Sr/$^{90}$Sr activity ratio of the FDNPP fuel was calculated as 0.43 by ORIGEN 2 code [19]. We began our measurement in 2015 (approximately 1500 days after the accident), and the $^{89}$Sr/$^{90}$Sr activity ratio was estimated as $1.6 \times 10^{-10}$.

The global background of $^{89}$Sr in the atmosphere is negligible in our study. The background $^{90}$Sr atmospheric concentration was $1 \times 10^{-6}$ Bq m$^{-3}$ in the Northern hemisphere in 1983 [20]. A sharp increase in the atmospheric concentration of $^{89}$Sr was observed in Europe after the Chernobyl nuclear accident. However, the $^{89}$Sr activity concentration of the global fallout returned to its pre-accident level [2] and has continued to decrease to date. The atmospheric concentration of $^{90}$Sr follows the fallout behavior and its level should be lower than $1 \times 10^{-6}$ Bq m$^{-3}$. However, in our data, the lowest $^{90}$Sr atmospheric concentration is $55 \times 10^{-6}$ Bq m$^{-3}$. Thus, we can conclude that the $^{90}$Sr activity detected in our study originates from the FDNPP accident.

Naturally occurring $^{210}$Pb provide severe background signals for $^{90}$Sr counting by Cherenkov lights measurements. The $^{90}$Sr activity concentrations obtained in the present study are $0.055–1.5$ mBq m$^{-3}$. Conversely, Tanahara et al. reported that the $^{210}$Pb activity concentrations in Okinawa Islands, Japan, were 0.06 to 1.98 mBq m$^{-3}$ from 2004 to 2011 [21]. Lead-210 only emits low-energy end-point beta-rays (0.06 MeV), but its daughter nuclide $^{210}$Bi emits relatively high-energy beta-rays (1.2 MeV). The beta-rays of $^{210}$Bi emit Cherenkov light in water, and its efficiency is approximately 30% that of $^{90}$Y. As we discussed above, our method eliminates the $^{210}$Pb contribution completely. In addition, we also confirm the possibility of $^{210}$Pb existence owing to radioactive equilibrium. If $^{210}$Pb contamination was present, more time would be required to achieve radioactive equilibrium relative to that required for the pure $^{90}$Sr-$^{90}$Y equilibrium, because $^{210}$Bi has a longer half-life than $^{90}$Y. However, all of our sequential measurement results show good agreement with the calculated $^{90}$Sr-$^{90}$Y radioactive equilibrium. Thus, we conclude that complete $^{210}$Pb elimination is achieved and thus does affect our determination of $^{90}$Sr in the aerosol samples.

Radium-226 and its daughter nuclides of $^{222}$Rn, $^{214}$Bi and $^{214}$Pb may cause background signals in the beta-ray measurements [22], because $^{226}$Ra is not eliminated by our method. However, the atmospheric concentration of $^{226}$Ra is approximately 1.5 μBq m$^{-1}$ [23], and the low-activity $^{226}$Ra and its daughters compared with that of $^{90}$Sr allow it to be ignored in the Cherenkov light measurements.

3.5. Activities of $^{90}$Sr in four aerosol samples collected after the FDNPP accident. The activities of $^{90}$Sr in the aerosol samples collected after the FDNPP accident were determined using the optimized separation method described above. The chemical separation method with solid-phase extraction was performed on each aerosol sample, and 15-mL 3.25 M HCl measurement samples were obtained. Cherenkov counting was performed over more than 2 weeks for each measurement sample. The $^{90}$Y growth curves for the aerosol samples are shown in Figure 4.

As shown in Table 2, the $^{90}$Sr activity concentrations are approximately 1/1000 those of $^{137}$Cs. Comparing the $^{90}$Sr/$^{137}$Cs activity concentrations of $^{90}$Sr in the aerosol samples collected after the FDNPP accident with those in the aerosol samples collected after the Chernobyl nuclear accident show that the activity of $^{90}$Sr in the aerosol samples collected after the FDNPP accident is lower than that in the aerosol samples collected after the Chernobyl nuclear accident. The $^{90}$Sr activity concentrations in the aerosol samples collected after the FDNPP accident are lower than those in the aerosol samples collected after the Chernobyl nuclear accident because the FDNPP accident released a smaller amount of $^{90}$Sr than the Chernobyl nuclear accident.
activity ratio in aerosols with those from soil and seawater samples provides us with a better understanding of the radioactivity discharge conditions in the FDNPP event. Soil samples collected in eastern Japan had the $^{90}\text{Sr}/^{137}\text{Cs}$ ratio of 1/10-1/1000 [10]. The Ministry of Education, Culture, Sports, Science & Technology in Japan (MEXT) reported that $^{90}\text{Sr}/^{137}\text{Cs}$ ratios in soils were approximately 1/1000 at many survey points in Fukushima Prefecture, including some points with ratios as high as 1/10-1/100 [24]. The soil $^{90}\text{Sr}/^{137}\text{Cs}$ ratios are comparable to our aerosol values; however, our data exhibit large variation. The $^{90}\text{Sr}/^{137}\text{Cs}$ ratio for seawater samples collected at 30-600 km offshore in June 2011 was reported to be 0.026±0.006 [25]. Povinec et al. reported that the $^{90}\text{Sr}/^{137}\text{Cs}$ ratio at the coast near the FDNPP varied between 0.09 and 64.5 from April 2011 to February 2012 [26]. The ratio is higher than that from our aerosol samples, probably because of the direct discharge of $^{90}\text{Sr}$ from the FDNPP site to the sea [27]. Because $^{90}\text{Sr}$ is less volatile than $^{137}\text{Cs}$, the $^{90}\text{Sr}/^{137}\text{Cs}$ ratio in aerosols would be lower. Water can contain additional $^{90}\text{Sr}$ in soluble chemical forms, which may also contribute to a higher $^{90}\text{Sr}/^{137}\text{Cs}$ ratio in seawater than in aerosols.

4. Conclusions

A rapid isolation method for $^{90}\text{Sr}$ in aerosol samples has been developed using Empore™ Sr Rad Disk. The problem of contamination by $^{210}\text{Pb}$ during solid extraction was resolved by cation exchange. The yield of Sr in this scheme was $>80\%$ and $^{210}\text{Pb}$ was completely eliminated. Because the separation method consists of only two steps, the $^{90}\text{Sr}$ chemical separation from the pretreated sample solution is achieved much faster (approximately 2.5 h) than in conventional methods. Aerosol samples collected in Mito City after the FDNPP accident were analyzed by the developed method, and the activities of $^{90}\text{Sr}$ were determined for the first time. The $^{90}\text{Sr}/^{137}\text{Cs}$ activity ratio in aerosol samples collected at Mito about 130 km south from the FDNPP site following the accident were found experimentally to be in the order of 1/1000.

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