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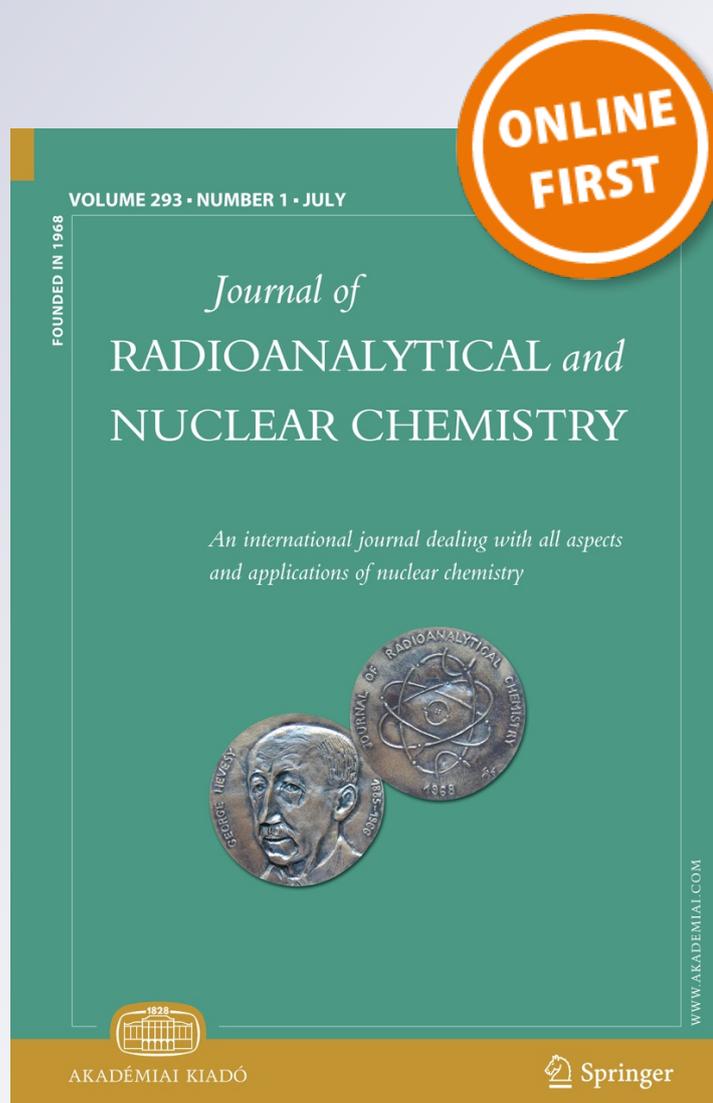
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Fast concentration of dissolved forms of cesium radioisotopes from large seawater samples

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Abstract The method developed for cesium concentration from large freshwater samples was tested and adapted for analysis of cesium radionuclides in seawater. Concentration of dissolved forms of cesium in large seawater samples (about 100 L) was performed using composite absorbers AMP-PAN and KNiFC-PAN with ammonium molybdophosphate and potassium–nickel hexacyanoferrate(II) as active components, respectively, and polyacrylonitrile as a binding polymer. A specially designed chromatography column with bed volume (BV) 25 mL allowed fast flow rates of seawater (up to 1,200 BV h⁻¹). The recovery yields were determined by ICP-MS analysis of stable cesium added to seawater sample. Both absorbers proved usability for cesium concentration from large seawater samples. KNiFC-PAN material was slightly more effective in cesium concentration from acidified seawater (recovery yield around 93 % for 700 BV h⁻¹). This material showed similar efficiency in cesium concentration also from natural seawater. The activity concentrations of ¹³⁷Cs determined in seawater from the central Pacific Ocean were 1.5 ± 0.1 and 1.4 ± 0.1 Bq m⁻³ for an off-shore (January 2012) and a coastal (February 2012) locality, respectively, ¹³⁴Cs activities were below detection limit (<0.2 Bq m⁻³).

Keywords Cesium · ¹³⁴Cs · ¹³⁷Cs · Seawater · Composite ion-exchangers

Introduction

Cesium concentration from larger sample volumes is usually required for radioanalytical determination of ¹³⁴Cs ($T_{1/2} = 2.07$ years) and ¹³⁷Cs ($T_{1/2} = 30.08$ years) in seawater. Some of the well-established methods include cesium adsorption on ammonium molybdophosphate (AMP) (e.g., [1–3]) and co-precipitation with various insoluble hexacyanoferrates(II) (e.g., [4–6]). Applications of these materials in chromatography columns for seawater analysis were reviewed by Gaur [7]. Insoluble hexacyanoferrates(II) were also used in impregnated cartridges (e.g., [1, 8, 9]) that as well as the chromatography columns usually reduced chemicals consumption and analysis time. Acidification of seawater is required in the methods where AMP is used due to its chemical stability. Insoluble hexacyanoferrates(II) can be used for concentration of cesium directly from non-acidified seawater, but lower pH may help reducing the uptake of other radionuclides [10] potentially interfering with gamma counting by scintillator detectors with lower energy resolution.

The historical principal sources of ¹³⁷Cs to seawater were nuclear tests atmospheric fallout, reprocessing plants, fallout from accident of Chernobyl nuclear power plant, and solid waste dumping at sea [11]. The level of ¹³⁷Cs in the North Pacific Ocean was 2.5 Bq m⁻³ in 2001 (median from decay corrected values from 1995 to 2001) [12], which correspond to 1.9 Bq m⁻³ in 2011 (before Dai-ichi accident). Lower real values could be expected due to the fact that ¹³⁷Cs apparent half-live in the Pacific is lower than the decay half-live [13]. Buessler et al. [14] estimated that

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the accident at the reactors of the Dai-ichi nuclear power plant in March 2011 became the largest accidental source of radionuclides to the ocean. Recently, Bailly du Bois et al. [11] proposed that 12–41 PBq of ^{137}Cs was discharged to the Pacific Ocean between March 25 and July 28, 2011 and predicted an increase of about 6 Bq m^{-3} after dilution in the surface ocean.

Use of inorganic ion-exchangers in the column application is often limited by their unsuitable granulometric and mechanical properties. The disadvantage could be overcome by preparation of composite inorganic ion-exchangers with modified polyacrylonitrile (PAN) as a binding matrix [15]. Composite absorbers of this type and their applications were summarized earlier [15, 16]. Based on previous studies (e.g., [17–19]) AMP-PAN and KNiFC-PAN were selected for cesium concentration from seawater. These materials were already used for fast determination of cesium radionuclides in large samples of various freshwater samples (AMP-PAN) [17] and in milk [20] and urine [21] samples (KNiFC-PAN).

The aim of this work was to test and compare AMP-PAN and KNiFC-PAN for sensitive determination of ^{134}Cs and ^{137}Cs in seawater. The main motivation was to develop a method that could monitor these radionuclides in the central Pacific Ocean after accidental releases in 2011. The work was focused on cesium concentration from about 100 L of sample of acidified seawater and in the case of KNiFC-PAN also from natural seawater. Stable cesium tracer was added for the determination of chemical recoveries.

Experimental

Materials and instruments

Seawater from the central Pacific Ocean was sampled offshore and at a coastal location. The offshore samples were collected on monthly cruises of Hawaii Ocean Time-Series (HOT) program to the Station ALOHA ($22^{\circ}45'\text{N}$, $158^{\circ}00'\text{W}$), the coastal samples were collected in south O'ahu, Hawaii ($21^{\circ}15.83'\text{N}$, $157^{\circ}49.33'\text{W}$).

For acidification of seawater nitric acid of technical grade (Fisher Chemical) was used. The working solution of stable cesium tracer was prepared from cesium chloride salt (ultra-pure grade, Amresco) by dissolving in deionized (DI) water. The solutions of ^{134}Cs and ^{137}Cs were prepared from Standard Radionuclide Sources (Eckert and Ziegler Analytics) by dilution in 0.5 M HNO_3 .

The composite inorganic ion-exchangers AMP-PAN and KNiFC-PAN were commercially produced at Czech Technical University in Prague. They were prepared by incorporation of “powdered” ($<0.1 \text{ mm}$) active component

AMP and KNiFC (potassium–nickel hexacyanoferrate(II)) into a binding matrix of PAN [15]. The AMP-PAN material was stored in 0.1 M HNO_3 and the KNiFC-PAN was stored in DI water.

Seawater samples were filtered using 125 mm long DCCPY wound cotton Micro-wynd II cartridge (Cuno) using Geopump Series II Peristaltic Pump (Geotech). The same pump was used for column experiments (silicone tubing Masterflex 15).

The column was originally developed for processing of about 100 L aqueous samples [17]. It was made from Plexiglas with internal diameter of 42 mm (Fig. 1). A perforated Teflon disk was used as the bed support together with a disk of either Whatman GF/C filter (pore size $1.2 \mu\text{m}$, diameter 47 mm) or polyamide sieve Uhelon (openings size 0.1 mm, diameter 50 mm).

The concentration of stable cesium used as a yield tracer was determined using a high performance double focusing magnetic sector field ICP-MS machine Element 2 (Thermo Finnigan). The sample inlet system included a Meinhard nebulizer with an uptake rate of approximately 1 mL min^{-1} and a Scott type spray chamber. A set of seawater samples (acidified to pH 1) of various cesium concentrations was used for the construction of calibration curves that were found linear for the range of cesium concentrations used ($0.5\text{--}40 \text{ ng mL}^{-1}$). The aliquots from experiments with natural seawater were acidified to pH 1 after sampling. All seawater samples were 25-fold diluted with ultra-pure DI water ($>18 \text{ M}\Omega \text{ cm}$) prior to cesium analysis.



Fig. 1 Column used for concentration of cesium from 100 L of seawater (internal diameter 42 mm, 25 mL of AMP-PAN)

Gamma-spectrometric measurements were performed using a coaxial HPGe detector (relative efficiency 43 %, resolution 1.76 keV for 1.33 MeV gamma line of ^{60}Co) connected to a DSPEC LF multichannel analyzer (both ORTEC). The recorded spectra were analyzed by Hypermet-PC ver 5.01 software [22]. The absorber in dry form was counted in a plastic Petri dish (47 mm diameter; FisherBrand) with the inner volume of about 12 mL that was sealed with an adhesive tape. The counting efficiency was determined by standard prepared from radionuclide solutions and the same amount of absorber. The minimum detectable activities (MDA) were calculated from the recorded spectra according to methods described by Currie [23].

Seawater pretreatment

The known amount of seawater sample was filtered to an empty 120 L plastic drum. For acidified samples, seawater was acidified by nitric acid (6 mL HNO_3 per 1 L of sample) and cesium carrier was added. The sample was mixed with a plastic rod and seawater was allowed to equilibrate for at least one hour.

Concentration of cesium

A disk of GF/C filter (alternatively polyamide sieve) was placed on the bottom part of the column and secured by screwing on the middle part of the column. Later, the column was filled with DI water and the bottom part of the column was degassed by the use of a syringe connected to the column outlet equipped with a stopcock. The bed was formed by sedimentation of 25 mL of the absorber in 0.1 M HNO_3 (alternatively in DI water for experiments with natural seawater) and tapping the column holder with still some layer of solution above the absorber. The KNiFC-PAN absorber was conditioned with 0.1 M HNO_3 solution for 20 min prior the bed formation for experiments with acidified seawater. Before the column was closed, it was carefully filled with the same solution that was used for bed formation. The top part of the column was filled by the use of the breather stopper and syringe with long needle. The inlet tubing filled with the sample solution was connected to the column, the outlet stopcock was opened, and the pumping was started. The downward flow-rate was regulated by pumping intensity and the stopcock on the column outlet. At the end of the experiment, the column was drained and the absorber was washed with two 10 mL portions of either 0.1 M HNO_3 (AMP-PAN) or DI water (KNiFC-PAN). For gamma counting the absorber was transferred into a disposable polystyrene beaker, homogenized by orbital shaking of the beaker, dried by elevated temperature (infra-red lamp), and transferred to a Petri

dish. A GF/C filter was placed on the top of the absorber to produce its uniform layer. Weighed part of the dry KNiFC-PAN resin was used for gamma-spectrometry to ensure the same counting geometry as for AMP-PAN (total dry volume of KNiFC-PAN was higher than volume of the counting dish).

Recovery yield and breakthrough determination

For the determination of cesium recovery yield R , 4.5 mL aliquots of the initial feed and seawater after treatment (well mixed) were sampled to glass vials. R (%) was calculated according to the Eq. (1)

$$R = (c_0 - c_f)/c_0 \times 100 \quad (1)$$

where c_0 is the initial cesium concentration in the feed and c_f is the final concentration in the eluate (ng mL^{-1}).

Similarly, several aliquots of the eluate were sampled during the experiment at certain intervals. Cesium breakthrough was calculated as the ratio of cesium concentration in the eluate c and in the initial feed c_0 (ng mL^{-1}) and it was related to the volume of processed sample estimated from average flow-rate and sampling time.

Results and discussion

In order to achieve short processing times and higher counting efficiencies, the flow-rate of the load solution should be optimized for low bed volume (BV) of the absorber used. Model experiments with cesium separation from drinking and surface freshwater by AMP-PAN (without stable cesium addition) showed that 99 % of cesium recovery could be achieved for a 4,000 BV sample if the flow-rate was not higher than $1,000 \text{ BV h}^{-1}$ [17]. Based on these results a column with 25 mL of AMP-PAN was proposed for large samples (about 100 L) with the maximum flow-rate of about 420 mL min^{-1} . The column was also used within this work for cesium concentration from seawater (Fig. 1). The dimensions of the column allowed to form a bed with a height of 1.8 cm ($\text{BV} = 25 \text{ mL}$). Mixing of the absorber bed was prevented by installing a water deflector to the column inlet. Seawater samples were run at a slightly reduced flow-rate in comparison to fresh-water samples [17] (approximately 300 mL min^{-1}) because of a higher content of alkali ions in seawater and the use of stable cesium tracer for recovery yield determination.

Efficiency of cesium extraction on the absorbers

The composite inorganic ion-exchangers studied here are listed in Table 1 together with their basic characterization.

Table 1 The composite ion exchangers used and their basic characterization

Material	Content of active component (%)	Grain size (mm)	Dry matter ^a (g mL ⁻¹)
AMP-PAN	80	0.1–0.7	0.27 ± 0.01 ^b
KNiFC-PAN	80	0.1–0.6	0.20 ± 0.01 ^c

^a Weight of dry absorber in 1 mL of original bed

^b Average ± standard deviation (7 determinations)

^c Average ± standard deviation (11 determinations)

The absorber materials were prepared with the same content of active component and with similar grain size. The dry matter in the BV was larger for AMP-PAN (6.8 g) than for KNiFC-PAN material (4.9 g).

The recovery yield of cesium was determined by the analysis of stable cesium in the initial load and final eluate solutions. Natural cesium concentration in seawater is approximately 0.4 µg L⁻¹ [24]. A stable cesium tracer was added in amounts exceeding this concentration so that a confident analysis of the eluate solution in which the cesium concentration was expected to be lower by one to two orders of magnitude in comparison to the initial concentration can be performed. On the other hand, the stable cesium quantity was kept lower than the sorption capacity of the absorber.

The influence of the added stable cesium concentration on cesium recovery yield was studied for AMP-PAN material. Two additions of cesium to seawater (25 and 4 mg) were equivalent to about 24 and 8 % of the sorption capacity. The capacities were estimated from the sorption isotherm in 0.1 M HCl presented earlier [18]. Cesium recovery yields were around 89 % in both experiments performed with similar seawater volume (100 L) and flow-rate (300 mL min⁻¹). In all later experiments the lower amount of cesium addition was used because it was expected to be less sensitive to flow-rate variability but still provided reproducible results on the ICP-MS analysis of the eluate.

Several replicate cesium separations from spiked acidified seawater by AMP-PAN were then performed and the determined recovery yields are listed in Table 2. For the tested flow-rate range of 240–320 mL min⁻¹ the recovery yield ranged 85–94 %. The variability in the flow-rate was due to partial clogging of the GF/C filter below the absorber bed possibly because of imperfect filtration of some samples. The presented results indicated that the recovery yield decreased with increasing sample flow-rate. The highest separation efficiency was obtained when filter clogging caused gradual decrease of the flow-rate. The effect was also obvious from the cesium breakthrough curve (Fig. 2, experiment D), which suggested that recovery yields could be improved by decreasing seawater flowrates. However, because the average cesium recovery

Table 2 Cesium recovery yields from seawater determined by ICP-MS

	Volume (L)	Average flow-rate (mL min ⁻¹)	Cs added (mg)	Recovery yield (%)
AMP-PAN (acidified seawater)				
A	106	280	25	88.7 ± 0.3
B	93	285	4	88.8 ± 0.4
C	101	319	4	84.9 ± 0.5
D	95	243	4	94.1 ± 0.3
E	98	326	4	86.3 ± 0.5
F	96	298	4	85.7 ± 0.6
KNiFC-PAN (acidified seawater)				
A	97	287	4	92.4 ± 0.4
B	98	288	4	94.4 ± 0.3
C	101	293	4	93.0 ± 0.4
D	102	260	4	91.5 ± 0.5
E	100	293	4	93.2 ± 0.5
KNiFC-PAN (natural seawater)				
A	102	279	4	91.9 ± 0.5
B	100	250	4	92.7 ± 0.5
C	98	276	4	88.3 ± 0.5
D	101	291	4	91.6 ± 0.5
E	100	315	4	90.9 ± 0.5
F	102	470	4	85.6 ± 0.6

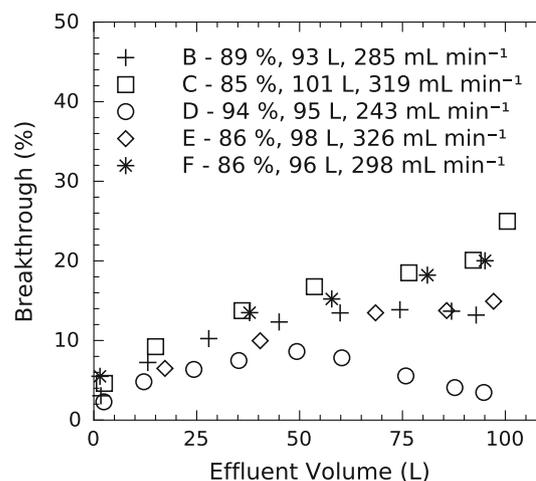


Fig. 2 Dependence of cesium breakthrough on effluent volume for cesium concentration by AMP-PAN from acidified seawater (BV = 25 mL, 4 mg of Cs added). The legend shows total recovery yield, total sample volume, and average flow-rate

yield of 87 % was acceptable the effect of flow-rate on cesium recovery yield was not tested in more details.

KNiFC-PAN was initially tested for cesium separation from acidified seawater under the same conditions as used for the AMP-PAN procedure. The results of several replicate analyses (Table 2) showed that KNiFC-PAN was more efficient in cesium separation from acidified seawater than

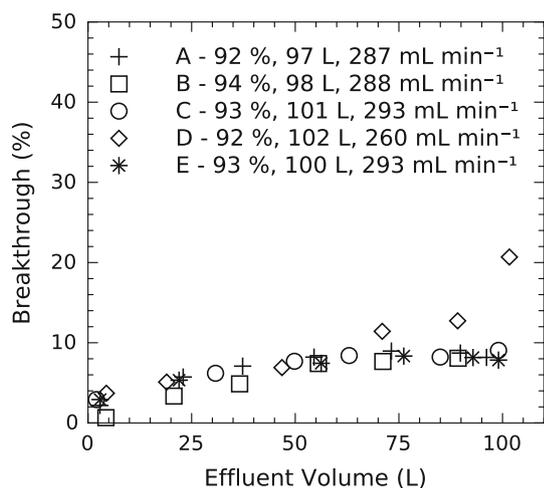


Fig. 3 Dependence of cesium breakthrough on effluent volume for cesium concentration by KNiFC-PAN from acidified seawater (BV = 25 mL, 4 mg of Cs added). The legend shows total recovery yield, total sample volume, and average flow-rate

AMP-PAN. The recovery yield was $93 \pm 1\%$ (average \pm standard deviation) at flowrates of about 280 mL min^{-1} . The pattern of cesium breakthrough (Fig. 3) indicated that cesium could be separated with similar efficiency from even larger seawater samples under the same conditions.

The composite material KNiFC-PAN was also tested for cesium separation from natural seawater without acidification. The advantage of such a procedure would be a possibility for a tandem column separation of cesium on KNiFC-PAN and actinides on material based on hydrated manganese dioxide [1]. The determined recovery yields for non-acidified seawater are presented in Table 2. The average value was $91 \pm 2\%$ (average \pm standard deviation) for flow-rate ranges of $250\text{--}315 \text{ mL min}^{-1}$. A higher extent of GF/C filter clogging was observed during the processing of filtered natural seawater and usually the filter had to be changed during the separation for the treatment of the complete sample volumes. The clogging effect was most pronounced if seawater was filtered about 3 days in advance of cesium separation, possibly because of fine particles formation by coagulation and microbiological growth (which could be disadvantage of non-acidified seawater). If a sieve was used as a bed support, the gradual decrease of flow-rate was not observed. The bed support clogging caused problems with estimation of processed sample volumes; hence breakthrough curves were not determined.

The promising results of cesium separation from natural seawater by KNiFC-PAN motivated a test with higher flow-rate (470 mL min^{-1}) and a sieve as a bed support. The flow-rate allowed the processing of 100 L of seawater in about 3.5 h with a cesium recovery yield of more than 85%. If the processing time was a limiting factor, such high flow-rate could be used.

Detection limits for ^{134}Cs and ^{137}Cs

It was expected that detection limits for both ^{134}Cs and ^{137}Cs would be influenced by the radiopotassium (naturally radioactive ^{40}K) in KNiFC-PAN material. Analysis of the measured gamma spectra of original material and material used for seawater cesium separation showed (after subtraction of the background signal) that potassium content in KNiFC-PAN decreased by the factor of 3.5 after processing 100 L of seawater. This was possibly due to changes of its chemical form by exchange of potassium for sodium ions. A change of absorber color from gray to green tones was noted during the experiment, which may indicate a change in ion content. In case of AMP-PAN, partial retention of potassium from seawater was observed but it was independent on the processed seawater sample volume (25, 50, and 100 L). Possibly, the sites available for potassium uptake in AMP-PAN were occupied early after contact with seawater. Accidentally, the total content of radiopotassium as measured by gamma-spectrometry was almost the same in the counted geometry (due to higher volume of dried KNiFC-PAN than of AMP-PAN). Thus the influence of ^{40}K on calculated minimal detectable activities (MDA) was the same. The MDA values for 100 L samples and 50–70 h of counting were around 0.18 and 0.15 Bq m^{-3} for ^{134}Cs and ^{137}Cs , respectively.

Analysis of cesium radionuclides in the central Pacific Ocean

The material AMP-PAN was used for the analysis of off-shore surface seawater samples collected at station Aloha. The sampling effort is part of the monitoring of Fukushima-derived radiation in the central Pacific Ocean. In the sample from January 23, 2012 the activities were 1.5 ± 0.1 and $<0.2 \text{ Bq m}^{-3}$ (counting time 60 h) for ^{137}Cs and ^{134}Cs , respectively. The coastal sample (O'ahu, February 14, 2012) was analyzed by cesium separation from natural seawater by KNiFC-PAN and its activity concentrations were 1.4 ± 0.1 and $<0.2 \text{ Bq m}^{-3}$ (counting time 70 h) for ^{137}Cs and ^{134}Cs , respectively. These results are in good agreement for roughly expected pre-Fukushima radiocesium levels in this region [12, 13].

Conclusions

The composite materials AMP-PAN and KNiFC-PAN performed very well for the separation of dissolved forms of radiocesium from 100 L of acidified seawater. The column with 25 mL of absorber allowed processing of 100 L samples in less than 6 h (flow-rate 280 mL min^{-1}) with both materials. Higher cesium recovery yields were

found for KNiFC-PAN material (93 ± 1 %) that exhibited similar efficiency for cesium concentration also from non-acidified seawater. More than 85 % of cesium recovery yield was found for cesium concentration from natural seawater by KNiFC-PAN even with higher flow-rate (470 mL min^{-1}). We also found that despite its potassium content, ^{40}K does not cause any background problems during gamma-spectrometric analysis.

The calculated MDA values were below 0.2 Bq m^{-3} for ^{137}Cs and ^{134}Cs for both composite materials tested. The methods developed here therefore allow a sensitive and fast analysis of radiocesium in seawater. The column method is a significant improvement over the historically used precipitation techniques because of the ease to handle large volumes of water for analysis. High recoveries can be achieved with KNiFC-PAN even from non-acidified seawater, which means that the method does not involve the use of any hazardous chemicals and is easy to perform in the field or aboard a scientific cruise. The activity concentrations of ^{137}Cs determined in seawater from Hawaiian Islands region were 1.5 ± 0.1 and $1.4 \pm 0.1 \text{ Bq m}^{-3}$ for the offshore (January 2012) and coastal (February 2012) locality, respectively. The content of ^{134}Cs was below detection limit ($<0.2 \text{ Bq m}^{-3}$) for both samples.

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