



Repeatability and reproducibility of measurements of low dissolved radiocesium concentrations in freshwater using different pre-concentration methods

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Abstract

We assessed the repeatability and reproducibility of methods for determining low dissolved radiocesium concentrations in freshwater in Fukushima. Twenty-one laboratories pre-concentrated three of 10 L samples by five different pre-concentration methods (Prussian-blue-impregnated filter cartridges, coprecipitation with ammonium phosphomolybdate, evaporation, solid-phase extraction disks, and ion-exchange resin columns), and activity of radiocesium was measured. The *z*-scores for all of the ¹³⁷Cs results were within ± 2 , indicating that the methods were a good degree of precision. The relative standard deviations (RSDs) indicating the reproducibility among different laboratories were larger than the RSDs indicating the repeatability in each separate laboratory.

Keywords Measurement precision · Fukushima · River water · ¹³⁷Cs · ¹³⁴Cs · Ge-detector

Introduction

It has been necessary to measure dissolved radiocesium (¹³⁴Cs and ¹³⁷Cs) concentrations in freshwater near the Fukushima Daiichi Nuclear Power Plant (FDNPP) since the FDNPP accident to provide information to allow root uptake by crops and accumulation in aquatic organisms to be assessed. Many institutions have therefore monitored dissolved radiocesium concentrations in freshwater in northeastern Japan since the FDNPP accident [1–12].

Dissolved and particulate-bound radiocesium concentrations in freshwater in the northeastern Japan are mostly < 0.1 Bq L⁻¹, and dissolved radiocesium concentrations

of 0.001–0.1 Bq L⁻¹ have been found at most monitoring points in the northeastern Japan [9, 12–15]. The low concentrations mean that effective quantitative measurements of dissolved radiocesium in freshwater require samples to be pre-concentrated before analysis. Environmental radioactivity measurement manuals published by the Japanese Ministry of Education, Culture, Sports, Science and Technology contain methods for measuring dissolved radiocesium in water involving either evaporating samples to dryness or ion exchange [16]. Other methods involve ammonium phosphomolybdate (AMP) coprecipitation, particularly for very saline samples, and instrumental analysis using a germanium semiconductor detector (Ge-detector) [17]. These methods have been used to quantitatively determine dissolved radiocesium in freshwater for a long time (since before the FDNPP accident). New pre-concentration methods were developed after the FDNPP accident, including using cartridges impregnated with Prussian blue (PB) [18–21] and solid-phase extraction disks [22, 23], to decrease the time required for pre-concentration and solid–liquid separation. These new concentration methods dramatically decreased

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the time required for analyzing samples, so have been used widely.

The precision achieved when analyzing radiocesium using different methods has been assessed previously. Five methods (involving evaporation to dryness, AMP coprecipitation, PB cartridges, solid-phase disks, and ion exchange resin) were used to analyze three water samples (two ion-exchanged water samples spiked with radiocesium and one freshwater sample) containing dissolved radiocesium at concentrations of $0.01\text{--}1.0\text{ Bq L}^{-1}$ by 18 laboratories [24]. The relative standard deviations (RSDs) were $0.08\text{--}0.119$, and the z -scores for $>80\%$ of the results were within ± 2 . The results indicated that the pre-concentration methods (including the new methods) are good degree of precision. However, only one sample was analyzed repeatedly, so the results did not indicate whether the slight variations in the concentrations were caused predominantly by variability in the results from each laboratory or by variability between the results from different laboratories. The results also did not indicate whether variability between the results from different laboratories may have been caused by different pre-concentration methods being used by the different laboratories.

In the study presented here, we evaluated the variability in dissolved ^{134}Cs and ^{137}Cs concentrations in freshwater samples determined by each participating laboratory (repeatability) and variability in the concentrations determined by the different laboratories (reproducibility). Each participating laboratory analyzed three samples under the same conditions, then all the samples were analyzed in one laboratory to remove the effect of using different instruments at different laboratories on the reproducibility of the results. We assessed the repeatability and reproducibility and the factors affecting the reproducibility of the results from different laboratories.

Materials and methods

Water sample collection

River water was collected from the Abukuma Kogen region in Japan on 25 October 2017 under base flow condition. Most radiocesium in the sample was supplied by the FDNPP accident in March 2011. Approximately 1200 L of river water was collected in six 200 L tanks. The water was pumped from the river at the center of a bridge. The water was immediately transported to the laboratory, where it was passed through A045A $0.45\text{ }\mu\text{m}$ membrane filters (Advantec MFS, Dublin, CA, USA) on 26 October 2017. The filtered water in a 1200 L tank was stirred by passing it through a pump at a flow rate of 3 L s^{-1} for $>12\text{ h}$. On 27 October 2017, 50 L of well-mixed water was removed using a washed plastic bucket, and three 10 L polyethylene flexible

tanks were filled with the water. The flexible tanks were then refrigerated until they were sent to a participating laboratory. This was repeated for each participating laboratory. The samples were sent to the participating laboratories on 7 November 2017. Each partitioning laboratory received its allocation of three 10 L samples on 8 November 2017 and immediately stored the samples in a refrigerator. We indicated the procedure for sample storage keeping in a refrigerator but did not indicate the acidic treatment because of limiting condition of pre-treatment method. Notable procedures for sample storage for each laboratory are shown in Table S1.

The mean \pm standard deviation Na^+ concentrations, Ca^{2+} concentrations, pH values, and electrical conductivities for the 10 L samples were $3.5 \pm 0.06\text{ mg L}^{-1}$, $2.3 \pm 0.06\text{ mg L}^{-1}$, 7.2 ± 0.05 , and $3.7 \pm 0.05\text{ mS m}^{-1}$, respectively. We concluded that all the samples sent to the participating laboratories were well mixed because similar Na^+ concentrations, Ca^{2+} concentrations, pH values, and electrical conductivities were found by the different laboratories.

Pre-concentration methods and radioactivity measurements

Five pre-concentration methods (PB filter cartridges, AMP coprecipitation, evaporation, solid-phase extraction disks, and ion-exchange resin) were used by the partitioning laboratories (Fig. 1). The test involved ^{134}Cs and ^{137}Cs concentrations in 68 water subsamples being determined by 21 Japanese laboratories. Each laboratory except one pre-concentrated and determined the radioactivity in three water samples (one laboratory only analyzed two samples, see Table 1). Two laboratories analyzed six water subsamples using two concentration methods, and these laboratories were each given two laboratory ID numbers.

The five pre-concentration methods are described below. The PB filter cartridge method takes advantage of PB ($\text{KFe}[\text{Fe}(\text{CN})_6]_3 \cdot x\text{H}_2\text{O}$) being able to adsorb dissolved radiocesium with a good degree of specificity [18–21]. The PB filter cartridge method involved removing dissolved radiocesium from water passed through a nonwoven fabric impregnated with PB in a cartridge. A water sample was passed through a cartridge at a flow rate of 2.5 L min^{-1} by pumping, and ^{134}Cs and ^{137}Cs radioactivity in each cartridge was then measured using a Ge-detector with a special cartridge container. As distribution of radiocesium in the cartridge is not uniform, geometric correction was performed to evaluate concentration [21]. Seven laboratories used potassium zinc ferrocyanide PB cartridges [21], and one laboratory used copper-substituted PB cartridges [25].

The AMP coprecipitation method takes advantage of the ability of AMP to adsorb dissolved radiocesium with a good degree of specificity [17]. Each water sample was adjusted

Fig. 1 Five pre-concentration methods (PB filter cartridges, AMP coprecipitation, evaporation, solid-phase extraction disks, and ion-exchange resin) used by the partitioning laboratories

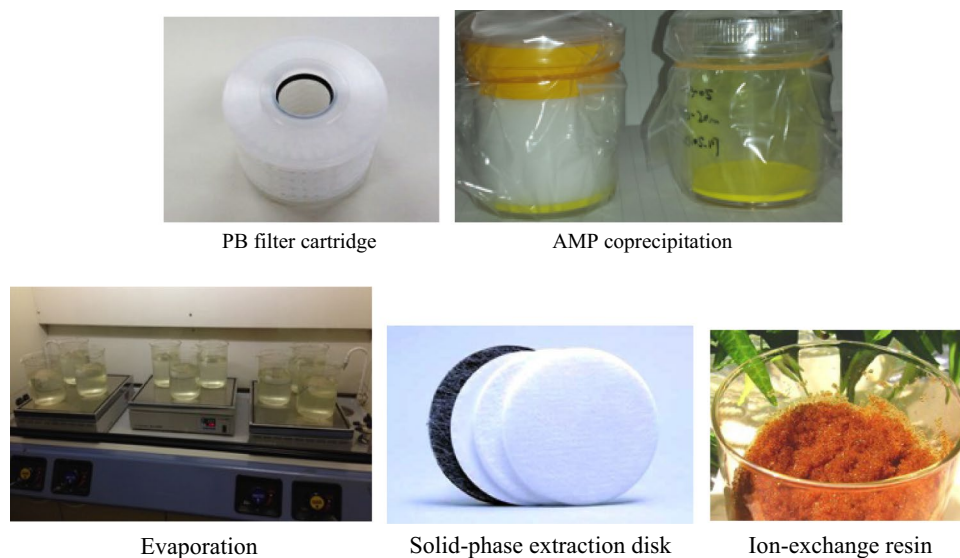


Table 1 Number of laboratories using each pre-concentration method and the number of samples analyzed using each pre-concentration method

Pre-concentration method	No. of laboratories ^a	No. of samples
Prussian blue filter cartridge	8	24
Ammonium phosphomolybdate coprecipitation	6	18
Evaporation	5	15
Solid-phase extraction disk	3	9
Ion exchange resin	1	2
Total	23	68

^aThe total number of laboratories is not the same as the number of participating laboratories

to ~pH 1.5 before analysis. The radiocesium in the sample was quantitatively removed from the water sample by coprecipitation as AMP/Cs compounds once AMP had been added. The AMP/Cs compounds were removed by filtering the sample, then dried and transferred to a small container suitable for radioactivity measurement (e.g. 100 ml with 5-cm diameter plastic container) before the radioactivity was measured using a Ge-detector.

The evaporation method is to measure concentrated radiocesium in a water sample by evaporation [16]. In the evaporation method in the present test, a water sample was evaporated in a glass beaker on a hot plate but not boiled excessively. Each sample was evaporated to a volume of between 50 mL and 2 L, then transferred to a 100 mL container or a 2 L Marinelli beaker before the radioactivity was measured using a Ge-detector. One laboratory acidified the samples before the evaporation method was performed.

In the solid-phase extraction disk method, dissolved radiocesium was removed from a water sample passed through a solid-phase extraction disk containing ion-selective resin on a Teflon support [26, 27]. A water sample was passed through a disk using a pressure filtration system, and the ¹³⁴Cs and ¹³⁷Cs radioactivity in the disk was measured using a Ge-detector.

In the ion exchange resin method, dissolved radiocesium is collected to a cation exchange resin from a water sample [16]. The ion exchange resin method in the present test was performed by passing a water sample through a 100 mL column containing a cation exchange resin in the Na⁺ form. The resin containing the radiocesium was then transferred to a 100 mL container and the radioactivity was measured using a Ge-detector.

The more detailed information of each pre-concentration methods have been described previously [21, 22, 24, 28, 29]. The specific differences in procedures among laboratories are summarized in Table S1.

The laboratories pre-concentrated the samples between 8 and 25 November 2017 and determined the radioactivity of ¹³⁴Cs and ¹³⁷Cs in the samples by 10 January 2018. The radioactivity measurements were made in the laboratories by using their own Ge-detectors. Each laboratory followed its usual operating procedures for determination of low dissolved radiocesium concentrations in freshwater (e.g., sample storage procedure and measurement time). Radioactivity of ¹³⁴Cs and ¹³⁷Cs were decay corrected to 12:00 JST 11 November 2017 to allow the results to be compared. The different pre-concentration methods gave samples with different shapes, so each laboratory calibrated its detector(s) using a standard radioactive source (a special customized source was used for each sample of the same type). Finally, in order to discriminate effects of radioactivity measurement

by different detectors from different procedures for sample treatment among laboratories, the samples were sent to one laboratory (Tokyo Power Technology Ltd., Chiba, Japan), where the ^{137}Cs radioactivity in each sample was measured using a single Ge-detector (using a 43,000 s measurement period).

Statistical analysis

The ^{134}Cs and ^{137}Cs concentrations found in the water samples were evaluated using z -scores, as described in a previous publication [30] and in the IAEA-JAPAN proficiency test [31]. Each z -score was calculated using the equation

$$Z = (x_i - X) / s, \quad (1)$$

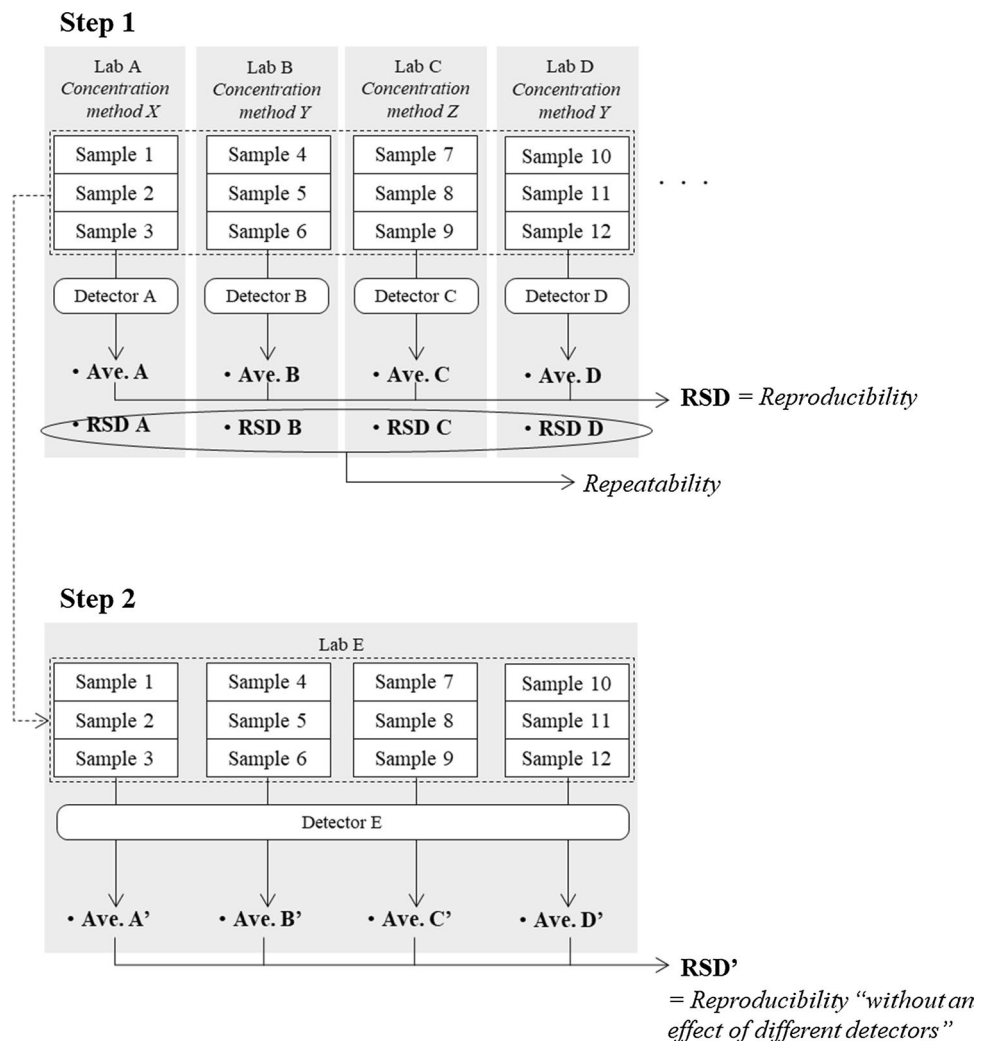
where x_i is the concentration in water sample i , X is the robust average, and s is the robust standard deviation. The robust average and robust standard deviation were calculated using algorithm A1, from JIS Z 8405 [32].

The repeatability and reproducibility were assessed using the relative standard deviation (RSD), as described in a previous publication [30]. Each RSD was calculated using the equation

$$\text{RSD} = s / X. \quad (2)$$

The repeatability (indicating the precision of repeated analyses) for each laboratory was assessed using the RSD for that laboratory ($n=3$). The reproducibility (indicating variability between the results from different laboratories) was assessed using the RSD calculated from the robust average concentrations produced by the laboratories, as shown in Fig. 2.

Fig. 2 Scheme used to determine the repeatability and reproducibility relative standard deviations (RSD)



Results and discussion

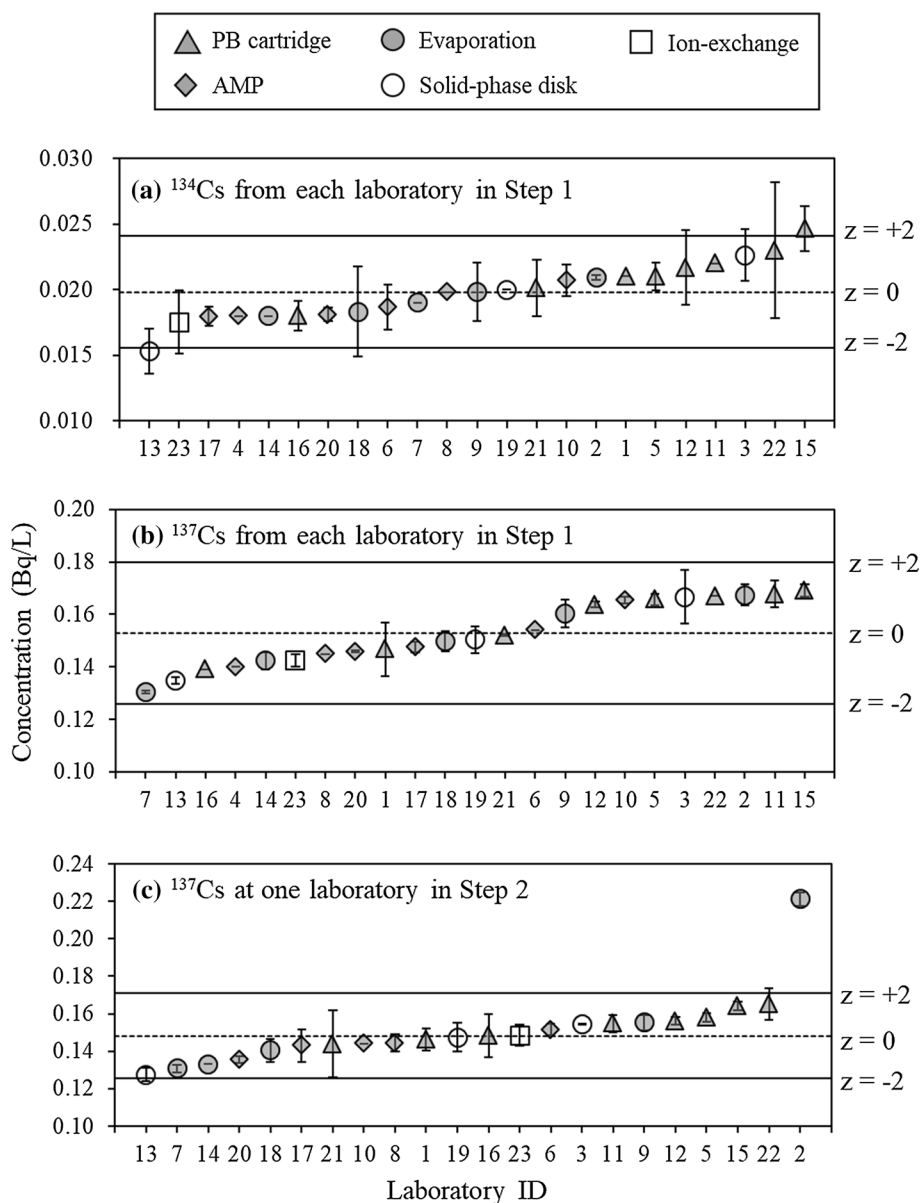
Assessment of the precision of the results

The robust averages \pm the standard deviations of the ^{134}Cs and ^{137}Cs concentrations determined by the laboratories were 0.020 ± 0.002 and 0.153 ± 0.013 Bq L^{-1} , respectively. The z -scores were within ± 2 . The ^{134}Cs and ^{137}Cs concentrations were 0.015 – 0.025 and 0.131 – 0.169 Bq L^{-1} , respectively, and the results produced by the different laboratories were not markedly different. The z -scores for $> 95\%$ of the ^{134}Cs results and all of the ^{137}Cs results were within ± 2 (Fig. 3a, b), suggesting that there was good degree of

reproducibility between the methods used, as has been found previously [24].

The robust average ^{134}Cs and ^{137}Cs concentrations determined using the five different pre-concentration methods were not markedly different (Fig. 4a, b). The ^{134}Cs and ^{137}Cs concentrations determined using the PB cartridge, AMP coprecipitation, evaporation, and solid-phase disk methods did not show statistically significant difference (one-way ANOVA, $p < 0.005$, the concentrations determined using the ion-exchange resin method was not included in the analysis because only one laboratory used for the determination). This indicated that differences in the concentrations found by the different laboratories were not caused by the laboratories using different pre-concentration methods. The concentrations found using the different pre-concentration methods

Fig. 3 Robust average **a** ^{134}Cs concentrations and **b** ^{137}Cs concentrations from each laboratory in Step 1, and **c** ^{137}Cs concentrations at laboratory in Step 2. The error bars indicate the standard deviations



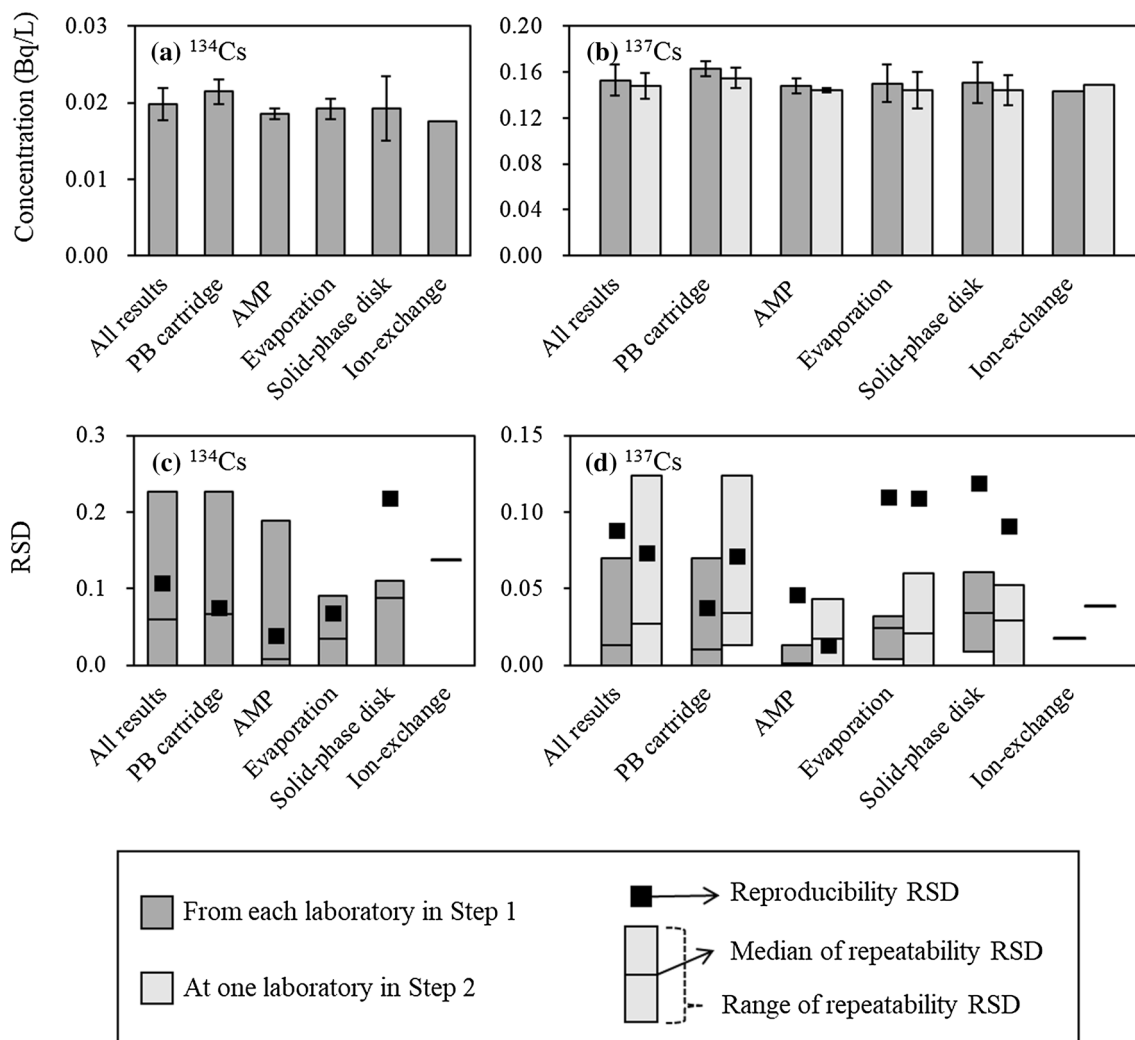


Fig. 4 Robust average **a** ^{134}Cs concentrations and **b** ^{137}Cs concentrations for the different pre-concentration methods. The error bars indicate the standard deviations. Comparison between repeatability and reproducibility relative standard deviations (RSDs) for **c** ^{134}Cs con-

centrations and **d** ^{137}Cs concentrations for the different pre-concentration methods. Bar charts indicate range of repeatability RSDs and middle lines of the bars are medians. Solid square symbols indicate reproducibility RSD

were not statistically significantly different, but the ^{134}Cs and ^{137}Cs concentrations were higher when the PB cartridge method was used (at 0.021 and 0.163 Bq L⁻¹, respectively) than when the other pre-concentration methods were used. Similar results were found in the previous multi-laboratory evaluation [24]. However, the participating laboratories did not perform the entire analytical processes following exactly the same procedures even when the same pre-concentration methods were used. We cannot therefore conclude that the different radiocesium concentrations found by the different laboratories were caused by different pre-concentration methods being used.

Repeatability RSD

The ranges and median repeatability RSDs for the participating laboratories are shown in bar charts in Fig. 4c, d. The repeatability for the laboratories can be compared from the figure. For all the results, the ^{134}Cs repeatability RSDs were 0–0.226 (median 0.059), and the ^{137}Cs repeatability RSDs were 0–0.070 (median 0.013). The repeatability RSDs were different for the different laboratories, suggesting that repeatability was different for different laboratories. Like for the concentrations, the repeatability RSDs for four of the pre-concentration methods did not show significant difference (one-way ANOVA, $p < 0.005$, the concentrations determined using the ion-exchange resin method

was not included in the analysis because only one laboratory used for the determination). The repeatability RSDs for the laboratories using the AMP method (median 0.008 for ^{134}Cs and median 0.001 for ^{137}Cs) were smaller (but not statistically significantly smaller) than the repeatability RSDs for the laboratories using the other pre-concentration methods. This may have been because the laboratories using the AMP method have used it for a long time (since before the FDNPP accident), meaning they have been able to minimize variability between repeat analyses [24].

Reproducibility RSD and analyses of all the samples by one laboratory

The reproducibility RSDs for each pre-concentration method and for the results for all the pre-concentration methods combined are shown in solid square symbols in Fig. 4c, d. The reproducibility RSDs indicate variability between the results produced by the different laboratories. The reproducibility RSDs for all the ^{134}Cs and ^{137}Cs results were 0.108 and 0.088, respectively. The median ^{134}Cs and ^{137}Cs reproducibility RSDs were 0.059 and 0.013, respectively, indicating that variability between laboratories was generally larger than variability within a laboratory. The reproducibility RSDs for the different pre-concentration methods were not markedly different, but the reproducibility RSDs were lower for the AMP method than for the other methods. This indicated that the methods offered good reproducibility between laboratories as well as within each laboratory.

The results shown above do not describe the separate effects of different pre-concentration methods, Ge-detectors, and other factors on reproducibility. Comparisons of ^{137}Cs concentration and RSD between from each laboratory and at one laboratory are shown in Figs. 3c and 4b, d. The robust average ^{137}Cs concentration \pm the standard deviation was $0.145 \pm 0.011 \text{ Bq L}^{-1}$ and the reproducibility RSD was 0.073. The average was not different from the averages for the different laboratories. The ^{137}Cs concentrations for the PB cartridge, AMP coprecipitation, evaporation, and solid-phase disk methods were not statistically significantly different (one-way ANOVA, $p < 0.005$).

The robust averages and standard deviations for the ^{137}Cs concentrations measured at a single laboratory are shown in Fig. 2c. The average concentrations were similar to the concentrations by each laboratory shown in Fig. 2b except for laboratory 2. The reproducibility RSDs measured at a single laboratory were almost the same as the reproducibility RSDs for the different laboratories (Fig. 3d). This suggests that the differences in the results from the different laboratories were not predominantly caused by the different pre-concentration methods and different Ge-detectors used but rather by other factors. The z -scores for $> 90\%$ of the ^{137}Cs results were within ± 2 , suggesting that the pre-concentration

methods used by the laboratories all offered a good degree of precision.

Conclusions

The measurement precision with which low dissolved radiocesium concentrations in freshwater samples were determined by 21 Japanese laboratories was assessed. The repeatability and reproducibility of the results were assessed. All the pre-concentration methods used by the different laboratories gave a good degree of precision, indicating that even techniques used by Japanese laboratories to quantitatively determine low dissolved radiocesium concentrations only since the FDNPP accident in 2011 were high precision. Variability in the concentrations found by different laboratories was larger than variability in the concentrations found by each laboratory. Variability between laboratories was probably not caused by different pre-concentration methods or Ge-detectors being used but by other differences in the sample treatment procedures. This should be investigated further. Few evaluations of the measurement precision of methods used to determine low dissolved radiocesium concentrations in freshwater focused on repeatability and reproducibility have been performed. The results presented here therefore improve our understanding of the precision of such measurements.

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Compliance with ethical standards

Conflict of interest The authors declare that they have no conflict of interest.

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