

**Supporting Information****Tracking the fate of particle associated Fukushima cesium in the ocean off Japan**

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## Analytical details

### Conversion factors for major flux components

The concentration of organic matter (OM) was estimated as follows:

$$\text{OM (\%)} = \text{POC} / 0.36 \quad (1)$$

where POC is concentration of organic carbon and the constant of 0.36 is the conversion coefficient estimated from the traditional stoichiometric formulation for the composition of marine algae [  $(\text{CH}_2\text{O})_{106}(\text{NH}_3)_{16}(\text{H}_3\text{PO}_4)$  ] based on Redfield et al<sup>1</sup>; Richards<sup>2</sup>; and Honda et al.<sup>3</sup>.

Based on crustal ratios<sup>4</sup>, concentrations of biogenic opal ( $\text{SiO}_2 \cdot 0.4\text{H}_2\text{O}$ ) and lithogenic materials (LM) were estimated with the following equations:

$$\text{Opal (\%)} = (\text{Si} - 3.42\text{Al}) \times 67.2/28 \quad (2)$$

$$\text{LM (\%)} = \text{Al} \times 100/8 \quad (3)$$

$\text{CaCO}_3$  was estimated with PIC as follows:

$$\text{CaCO}_3 (\%) = \text{PIC} \times 100/12 \quad (4)$$

## Radionuclides

Gamma efficiencies were determined from mixed standards spiked with  $^{226}\text{Ra}$ ,  $^{238}\text{U}$ ,  $^{235}\text{U}$ ,  $^{134}\text{Cs}$  and  $^{137}\text{Cs}$  (Eckert & Ziegler Isotope Products, EG-ML) and covering a range of volumes and densities. Activities for  $^{134}\text{Cs}$  were calculated using the average of the 604 and 795 keV energy peaks. The  $^{134}\text{Cs}$  activity was corrected for cascade summing (43–49%). The errors on  $^{134}\text{Cs}$ ,  $^{137}\text{Cs}$  and  $^{210}\text{Pb}$  were propagated from the one sigma counting error plus errors due to calibration. In the case of excess  $^{210}\text{Pb}$ , the error also includes an uncertainty from the difference between total and supported  $^{210}\text{Pb}$  activities and their associated errors.

## Sediment inventories and radionuclide ratios

For radionuclide inventories at F1, sediment cores of 10 and 18 cm length were collected in July 2012 and May 2013, respectively, and sectioned every 0.5 cm for the first 2 cm and in 1-2 cm increments for the rest of the core length. Bulk density was estimated with wet/dry weights.  $^{210}\text{Pb}_{\text{ex}}$ ,  $^{137}\text{Cs}$ , and  $^{134}\text{Cs}$  inventories for each layer were calculated using layer thickness, bulk density, and activity, and then decay-corrected as detailed in the previous section.

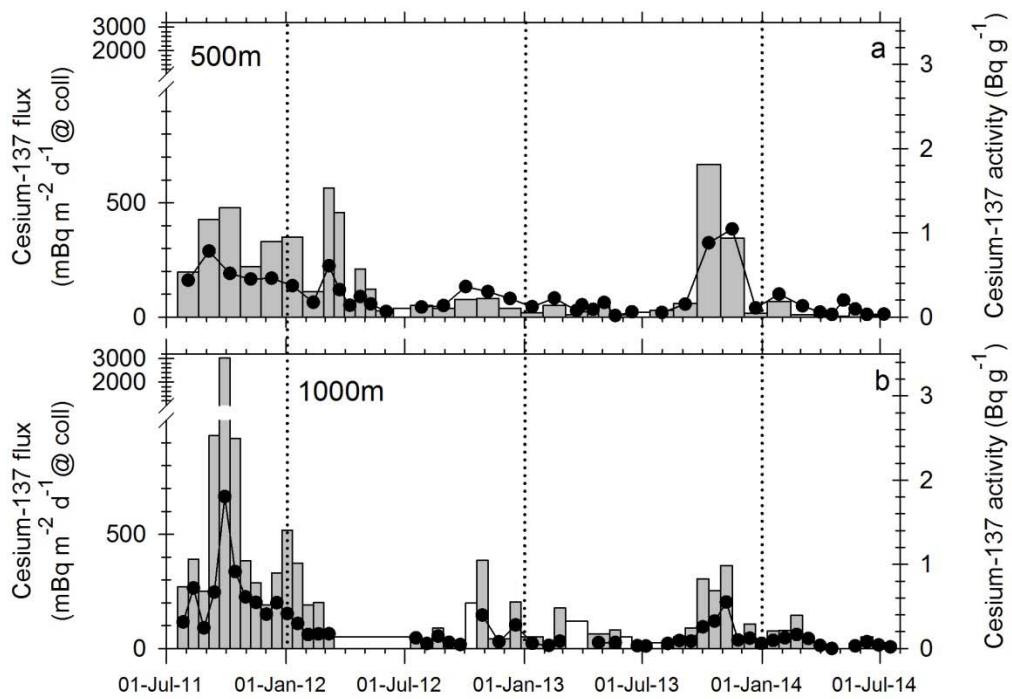


Figure S1. Three year record of  $^{137}\text{Cs}$  flux at time of sampling at a) 500m and b) 1000m shown activity flux (gray bars = measured; white bars are missing samples) in units of  $\text{mBq m}^{-2} \text{d}^{-1}$  (left Y-axis) and specific activity (filled circles and line) in units of  $\text{Bq g}^{-1}$ .

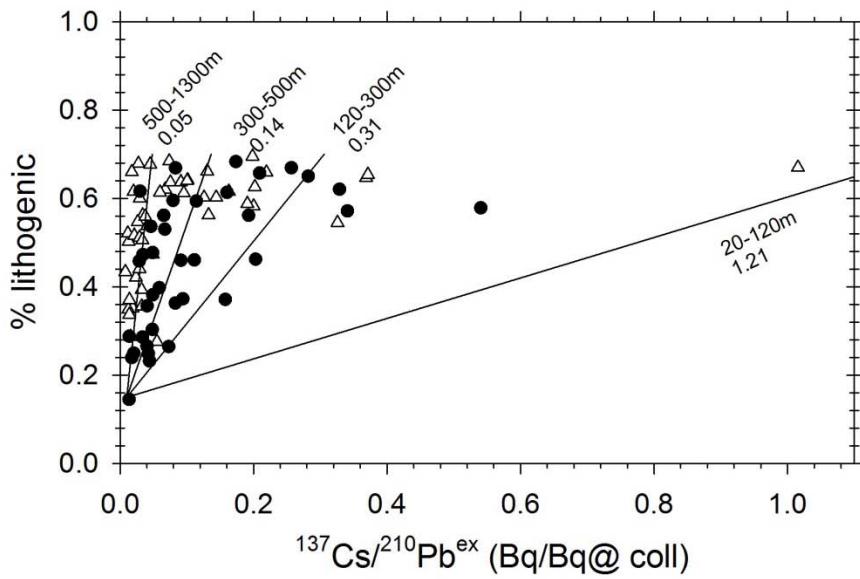


Figure S2. Percent lithogenic fraction vs.  $^{137}\text{Cs}/^{210}\text{Pb}_{\text{ex}}$  activity ratio at time of sampling (@ coll) for 500 m (filled circles) and 1000 m (open triangles) at F1. Lines are drawn to show mixing trends between different end-member  $^{137}\text{Cs}/^{210}\text{Pb}_{\text{ex}}$  ratios using % lithogenic variations to model fraction transported from lateral input sources. See text for details.

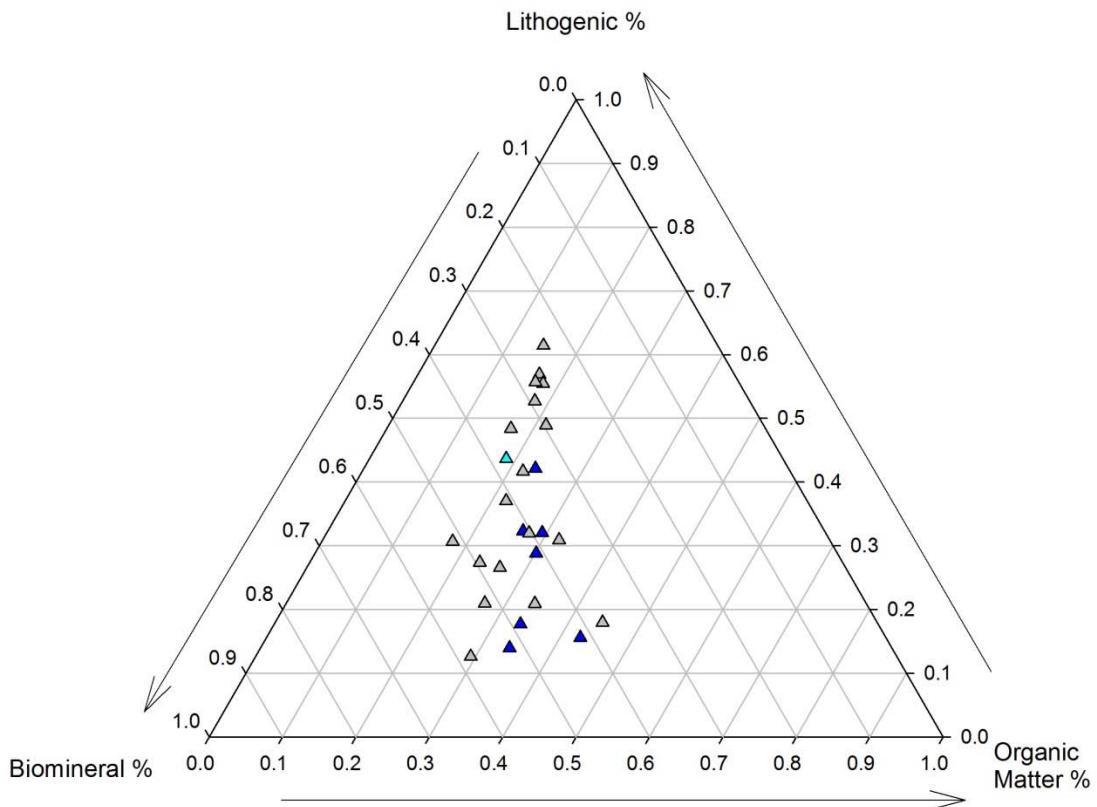


Figure S3. Ternary plot with percent by mass of lithogenic, biomineral and organic matter fractions for FS1 (data from Otosaka et al.<sup>5</sup>). Arrows along the 3 axes point to 100% of a given fraction. Triangles are for a single trap at 870 m. Symbols are color coded by  $^{137}\text{Cs}$  activity using the same scale as for trap F1 (Fig. 5): red > 0.5  $\text{Bq g}^{-1}$ ; cyan 0.3-0.5  $\text{Bq g}^{-1}$ ; blue 0.1-0.3  $\text{Bq g}^{-1}$ ; gray <0.1  $\text{Bq g}^{-1}$ .





**Supplemental Table S2: Comparison of Annual Isotope Fluxes from Sediment Trap F1 and Underlying Sediment Cores**

Collection Method	Location	Sampling Depth (m)	Sampling Period	$^{210}\text{Pb}_{\text{ex}}$ Flux (Bq m $^{-2}$ yr $^{-1}$ )	Calculated $^{210}\text{Pb}_{\text{ex}}$ Inventory (Bq m $^{-2}$ )	$^{137}\text{Cs}$ Flux (Bq m $^{-2}$ yr $^{-1}$ )	$^{134}\text{Cs}$ Flux (Bq m $^{-2}$ yr $^{-1}$ )
Sediment Trap	F1	500	2011-2012	500 $\pm$ 30	16,000 $\pm$ 800	92 $\pm$ 5	98 $\pm$ 4
			2012-2013	210 $\pm$ 10	6,800 $\pm$ 300	15 $\pm$ 0.8	11 $\pm$ 0.5
			2013-2014	270 $\pm$ 10	8,500 $\pm$ 400	44 $\pm$ 2	46 $\pm$ 2
		<b>3-Year Average (<math>^{210}\text{Pb}_{\text{ex}}</math>) or 2-Year Total (Cs)</b>				<b>10,500</b>	<b>110</b>
		1000	2011-2012	600 $\pm$ 30	19,100 $\pm$ 1000	140 $\pm$ 7	143 $\pm$ 5
			2012-2013	720 $\pm$ 40	23,100 $\pm$ 1200	32 $\pm$ 2	30 $\pm$ 2
			2013-2014	580 $\pm$ 30	18,500 $\pm$ 1000	31 $\pm$ 2	33 $\pm$ 1
		<b>3-Year Average (<math>^{210}\text{Pb}_{\text{ex}}</math>) or 2-Year Total (Cs)</b>				<b>20,200</b>	<b>170</b>
						$^{210}\text{Pb}_{\text{ex}}$ Inventory (Bq m $^{-2}$ )	$^{137}\text{Cs}$ Inventory (Bq m $^{-2}$ )
Sediment Cores	Core 5	1300	7/7/2012		15,400 $\pm$ 500	220 $\pm$ 10	190 $\pm$ 10
	Core 6	1260	5/22/2013		16,800 $\pm$ 500	370 $\pm$ 10	280 $\pm$ 20
	<b>Average Total Inventory</b>				<b>16,100 <math>\pm</math> 700</b>	<b>300 <math>\pm</math> 80</b>	<b>240 <math>\pm</math> 50</b>

**Supplemental Table S3: Comparison between Sediment Traps**

Trap	Depth (m)	Mass Flux (mg m <sup>-2</sup> d <sup>-1</sup> )	% CaCO <sub>3</sub>	% Biogenic Silica	% Lithogenic	% Organic Matter	<sup>137</sup> Cs Flux (mBq m <sup>-2</sup> d <sup>-1</sup> )	<sup>134</sup> Cs Flux (mBq m <sup>-2</sup> d <sup>-1</sup> )	<sup>210</sup> Pb <sub>ex</sub> Flux (mBq m <sup>-2</sup> d <sup>-1</sup> )	<sup>137</sup> Cs/ <sup>210</sup> Pb <sub>ex</sub> (Bq Bq <sup>-1</sup> )	Trap Dates
F1	500	482	7.8%	30.1%	51.0%	19.6%	160 ± 8	166 ± 7	1.02 ± 0.05	0.15 ± 0.01	7/19/2011 to 6/15/2013
F1	1000	838	7.3%	24.6%	61.7%	14.9%	340 ± 20	330 ± 10	2.00 ± 0.1	0.17 ± 0.01	8/4/2011 to 7/2/2013
FS1	875	419	12.0%	28.0%	35.0%	26.0%	31 ± 2	32 ± 3	0.98 ± 0.02	0.03 ± 0.002	8/5/2011 to 6/22/2013

NOTES: Percentages and fluxes are averages for the overlapping time period of close to two years when these traps were deployed at the same time.

**References for supplemental text**

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4. Taylor, S. R., Abundance of chemical elements in the continental crust: a new table. *Geochimica et Cosmochimica Acta* **1964**, 28, (8), 1273-1285.
5. Otosaka, S.; Nakanishi, T.; Suzuki, T.; Satoh, Y.; Narita, H., Vertical and Lateral Transport of Particulate Radiocesium off Fukushima. *Environmental Science & Technology* **2014**, 48, (21), 12595-12602.