Research Square

Preprints are preliminary reports that have not undergone peer review.
They should not be considered conclusive, used to inform clinical practice,
or referenced by the media as validated information.

Development of a Rapid and Safe Analytical Method for Radiostrontium using a Barium Silicate-based Sr Sorbent

Haruka Minowa

minowa@jikei.ac.jp

The Jikei University School of Medicine

Yoshimune Ogata

Aichi Medical University

Sadao Kojima

Aichi Medical University

Yuka Kato

ALOKA Co., Ltd

Keisuke Sueki

Tokyo Metropolitan University

Shinji Sugihara

Oita University

Tetsuya Arinobu

Aichi Medical University

Research Article

Keywords: radiostrontium, environmental water, barium silicate, strontium sorbent, analytical method development, liquid scintillation counting

Posted Date: August 7th, 2026

DOI: https://doi.org/10.21203/rs.3.rs-10544475/v1

License: This work is licensed under a Creative Commons Attribution 4.0 International License.
Read Full License

Additional Declarations: No competing interests reported.

Title page

Names of the authors: Haruka Minowa[1], Yoshimune Ogata[2], Sadao Kojima[2], Yuka Kato[3], Keisuke Sueki[4], Shinji Sugihara[5], Tetsuya Arinobu[2]

Title: Development of a Rapid and Safe Analytical Method for Radiostrontium using a Barium Silicate-based Sr Sorbent

Affiliation(s) and address(es) of the author(s):

[1]The Jikei University School of Medicine, 3-25-8, Nishi-sinbashi, Minato-ku, Tokyo, 105-8461, Japan

[2]Aichi Medical University, 1-1, Yazakokarimata, Nagakute, Aichi, 480-1195, Japan

[3]ALOKA Co., Ltd., 1-20-8, Nakacho, Musashino-shi, Tokyo, 180-0006, Japan

[4]Tokyo Metropolitan University, 1-1, Minami-osawa, Hachioji, Tokyo, 192-0364, Japan

[5]Oita University, 1-1, Idaigaoka, Hasama-machi, Yufu, Oita, 879-5593, Japan

E-mail address of the corresponding author: minowa@jikei.ac.jp

Development of a Rapid and Safe Analytical Method for Radiostrontium using a Barium Silicate-based Sr Sorbent

Yoshimune Ogata[1], Haruka Minowa[2], Sadao Kojima[1], Yuka Kato[3], Keisuke Sueki[4], Shinji Sugihara[5], Tetsuya Arinobu[1]

1Aichi Medical University, 1-1, Yazakokarimata, Nagakute, Aichi, 480-1195, Japan
2The Jikei University School of Medicine, 3-25-8, Nishisinbashi, Minato, Tokyo, 105-8461, Japan
3ALOKA Co. Ltd., 1-20-8, Nakacho, Musashino-shi, Tokyo, 180-0006, Japan
4Tokyo Metropol-itan Uni-versity, 1-1, Minamiosawa, Hachioji, Tokyo, 192-0364, Japan
5Oita University, 1-1, Idaigaoka, Hasama-machi, Yufu, Oita, 879-5593, Japan

Abstract (50 to 100 words.)

A new analytical method for radiostrontium in environmental water using a Sr sorbent was developed. In this method, the sorbent is added to sample solution, stirred for four hours, separated, and measured for 60 minutes. The minimum detectable concentration (MDC) was lower than 0.4 Bq L^{-1}, corresponding to 1／75 of the permissible effluent standard of ^{90}Sr for wastewater in Japan (30 Bq L^{-1}). The procedure provides within six hours and does not require any hazardous reagents. This method is applicable to the rapid and safe analysis of radiostrontium in environmental water.

Keywords

radiostrontium, environmental water, barium silicate, strontium sorbent, analytical method development, liquid scintillation counting

1. Introduction

Radiostrontium is known to accumulate in bone tissue, thereby increasing the risk of solid cancers and leukaemia. There is substantial societal and scientific interest in its accurate and timely measurement. However, ^{89}Sr and ^{90}Sr, are pure beta emitters, requiring chemical separation from other radionuclides prior to radioactivity determination. Conventional analytical procedures typically involve large quantities of hazardous reagents, demand skilled techniques, and require two to three weeks to obtain results [1,2]. Thus, a rapid, simple, and safe analytical method is strongly desired.

In recent years, several approaches employing crown ether-based materials, such as the Sr RadDisk (CDS Empore 3290 Strontium RAD DISK, CDS Analytical LLC) [3,4] and Sr-selective resin, have been reported [5]. Although these methods offer improved simplicity and speed, the reagents are relatively expensive for routine monitoring applications. Rapid analytical techniques using ICP-MS have also been proposed [6], ICP-MS offers high sensitivity, but its costly instrumentation, maintenance requirements, and operational complexity make it impractical for monitoring or screening applications. Another method based on measuring ^{90}Y, the progeny of ^{90}Sr, using solid-phase extraction has been described [7], however, this approach requires several weeks for ^{90}Y ingrowth and cannot be applied to ^{89}Sr.

To address these limitations, we have been investigating rapid, safe, and cost-effective radiostrontium measurement techniques using commonly available detectors. In this study, we propose a new analytical method employing a Sr-selective sorbent. The characteristics of the sorbent are evaluated, and recommended analytical procedures are demonstrated.

2. Materials and methods

2.1 Strontium sorbent

The Sr sorbent [8,9] used in this study (PURECERAM MAq®, hereafter referred to as P-MAq) was supplied by Nippon Chemical Industrial Co., Ltd. P-MAq is a white powder whose primary component is barium silicate. The particle size ranges from 5 to 125 μm, with an average diameter of 24.8 μm. This material is insoluble in water and selectively sorbs strontium. Figure 1 presents photographs of the sorbent. As shown in the SEM image

(Fig. 1c), P-MAq exhibits a complex microstructure composed of both crystalline and amorphous phases.



(a) (b) (c)

Fig. 1 PURECERAM MAq; (a) product packaging, (b) photograph, and (c) SEM image.

2.2 Summary of previous experiments

Previous experiments and their results have been reported in our earlier publications [10-14]. A summary is provided here for clarity. When 150 mg of P-MAq was added to 100 mL of simulated seawater and stirred for four hours, the average sorption ratio of Sr reached approximately 90%. The four major seawater constituents - Na, K, Mg, and Ca- were hardly sorbed to the sorbent. Cesium, a nuclide of concern in nuclear accidents, also showed no sorption. The sorption behavior of Ra, Pb, and Ba was additionally examined. Their sorption ratio exceeded 98% after four hours of stirring. These results demonstrate that P-MAq selectively sorbs Sr, Ra, Pb, and Ba, while exhibiting negligible sorption of Na, K, Mg, Ca, and Cs.

2.3 Required amount of Sr sorbent

To determine the amount of Sr sorbent required for effective strontium sorption, 25, 50, 100, 120, 130, 140 and 150 mg of P-MAq were added to 100 mL of simulated seawater spiked with several kBq of ^{85}Sr. The solutions were stirred for 240 min. During stirring, 2-mL aliquots were collected at intervals between 15 and 240 min and filtered using a syringe filter (0.45 μm pore size; DISMIC-13HP, Advantech Co., Ltd.). The radioactivity of the filtrates (liquid phase) was measured using a gamma counter (WIZARD^2, PerkinElmer Japan G.K.).

2.4 Effect of Ca concentration on Sr sorption

To evaluate the effect of calcium concentration on Sr sorption, simulated seawater solutions containing different Ca concentrations (0, 2.5, 5, 10 [equivalent to typical surface seawater], and 20 mM) were prepared. Artificial seawater (Daigo's Artificial Seawater SP, LABOLOGY Co., Ltd.) and natural seawater (collected in Ogasawara) were also analyzed for comparison. Each 100-mL sample was spiked with several kBq of ^{85}Sr, followed by the addition of 150 mg of P-MAq. The mixtures were stirred, and 2-mL aliquots were collected between 10 and 240 min, and subsequently filtered. The radioactivity of the filtrates was measured using the gamma counter.

2.5 Effect of Sr concentration on Sr sorption

To examine the influence of strontium concentration on sorption behaviour, simulated seawater solutions containing different stable Sr concentrations (0, 0.06, 0.09 [equivalent to typical surface seawater], 0.15, and 0.3 mM) were prepared. Each 100-mL sample was spiked with several kBq of ^{85}Sr. Following this, 150 mg of P-MAq was added and the mixture was stirred. During stirring, 2-mL aliquots were collected at intervals between 10 and 240 min and filtered. The radioactivity of the filtrates was measured using the gamma counter.

2.6 Effect of sulfate ion on Sr sorption

To investigate the effect of sulfate ions on Sr sorption, three types of solutions were prepared: sulfate-free freshwater, freshwater containing 25 mM sulfate (a typical concentration in surface seawater), and simulated seawater with varying sulfate concentrations. Each 100-mL sample was spiked with several kBq of ^{85}Sr, followed by the addition of 150 mg of P-MAq. The mixtures were stirred for 240 min, filtered, and the radioactivity of the filtrates was measured using the gamma counter.

2.7 The minimum detectable concentration

The minimum detectable concentration (MDC, Bq L^{-1}) was calculated by the Currie's equation [16];

$$MDC = \frac{2.33\sqrt{n_{BG} \times T}}{T} \times \frac{100}{Y} \times \frac{100}{\varepsilon} \times \frac{1}{V} \quad \text{(Bq L}^{-1}\text{)} \qquad (1)$$

where, T is the measurement time (s), n_{BG} is the background counting ratio (cps), Y is the chemical yield (%), ε is the counting efficiency (%), and, V is the sample volume (L).

3. Results and discussions

3.1 Effects of chemical parameters on Sr sorption

Figure 2 shows the time profile of the residual ^{85}Sr in the sample solution for different amounts of P-MAq. As shown in the figure, when 130 mg or more of P-MAq was used, the residual ^{85}Sr ratio decreased to approximately 10% after 4 h of stirring, corresponding to a sorption ratio of about 90%. These results indicate that 130 mg of P-MAq is sufficient for effective Sr sorption in seawater.

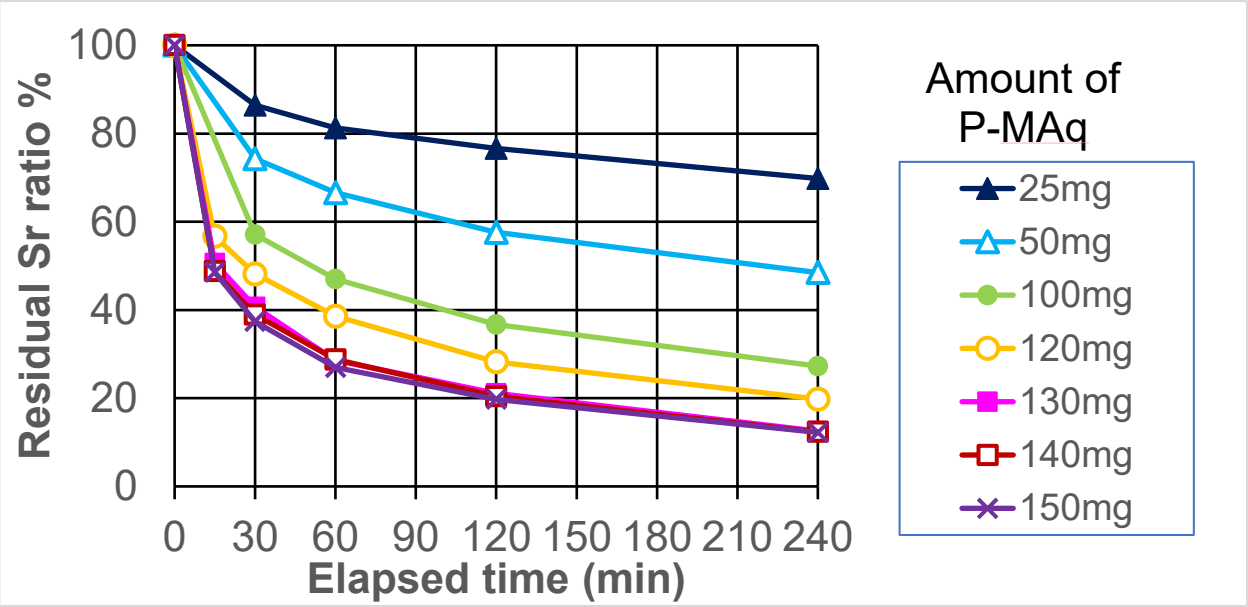


Fig. 2 Time profile of the residual Sr ratio in the sample solution during stirring.

Figure 3 shows the effect of Ca concentration on the sorption ratio of Sr on P-MAq. The higher the Ca concentration, the lower the Sr sorption ratio. There is a strong correlation between Ca concentration and Sr sorption. This relationship is expressed by the following equation:

$$S = -0.877C + 98.4 \quad (2)$$

where S is the Sr sorption ratio (%), C is the Ca concentration (mM) and the correlation coefficient was 0.9512, indicating a strong linear dependence. Because Ca concentrations in natural seawater vary slightly depending on sampling location, attention must be paid to Ca concentration when analyzing radiostrontium using this sorbent.

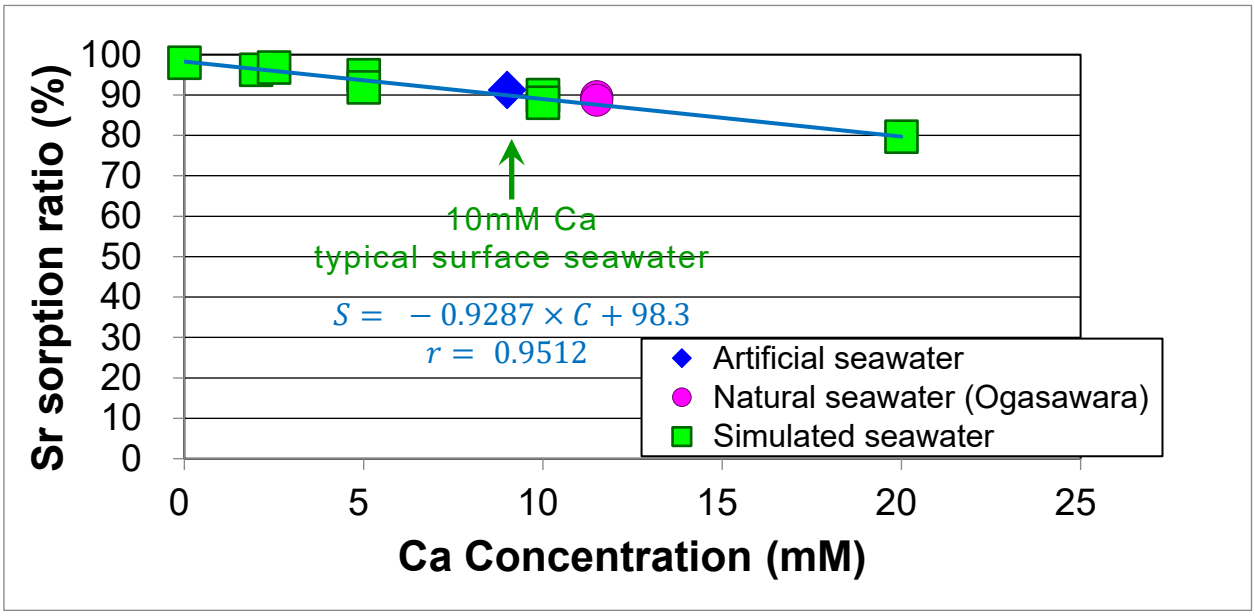


Fig. 3 Effect of Ca concentration on Sr sorption.

Figure 4 shows the time profiles of Sr sorption at different Sr concentrations. As observed in the figure, lower Sr concentrations resulted in faster sorption ratio. However, after 240 min of stirring, the sorption ratio reached similar final values regardless of the initial Sr concentration.

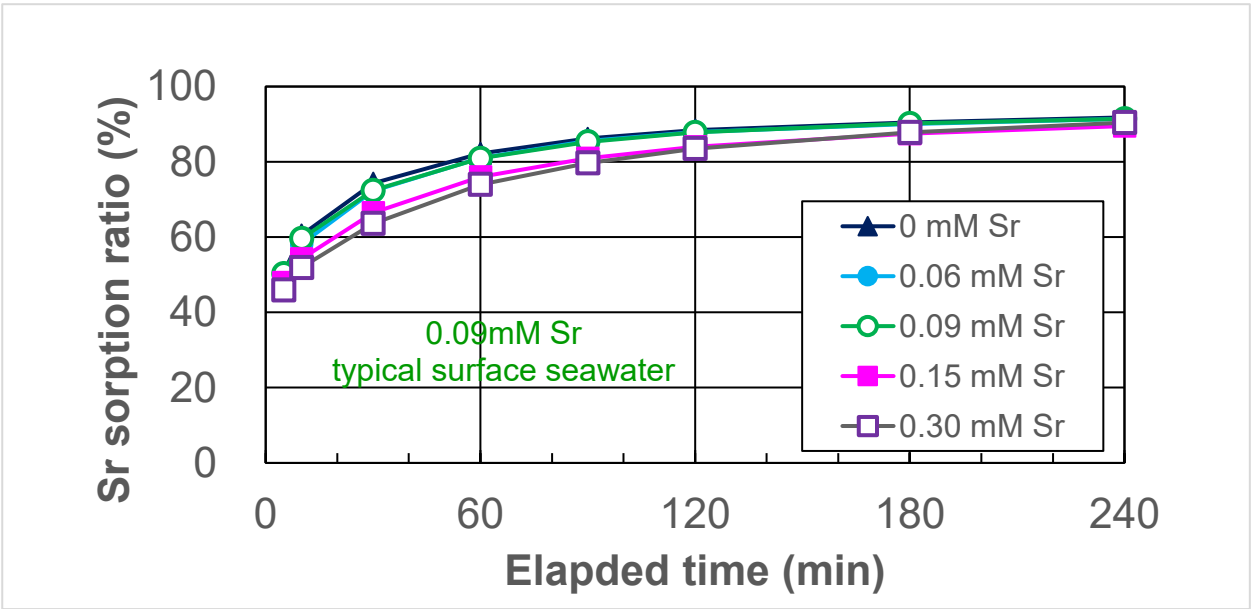


Fig. 4 Time profiles of Sr sorption ratio at different Sr concentrations.

Figure 5 illustrates the effect of sulfate ions. Sulfate-free freshwater exhibited no Sr sorption, whereas freshwater containing 25 mM sulfate showed more than 98% Sr sorption. In contrast, sulfate-free simulated seawater sorbed only about 20%, while simulated seawater with sulfate concentrations exceeding 10% of natural seawater achieved approximately 90% Sr sorption. Sr readily forms a sparingly soluble sulfate ($SrSO_4$) in aqueous solution. These results suggest that Sr precipitates in the form of sulfate complexes on the surface of P-MAq, and is subsequently adsorbed onto P-MAq.

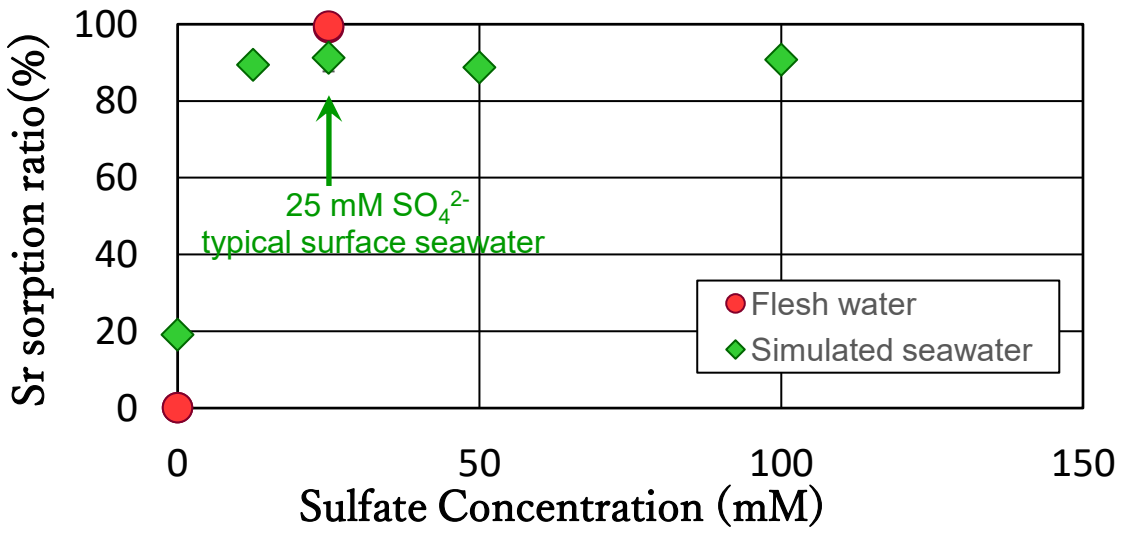


Fig. 5 Effect of sulfate ions on Sr sorption.

The P-MAq sorbs not only Sr but also Ra, Pb, and Ba. Naturally occurring radionuclides such as ^{226}Ra, ^{228}Ra and ^{210}Pb may potentially interfere with radiostrontium measurements. However, given the small sample volumes (a few liters of seawater) used in this method, their radioactivity is relatively low [17], and their presence does not significantly affect the radiostrontium analysis. Regarding barium, ^{140}Ba is a fission product that may be released during nuclear accident. Since ^{140}Ba is gamma emitter, it can be quantified by gamma-ray spectrometry and its contribution can be corrected accordingly.

3.2 The analytical methods

The recommended procedure for Sr analysis using P-MAq is as follows.

(1) Environmental waters are filtered through a membrane filter to remove suspended solids, and to obtain a solution equivalent to 0.1 M HCl, 2.5 mL of 4 M HCl was added to 1 L of sample.
(2) P-MAq (150 mg per 100 mL of solution) is added to each sample, and the mixture is stirred for 4 hours.
(3) The P-MAq is subsequently separated by suction filtration or centrifugation.
(4) The radioactivity of the P-MAq with adsorbing Sr is measured using a suitable detector.

Depending on the measurement equipment, these analytical methods are available:
(1) Low-Background Gas-Flow Counter Method (LBC),
(2) Plastic Scintillation Bottle Method (PSB), and
(3) Liquid Scintillator Method (LS)

Figure 6 illustrates the workflow of the low-background gas-flow counter method (LBC). P-MAq with sorbed Sr was collected on a filter paper (5C, 22-mm diameter; Advantech Co., Ltd.) by suction filtration. The filter paper was mounted on a planchet with adhesive, dried, and coated with a diluted collodion solution for protection. Radioactivity was measured using a low-background gas-flow counter (LBC-4601; ALOKA Co., Ltd.). As the permissible amount of P-MAq was 75 mg, the applicable volume of sample solution was 50 mL.

Figure 7 illustrates the workflow of the plastic scintillation bottle method (PSB) [7,10]. P-MAq with sorbed Sr was collected on a membrane filter (44-mmϕ diameter, 0.45-μm pore size; MF-Millipore HVLP04700, Millipore, Merck KGaA) by suction filtration. The filter was dried and sealed with polyethylene film to prevent contamination. Following this, it was sandwiched between two plastic scintillators and placed inside a plastic bottle. Radioactivity was measured using a low-background liquid scintillation counter (LSC-LB7, ALOKA Co., Ltd.). As the permissible amount of P-MAq was 150 mg, the applicable volume of sample solution was 100 mL.

Figure 8 shows the workflow of the liquid scintillator method (LS). P-MAq was separated by centrifugation and transferred into a counting vial. Following this, a scintillation cocktail, either an emulsion scintillator (Clear-sol II®, Nacalai Tesque, Inc.) or a gel scintillator (Ecoscint GL®, National Diagnostics), was added and mixed. Radioactivity

was measured using either a conventional liquid scintillation counter (LSC-6100, ALOKA Co., Ltd.) or a low-background liquid scintillation counter (LSC-LB7, ALOKA Co., Ltd.). Conventional LSCs accommodate the standard 20-mL vials, whereas low-background LSCs can measure large 140-mL vials. For the standard vial, the permissible amount of P-MAq is 1,500 mg, corresponding to an applicable sample volume of 1,000 mL. For the large vial, the permissible amount of P-MAq is 7,500 mg, corresponding to an applicable sample volume of 5,000 mL.

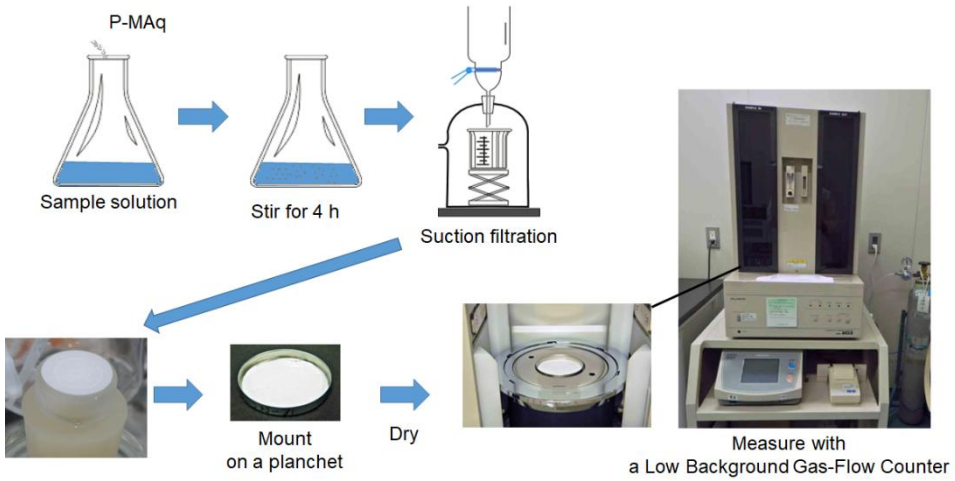


Fig. 6 Workflow of the Low Background Gas-Flow Counter method (LBC).

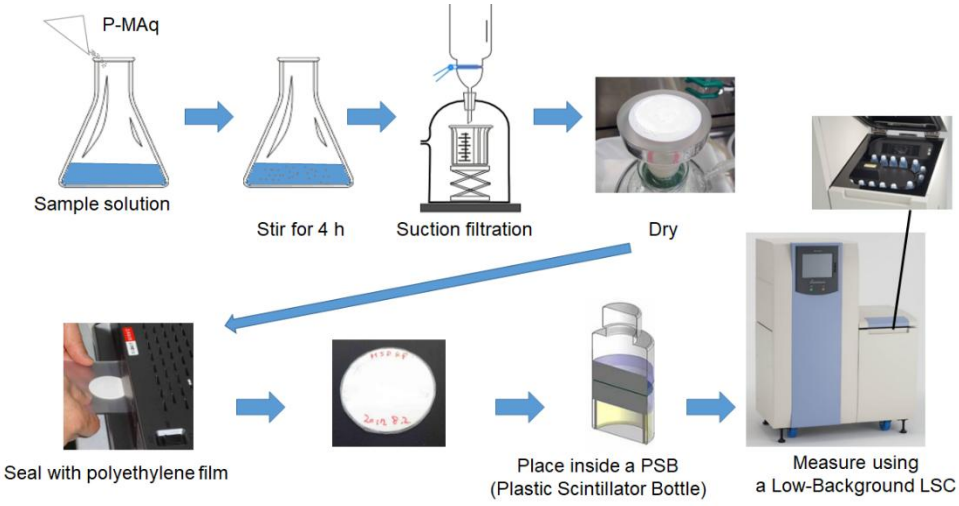


Fig. 7 Workflow of the Plastic Scintillation Bottle method (PSB).

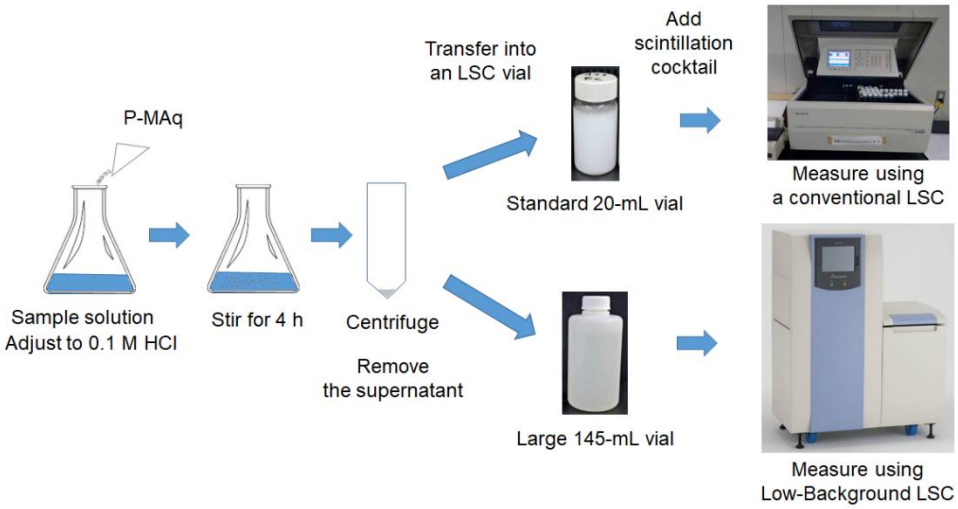


Fig. 8 Workflow of the Liquid Scintillator method (LS)

3.3 The minimum detectable concentration

The average Sr sorption ratio after 4 hours of stirring in simulated seawater was 90%. The MDC depends primarily on the volume of the sample solution, which is proportional to the amount of P-MAq used. Table 1 summarizes the MDC values for each method. The LBC method exhibited a maximum MDC of 0.4 Bq L^{-1}, whereas the LS method using large vials achieved a minimum MDC of 0.013 Bq L^{-1}.

Table 1 Minimum Detectable Concentration (MDC) for each method

Method	Counting efficiency (%)	BG (cpm)	P-MAq (mg)	Sample volume (mL)	MDC (Bq L^{-1})	MDC /Permissible value
LBC	10	0.12	75	50	0.4	1/75
PSB	40	8	150	150	0.4	1/75
LS (standard vial)	90	56	1,500	1,500	0.04	1/750
LS (large vial)		134	7,500	7,500	0.013	1/2,300

LBC: Low-Background Gas-Flow Counter; PSB: Plastic Scintillation Bottle; LS: Liquid Scintillator.

As shown in Table 1, the LBC method yielded the highest MDC of 0.4 Bq L^{-1},

corresponding to 1/75 of the permissible effluent standard of ^{90}Sr for wastewater in Japan (30 Bq L^{-1}). Because the required sample volume is small, the method is easy to perform and has the lowest operational cost. The LS method provides the lowest MDC and can be conducted using a conventional liquid scintillation counter. However, it generates organic liquid waste. In contrast, the PSB method does not generate organic liquid waste, and plastic scintillation bottles are reusable. Therefore, the appropriate method can be selected based on the target MDC, available measurement equipment, and facility constraints.

This analytical approach is simple: the procedure involves adding P-MAq to the liquid sample, stirring, and subsequently separating it. It requires no hazardous reagents and does not demand specialized skills. The procedure can be carried out using commonly available laboratory equipment. Considering all aspects, it is a rapid, simple, and safe method.

4. Conclusions

A newly developed analytical method for radiostrontium in environmental water using a Sr sorbent has been proposed. In this method, the sorbent is added to the sample solution, stirred for 4 hours, separated, and subsequently measured for 60 minutes. The MDC was less than 0.4 Bq L^{-1}, corresponding to a value lower than 1/75 of the permissible effluent standard for wastewater in Japan (30 Bq L^{-1}). The procedure provides results within 6 hours and does not use any hazardous reagents. Therefore, this method demonstrates its applicability to the rapid and safe analysis of radiostrontium in environmental water.

Acknowledgements

We express our deepest gratitude to the late Prof. Michio Aoyama for his invaluable cooperation and advice. We also thank Nippon Chemical Industrial Co., Ltd. for providing P-MAq for research purposes.

This research was conducted at the Radioisotope Center Medical Branch, Nagoya University; the Radioisotope Research Facility, the Jikei University School of Medicine; the Nuclear Medicine Laboratory, Aichi Medical University; and the Radioisotope Center, Ito Laboratory, Kyushu University. We are grateful to the staff of these facilities for their

support.

This research was partially supported by the following grants:

(1) Nuclear Regulation Authority, FY2021 Radiation Countermeasures Commission, "Development of a safe, rapid, and inexpensive analytical method for strontium in water";

(2) Environmental Radioactivity and Impact Assessment Network Joint Research Center (ERAN, Grant No. F-21-19, F-22-06, P-23-12); and

(3) JST FY2024 Grant-in-Aid for Scientific Research (C), Grant No. 24K08310.

References

[1] Ministry of Education, Culture, Sports, Science and Technology, Radioactivity Measurement Method Series 2: Radioactive Strontium Analysis Method (2003)

[2] Nora Vajda, Chang-KyuKim., Determination of radiostrontium isotopes: A review of analytical methodology, Applied Radiation and Isotopes, 68, 2306-2326 (2010)

[3] Empore Rad Disk, GL Sciences Inc. https://www.gls.co.jp/product/spe_columns/3m_empore/01163.html

[4] Battelle Press, Columbus, OH. Method RP515, Rapid determination of radiostrontium using Empore™ Strontium RAD Disks. (1997)

[5] Ometáková, J et. al., A comparison of classical ^{90}Sr separation methods with selective separation using molecular recognition technology products Analig® SR-01 gel, 3M Empore™ Strontium Rad Disk and extraction chromatography Sr Resin. J Radioanal. Nucl. Chem 290, 319–323. DOI 10.1007/s10967-011-1338-x (2011)

[6] M. Furukawa, Y. Takagai, Rapid Measurement of Strontium-90 Using ICPMS — HighPerformance Sequential Analyzer for β-ray Emitter Nuclide of Several Ten-Years Half-Life—, Radioisotopes, 67, 17-30 (2018) (in Japanese)

[7] H. Tazoe, et. al., Determination of strontium-90 from direct separation of yttrium-90 by solid phase extraction using DGA Resin for seawater monitoring, Talanta, 152, 219-277 (2016)

[8] Avramenko, V. A. et.al. Sorption Recovery of Strontium from Seawater. Radiochemistry 43, 433-436 (2001)

[9] Avramenko, V. A. et.al. Management of Liquid Radioactive Wastes Containing Seawater.

Pacific Science Review 15, 40-47 (2013)

[10] H. Minowa, Y. Ogata, Y. Kato, S. Kojima, Characterization of barium silicate-based adsorbent for Sr in seawater and terrestrial water, Proceedings of the 21st Workshop on Environmental Radioactivity, KEK Proceedings 2020-4, 176-179 (2020)

[11] S. Kojima, Y. Ogata, H. Minowa, Y. Kato, Characterization of barium silicate-based adsorbent for Ra and Pb in seawater, Proceedings of the 21st Workshop on Environmental Radioactivity、KEK Proceedings 2020-4, 186-191 (2020)

[12] Y. Ogata, S. Kojima, H. Minowa, Y. Kato, T. Arinobu, S. Sugihara, Rapid Measurement Method for Radiostrontium in Seawater and Land, Water using an Adsorbent composed with Barium Silicate, -Comparison of Measurement Methods-, Proceedings of the 24th Workshop on Environmental Radioactivity, KEK Proceedings 2023-2, 150-154 (2023)

[13] Y. Ogata, S. Kojima, H. Minowa, Y. Kato, T. Arinobu, S. Sugihara, Rapid measurement of radiostrontium in seawater and inland water using strontium sorbent, Proceedings of the 25th Workshop on Environmental Radioactivity, KEK Proceedings 2024-6, 129-133 (2024)

[14] H. Minowa, Y. Ogata, S. Kojima, Y. Kato, S. Sugihara, T. Arinobu, Development of rapid analysis of radiostrontium in environmental water using Sr sorption-reagent mainly composed with barium silicate I, Radioisotopes (Submitting).

[15] Y. Nozaki, Updated (1997 version) Table of the Elements in Seawater and its Remarks, Special issue on "Ocean Resources: Utilization and Issues", Bulletin of the Society of Sea Water Science, Japan, Vol 51, (5) 302-308 (1997)

[16] Currie LA. Limits for qualitative detection and quantitative detection. Anal. Chem. 40:586-593; 1968

[17] IAEA-TECDOC-1429, Worldwide marine radioactivity studies (WOMARS) Radionuclide levels in oceans and seas, Vienna, (2005)