

1 **Supplementary information:**

2 **Table 1S.** Analytical results of $^{239+240}\text{Pu}$ and ^{241}Pu activity, $^{240}\text{Pu}/^{239}\text{Pu}$ and $^{241}\text{Pu}/^{239}\text{Pu}$ atom
3 ratio in ocean sediment reference materials, NIST-4357 and IAEA-368.

Sample	Remarks	$^{239+240}\text{Pu}$ activity (mBq/g)	$^{240}\text{Pu}/^{239}\text{Pu}$ atom ratio	^{241}Pu activity (mBq/g) ^a	$^{241}\text{Pu}/^{239}\text{Pu}$ atom ratio ^a
NIST-4357	Our modified method	9.70±0.13	0.235±0.006	112.3±13.6	0.0135±0.0020
	Certified values	9.2-13.3	–	–	–
	Zhang et al., 2010	9.44±0.19	0.236±0.010	111.5±9.3	0.0132±0.0007
	Hrnecek et al., 2008	10.1±0.5	0.233±0.008	90.1±5.6	–
	Yoshida et al., 2007	9.3±0.16	0.244±0.004	–	–
IAEA-368	Our modified method	33.3±1.6	0.033±0.002	10.53±1.87	0.00019±0.00002
	Certified values	29.0-34.0	–	–	–
	Zhang et al., 2010	31.8±0.6	0.032±0.002	12.01±8.27	0.00026±0.00010
	Donard et al., 2007	30.8±3.5	0.030±0.004	–	–
	Kim et al., 2000	29.9±0.65	0.04±0.01	–	–
	Ohtsuka et al., 2005	30.0±1.6	0.042±0.003	–	–

4 a: Decay corrected to 1 January 2000.

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6 **References**

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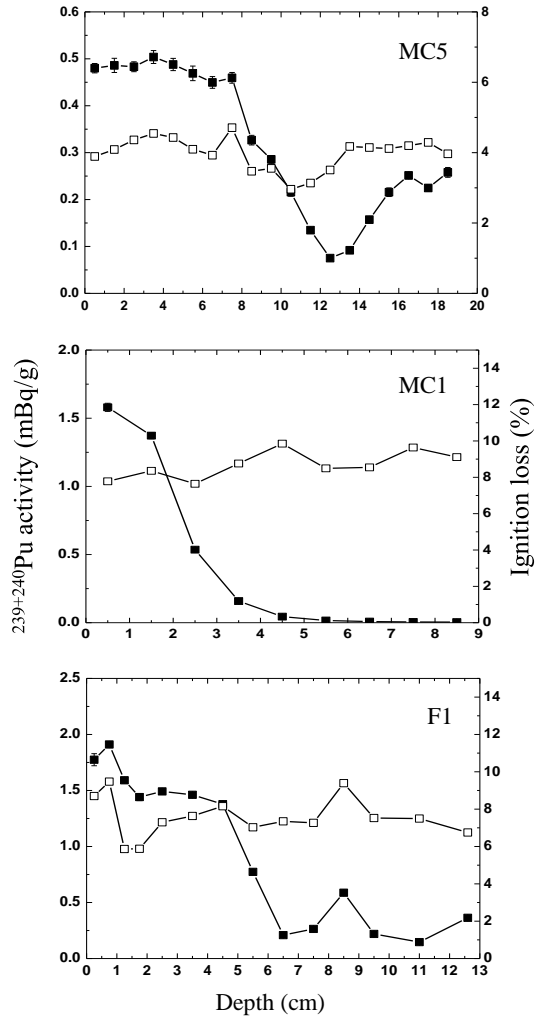
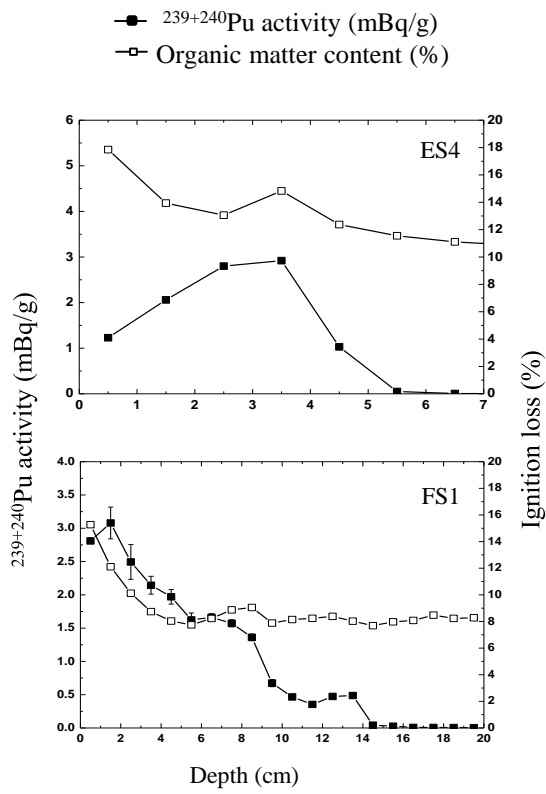
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1 **Table 2S.** Analytical results of Pu isotopes in surface sediments from Japanese estuaries,
 2 investigated in this study.

Estuary	Sample	Location	²³⁹⁺²⁴⁰ Pu activity (mBq/g)	²⁴¹ Pu activity (mBq/g)	²⁴⁰ Pu/ ²³⁹ Pu atom ratio	²⁴¹ Pu/ ²³⁹ Pu atom ratio
Naka River Estuary	2009IBT 01	36°20'6"N, 140°35'36"E	0.117±0.006	ND	0.244±0.024	ND
	2009IBT 02	36°20'6"N, 140°36'18"E	0.198±0.006	ND	0.256±0.015	ND
	2009IBT 03	36°20'6"N, 140°37'0"E	0.263±0.008	ND	0.254±0.015	ND
	2009IBT 04	36°20'6"N, 140°37'36"E	0.351±0.010	ND	0.243±0.018	ND
Kitakami River Estuary	2010MIT 01	38°34'39"N, 141°28'6"E	0.050±0.004	ND	0.205±0.037	ND
	2010MIT 02	38°34'39"N, 141°28'27"E	0.066±0.005	ND	0.214±0.039	ND
	2010MIT 03	38°34'39"N, 141°29'8"E	0.172±0.005	ND	0.208±0.015	ND
Kako River Estuary	2010HYT 01	34°43'23"N, 134°48'1"E	0.129±0.005	ND	0.201±0.018	ND
	2010HYT 02	34°42'55"N, 134°47'41"E	0.090±0.005	ND	0.220±0.024	ND
Sagami River Estuary	2008KAT 01	35°19'12"N, 139°22'6"E	0.010±0.002	ND	ND	ND
	2008KAT 02	35°18'48"N, 139°22'6"E	0.033±0.003	ND	0.261±0.057	ND
	2008KAT 03	35°19'36"N, 139°22'6"E	0.077±0.004	ND	0.258±0.032	ND
Oyodo River Estuary	2009MIT 01	31°53'30"N, 131°28'36"E	0.196±0.006	ND	0.270±0.017	ND
	2009MIT 02	31°53'30"N, 131°29'12"E	0.346±0.010	ND	0.254±0.014	ND
	2009MIT 03	31°53'30"N, 131°29'48"E	0.289±0.006	ND	0.269±0.011	ND
Yoshino River Estuary	2009TOT 01	34°4'21"N, 134°36'21"E	0.049±0.003	ND	0.229±0.023	ND
	2009TOT 02	34°4'15"N, 134°36'39"E	0.060±0.003	ND	0.232±0.021	ND
	2009TOT 03	34°4'45"N, 134°37'18"E	0.091±0.004	ND	0.211±0.024	ND
	2009TOT 04	34°4'15"N, 134°37'57"E	0.114±0.006	ND	0.209±0.025	ND
Oi River Estuary	2009SHT 01	34°46'0"N, 138°18'0"E	0.063±0.003	ND	0.263±0.026	ND
	2009SHT 02	34°46'0"N, 138°18'18"E	0.066±0.004	ND	0.236±0.033	ND
	2009SHT 03	34°46'0"N, 138°18'54"E	0.137±0.005	ND	0.257±0.022	ND

3 ND: Not detected.

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2 **Figure 1S.** The vertical profiles of $^{239+240}\text{Pu}$ activity and ignition loss in the sediment core
 3 samples.

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