Supporting Information

Sensitive iodine speciation in seawater by multi-mode size-exclusion chromatography with sector-field ICP-MS

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Table 1S Analytical results of total iodine and dissolved iodine species in seawater collected at Stn. 1 in the North Pacific off Aomori, Japan

| Depth (m) | ΤΙ(μΜ) | IO ₃ (μΜ) | Ι ˙ (μΜ) | TII (μM) | DOI (µM) | lodate (%) | lodide (%) | DOI (%) |
|-----------|-------------------|----------------------|----------|----------|----------|------------|------------|---------|
| 0 | 0.499 ± 0.010 | 0.331 | 0.094 | 0.425 | 0.074 | 66.3 | 18.8 | 14.9 |
| 5 | 0.495 ± 0.009 | 0.328 | 0.100 | 0.428 | 0.066 | 66.4 | 20.2 | 13.4 |
| 10 | 0.500 ± 0.010 | 0.389 | 0.096 | 0.485 | 0.015 | 77.7 | 19.3 | 3.0 |
| 20 | 0.497 ± 0.009 | 0.376 | 0.065 | 0.441 | 0.055 | 75.8 | 13.1 | 11.2 |
| 30 | 0.486 ± 0.006 | 0.385 | 0.064 | 0.449 | 0.037 | 79.2 | 13.1 | 7.7 |
| 40 | 0.502 ± 0.013 | 0.345 | 0.066 | 0.412 | 0.091 | 68.8 | 13.2 | 18.1 |
| 50 | 0.504 ± 0.009 | 0.366 | 0.065 | 0.431 | 0.073 | 72.8 | 12.8 | 14.4 |
| 60 | 0.503 ± 0.006 | 0.370 | 0.062 | 0.432 | 0.071 | 73.6 | 12.3 | 14.2 |
| 75 | 0.483 ± 0.007 | 0.378 | 0.059 | 0.437 | 0.046 | 78.3 | 12.2 | 9.5 |
| 100 | 0.503 ± 0.017 | 0.379 | 0.058 | 0.437 | 0.066 | 75.4 | 11.5 | 13.1 |
| 150 | 0.505 ± 0.008 | 0.361 | 0.057 | 0.418 | 0.087 | 71.5 | 11.3 | 17.2 |
| 200 | 0.514 ± 0.012 | 0.382 | 0.044 | 0.426 | 0.087 | 74.4 | 8.6 | 17.0 |
| 250 | 0.509 ± 0.020 | 0.401 | 0.044 | 0.445 | 0.063 | 78.9 | 8.7 | 12.4 |
| 300 | 0.507 ± 0.007 | 0.392 | 0.042 | 0.433 | 0.074 | 77.3 | 8.2 | 14.5 |
| 500 | 0.527 ± 0.010 | 0.403 | 0.041 | 0.444 | 0.083 | 76.5 | 7.8 | 15.7 |
| 700 | 0.537 ± 0.013 | 0.413 | 0.009 | 0.423 | 0.114 | 77.0 | 1.7 | 21.3 |

TI: total iodine; TII: total inorganic iodine, which equals to the sum of iodate and iodide concentrations. DOI: dissolved organic iodine. Concentrations of iodate and iodide are the mean of duplicate determinations. DOI was calculated from the difference between TI and TII. TI was obtained with triplicate measurements. Concentrations of IO_3^- and I^- were obtained with duplicate measurements.

Table 2S Comparison of inorganic iodate and iodide separation using different chromatographic columns

| Column | Separation mode | Mobile phase | IO ₃ RT(s) | I RT(s) | |
|----------------------|---------------------|---------------------|-----------------------|-------------|--|
| | | 30 mM ammonium | | | |
| IonPac CG 5A | cation-exchange | bicarbonate, | 23 ± 1 | 183 ± 1 | |
| | | 1.1 ml/min | | | |
| | | 30 mM ammonium | | 98 ± 1 | |
| ICS-A2H | anion-exchange | bicarbonate, | 19 ± 1 | | |
| | | 1.1 ml/min | | | |
| | cation-exchange | 30 mM ammonium | | 272 ± 2 | |
| IonPac CG 5A+ICS-A2H | +anion-exchange | bicarbonate, | 36 ± 1 | | |
| | +amon-exchange | 1.1 ml/min | | | |
| | | 30 mM ammonium | | 867 ± 1 | |
| ExcelPak ICS-A23 | anion-exchange | bicarbonate, | 144 ± 1 | | |
| | | 1.1 ml/min | | | |
| | cation concentrator | 30 mM ammonium | | 12 ± 1 | |
| TCC2 | | bicarbonate, | 11 ± 1 | | |
| | | 1.1 ml/min | | | |
| | cation concentrator | 30 mM ammonium | | | |
| TCC2+ICS-A2H | +anion-exchange | bicarbonate, | 25 ± 1 | 105 ± 1 | |
| | +amon-exchange | 1.1 ml/min | | | |
| | multi-mode | 50 mM malonic acid- | | 312 ± 1 | |
| AsahiPak GS-220 7C | size-exclusion | 37.5 mM TMAH, | 157 ± 1 | | |
| | SIZE-CACIUSIOII | pH 6.9; 1.0 ml/min | | | |

^{*}RT: retention time, measured with 3 consecutive analyses.

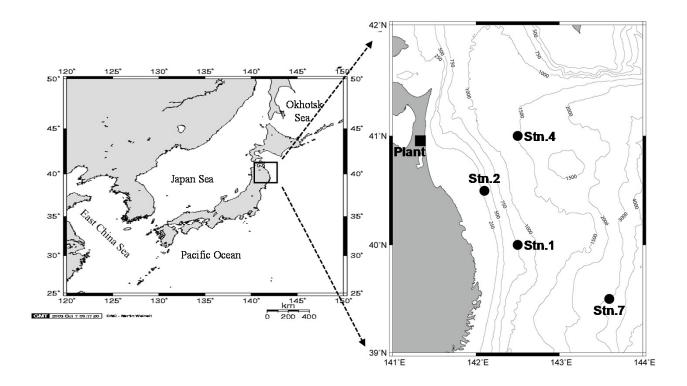


Fig. 1S Map showing the location sites for coastal seawater samples and the location of nuclear fuel reprocessing plant. Seawater samples of Stn.1 were used in this study for iodine speciation, and samples from other stations were used for the analysis of radionuclides, such as ⁹⁹Tc, ¹³⁷Cs and Pu isotopes.

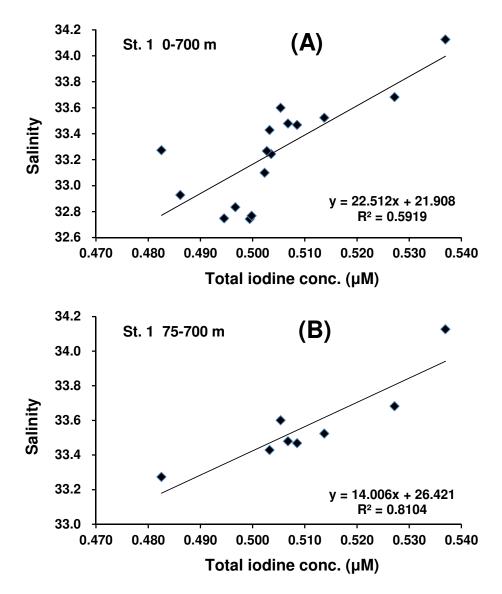


Fig. 2S Correlation between salinity and total iodine concentrations. (A) In the whole water column (0-700 m); (B) For the depths of 75-700 m. Total iodine concentrations were obtained with triplicate measurements using SF-ICP-MS.