Supplementary Information

Novel ^{90}Sr analysis of environmental samples by ion-laser interaction mass spectrometry

Maki Honda,[*ab] Martin Martschini,[a] Oscar Marchhart,[a] Alfred Priller,[a] Peter Steier,[a] Robin Golser,[a] Tetsuya K. Sato,[b] Tsukada Kazuaki,[b] Aya Sakaguchi[c]

[a] University of Vienna, Faculty of Physics, Isotope Physics, Währinger Street 17, Vienna 1090, Austria

[b] Japan Atomic Energy Agency, 2-4 Shirakata, Tokai, Ibaraki 319-1195, Japan

[c] Center for Research in Isotopes and Environmental Dynamics, University of Tsukuba, 1-1-1 Tennodai, Tsukuba, Ibaraki 305-8577, Japan

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Corresponding Author

[*] Maki Honda – honda.maki@jaea.go.jp

Present Addresses

[b] Japan Atomic Energy Agency, 2-4 Shirakata, Tokai, Ibaraki 319-1195, Japan

EXPERIMENTAL SECTION

Reagents. The chemical reagents were 1000 mg L^{-1} Strontium standard solution (ARISTAR® standard for ICP, VWR Chemicals; this Sr standard solution was used for Sr carrier and blank samples), 15 M HNO_3 (extra pure, Carl Roth GmbH), 11 M HCl (extra pure, Carl Roth GmbH), 48% HF (ARISTAR® for trace analysis, VWR Chemicals), 98.7% NaOH (VWR Chemicals), 99.55% NaCl (Merck KGaA), 99.99% PdF_2 (powder, Sigma-Aldrich, and high-purity deionized water (18.2 MΩ/cm) obtained from a water purification system of Milli-Q (Merck KGaA) and Y_2O_3 (99.99%, Rare metallic Co., Ltd.)

Pre-conditioning of Sr resin® (pre-packed column, 50-100 µm, 2 mL, Eichrom Technologies). Ten milliliter of water was passed through, followed by 10 mL of 8 M HNO_3.

Pre-conditioning of anion exchange resin (MCl Gel CA08P, 120 µm, Mitsubishi Chemicals). Pour 2 mL of slurry anion exchange resin into a polypropylene column (6.5 – 8.5 mm i.d. × 58 mm length, Muromachi Chemicals). 10 mL of 1 M NaOH was passed through, followed by 10 mL of 1 M NaCl. This process was repeated three times in total.

The liquid chromatographic technique of Zr using an anion exchange resin column is summarized in J. V. Kratz and Y. Nagame (2014)1. The affinity of Zr for anion exchange resins is expressed as a function of HCl concentration. Zr (IV) in HCl solution forms a chloride complexation (e.g. $[ZrCl_6]^{2-}$), which shows high distribution coefficients (K_d) under solution conditions of high HCl concentration, and K_d decreases steeply with decreasing HCl concentration.

1. J.V. Kratz, Y. Nagame, *Liquid-Phase Chemistry of Superheavy Elements*. In: M. Schädel, D. Shaughnessy. (eds) The Chemistry of Superheavy Elements. Springer, Berlin, Heidelberg, 2014.

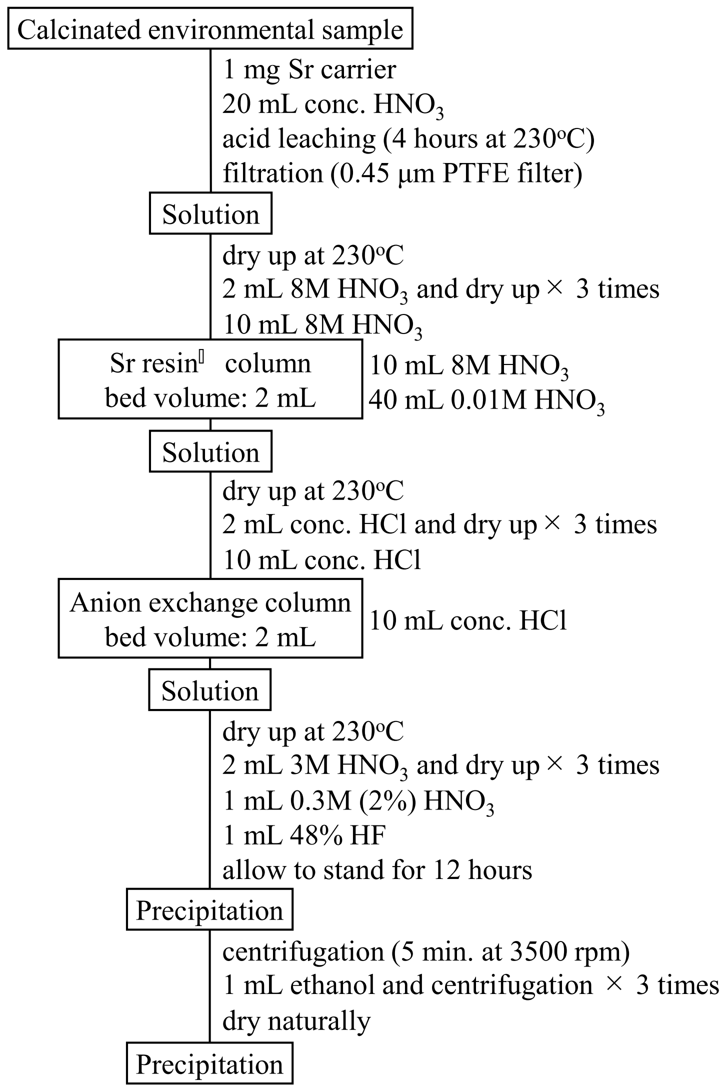

Figure. S1 Separation scheme for ^{90}Sr AMS.

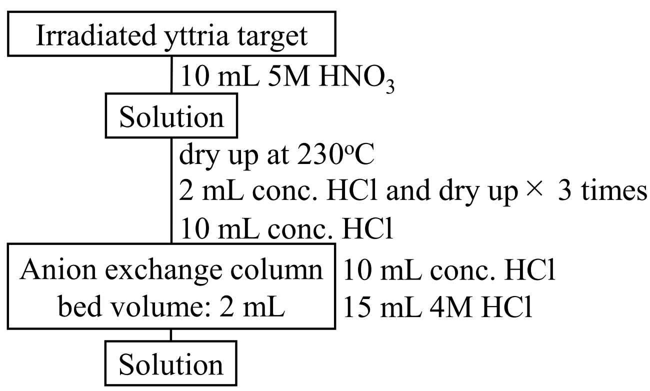

Figure. S2 Separation scheme of Zr using anion exchange resin (CA08P, 120 μm, Mitsubishi Chemicals).

RESULTS AND DISCUSSION

Table S1 Elemental composition of the leachate obtained by acid leaching of IAEA sample

Sample	Treated [g] (Dry)	Unit: g					
		Na [a]	K [a]	Rb [b]	Cs [b]	Mg [a]	Ca [a]
IAEA-447	1.28	(5.37±0.04) $\times 10^{-4}$	(8.95±0.30) $\times 10^{-3}$	(5.13±0.12) $\times 10^{-7}$	(4.76±0.11) $\times 10^{-6}$	(7.87±0.04) $\times 10^{-3}$	(2.20±0.01) $\times 10^{-1}$
IAEA-A-12	1.01	(1.12±0.04) $\times 10^{-3}$	(3.83±0.05) $\times 10^{-4}$	(1.61±0.03) $\times 10^{-5}$	(4.01±0.25) $\times 10^{-9}$	(6.14±0.04) $\times 10^{-3}$	(9.71±0.33) $\times 10^{-1}$
IAEA-TEL-2015-03-S5	1.11	(6.43±0.07) $\times 10^{-4}$	(2.28±0.06) $\times 10^{-3}$	(5.35±0.02) $\times 10^{-5}$	(1.19±0.01) $\times 10^{-6}$	(2.74±0.03) $\times 10^{-3}$	(4.17±0.04) $\times 10^{-3}$
		Sr [b]	Ba [b]	Fe [a]	Zr [b]	Pb [a]	
IAEA-447		(1.18±0.03) $\times 10^{-4}$	(1.02±0.02) $\times 10^{-4}$	(3.21±0.02) $\times 10^{-3}$	(3.66±0.09) $\times 10^{-5}$	(1.72±0.02) $\times 10^{-5}$	
IAEA-A-12		(2.47±0.06) $\times 10^{-4}$	(1.13±0.03) $\times 10^{-4}$	(7.71±0.09) $\times 10^{-5}$	(3.07±0.16) $\times 10^{-8}$	(3.52±0.04) $\times 10^{-5}$	
IAEA-TEL-2015-03-S5		(1.68±0.02) $\times 10^{-4}$	(5.53±0.05) $\times 10^{-4}$	(1.35±0.01) $\times 10^{-2}$	(1.11±0.07) $\times 10^{-4}$	(1.59±0.03) $\times 10^{-5}$	

[a] Elements (Na, K, Mg, Ca, Fe and Pb) were measured by ICP-AES (SPS7800, Hitachi High-Tech Science). The elements were quantified with the standard solutions prepared from XSTC-622 (SPEX).

[b] Elements (^{85}Rb, ^{133}Cs, ^{88}Sr, ^{135}Ba and ^{90}Zr) were measured by ICP-MS (Agilent 7700, He collision and Agilent 8800, MS/MS mode, He collision). Measurements were corrected by an internal standard of ^{115}In, and the elements were quantified with the standard solutions prepared from XSTC-622 (SPEX).

Table S2 ^{90}Sr concentration of IAEA sample and atomic ratio of the leachate obtained by acid leaching

Sample	Reference date	Calculated			Measured by AMS		
		^{90}Sr [b] [Bq/g]	$^{90}Sr/^{88}Sr$ [a] [atoms/atoms]	$^{90}Sr/Sr$ [a] [atoms/atoms]	^{90}Sr [b] [Bq/g]	$^{90}Sr/^{88}Sr$ [a] [atoms/atoms]	$^{90}Sr/Sr$ [a] [atoms/atoms]
IAEA-447	11 Dec. 2019	0.0039±0.0002	$(1.01\pm0.06)\times10^{-12}$	$(8.38\pm0.50)\times10^{-13}$	0.0033±0.0004	$(8.66\pm0.97)\times10^{-13}$	$(7.15\pm0.78)\times10^{-13}$
IAEA-A-12	15 Nov. 2019	$0.0220^{+0.0018}_{-0.0034}$	$(1.02^{+0.10}_{-0.19})\times10^{-12}$	$(9.90^{+0.80}_{-1.54})\times10^{-13}$	0.0226±0.0023	$(1.23\pm0.13)\times10^{-12}$	$(1.02\pm0.10)\times10^{-12}$
IAEA-TEL-2015-03-S5	2 Sep. 2020	0.0316±0.0024	$(6.77\pm0.51)\times10^{-12}$	$(5.59\pm0.42)\times10^{-12}$	0.0278±0.0031	$(5.96\pm0.67)\times10^{-12}$	$(4.92\pm0.55)\times10^{-12}$

[a] The $^{90}Sr/^{88}Sr$ and $^{90}Sr/Sr$ do not represent the atomic ratios of the IAEA samples because natural Sr is not fully extracted from the soil sample by nitric acid leaching. Instead, it is the atomic ratio of the leachate containing ^{90}Sr and Sr (some natural Sr + Sr carrier) leached from the IAEA sample by acid leaching. Nevertheless, these calculated and measured isotopic ratios are given to demonstrate the validity of the AMS measurement of Sr isotopic ratios. $^{90}Sr/Sr$ atomic ratio was calculated from $^{90}Sr/^{88}Sr$ with natural $^{88}Sr/Sr$ as 0.8258.

[b] ^{90}Sr was calculated from measured $^{90}Sr/^{88}Sr$ atomic ratio and total Sr (natural Sr + 1 mg of Sr).