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1 **Input of I-129 into the western Pacific Ocean resulting from the Fukushima**
2 **nuclear event.**

3

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11

12 **Abstract:** We present an initial characterization of the input of ¹²⁹I into the Pacific
13 Ocean resulting from the 2011 Fukushima nuclear accident. This characterization is
14 based primarily on ¹²⁹I measurements on samples collected from a research cruise
15 conducted in waters off the eastern coast of Japan in June 2011. These
16 measurements were compared with samples intended to reflect pre-Fukushima
17 background that were collected during a May 2011 transect of the Pacific by a
18 commercial container vessel. In surface waters, we observed peak ¹²⁹I
19 concentrations of ~300 μBq/m³ which represents an elevation of nearly three
20 orders of magnitude compared to pre-Fukushima backgrounds. We coupled our ¹²⁹I
21 results with ¹³⁷Cs measurements from the same cruise and derived an average
22 ¹²⁹I/¹³⁷Cs activity ratio of 0.442x10⁻⁶ for the effluent from Fukushima. Finally, we
23 presented ¹²⁹I depth profiles from five stations from this cruise which form the basis

24 for future studies of ocean transport and mixing process as well as estimations of
25 the total budget of ^{129}I released into the Pacific.

26

27 **Keywords:** Iodine-129, Fukushima, Environmental Radioactivity, Accelerator Mass
28 Spectrometry, Ocean Tracers

29

30 **Introduction**

31 The tsunami resulting from the March 11, 2011 Tōhoku earthquake triggered a
32 catastrophic accident at the Fukushima Daiichi nuclear power plant that caused the
33 release of radioactive elements into the environment [1-5]. Some of these
34 radioactive species (e.g., ^{131}I , ^{134}Cs , ^{137}Cs) are of major concern because they present
35 a hazard associated with radiological dose to surrounding plants, animals, and
36 human populations. Significant effort has been directed to assess the magnitude of
37 the release of radioactivity to the surrounding environment and incorporation into
38 plant and animal food supplies. Such studies are important for estimating the
39 radiological dose exposure to local human populations. The primary input of
40 radioactivity into the Pacific Ocean was a combination of direct discharge from the
41 cooling ponds of the power plant and deposition resulting from atmospheric
42 releases caused by explosions during the nuclear disaster. Determining the relative
43 proportion of these two release routes is important for accurate dose assessments.

44

45 Iodine-129 is another radioactive species that was released by the Fukushima
46 nuclear accident. Owing to an extremely long radioactive half-life (15.7 Ma), ^{129}I

47 presents virtually no radiological hazard so it has understandably been given less
48 focus thus far. It is nonetheless valuable to assess the environmental release of ^{129}I
49 for a number of reasons.

50

51 First, ^{129}I is a powerful tool for studying ocean transport and mixing processes [6-
52 12]. In addition to the long radioactive half-life of ^{129}I , the long residence time
53 (~ 245 ka) and relatively low bioavailability of iodine enable ^{129}I to behave as a
54 nearly conservative tracer in the ocean. The utility of ^{129}I has been extensively
55 demonstrated in many studies in which releases from the Sellafield and La Hague
56 reprocessing plants were traced into the surrounding seas (e.g., Norwegian, Baltic,
57 etc.) and further the North Atlantic Ocean. The Fukushima releases represent a
58 similar, albeit less intense and more discrete, point source for tracing water mass
59 movement in the Western Pacific Ocean.

60

61 Another valuable reason for investigating the release of ^{129}I from Fukushima is its
62 potential for differentiating between atmospherically deposited and directly
63 discharged radioactivity in the Pacific Ocean. It has been demonstrated that
64 atmospherically-released radioiodine and radiocesium from the 1986 Chernobyl
65 nuclear accident were chemically fractionated because of differences in volatility
66 [13, 14]. A similar fractionation can be expected in the atmospheric release from
67 Fukushima, while the direct discharge should more closely reflect the relative ratio
68 of radioiodine and radiocesium in the damaged fuel rods. If the degree of
69 fractionation is sufficiently high, then it should be possible to assess the relative

70 contributions of atmospherically deposited and directly discharged radioactivity
71 from Fukushima in the Pacific Ocean. Iodine-129 is the only radioactive isotope of
72 iodine with a sufficiently long half-life to allow this type of investigation.

73

74 The purpose of the present work is to present an initial assessment of the input of
75 Fukushima-derived ^{129}I into the Western Pacific Ocean. This assessment is
76 primarily based on measurements of water samples collected during a research
77 cruise conducted off the eastern coast of Japan during June 2011. In order to put
78 these measurements into proper context, we establish a pre-Fukushima background
79 of ^{129}I through measurements of water samples collected from a container ship that
80 crossed the Pacific Ocean in May 2011. We also present an initial estimate of the
81 $^{129}\text{I}/^{137}\text{Cs}$ activity ratio from the direct discharge of the Fukushima effluent and
82 compare this to values from Chernobyl.

83

84 **Experimental**

85 To establish a pre-Fukushima background of ^{129}I in the Pacific Ocean, we collected
86 water samples from a container ship (OOCL Tokyo) during a May 2011 crossing of
87 the Pacific from the Port of Hong Kong (China) to the Port of Long Beach (USA). The
88 exact coordinates and time of collections are listed in Table 1. In brief, the OOCL
89 Tokyo's course took her from the South China Sea, through the Taiwan Strait into
90 the East China Sea, then into the Philippine Sea just south of Kyushu, where she
91 began a great circle route to a latitude of $\sim 43^\circ\text{N}$. Samples were collected using a
92 continuous flow surface seawater line.

93

94 To assess the input of Fukushima-derived ^{129}I into the Pacific Ocean, water samples
95 were collected during a research cruise conducted on board the R/V Ka'imikai-o-
96 Kanaloa (KOK) from June 3 to June 17, 2011. The cruise plan has been previously
97 published [15], but in short, consisted of 32 major sampling stations ranging from
98 50 to 650 m from the Fukushima Daiichi nuclear power plant. Stations were
99 selected to focus on the area north of the Kuroshio current, but a few stations were
100 located within the current. A standard Rosette water sampler fitted with Niskin
101 bottles and conductivity, temperature, and depth (CTD) sensors was deployed at
102 each station. Water samples were collected at discrete depths ranging from 1000 m
103 up to near the sea surface (5 to 20 m).

104

105 Samples from both the OOCL Tokyo and the KOK were collected in a similar manner.
106 Prior to collection, the supply tap (from either the continuous flow surface sampler
107 or a Niskin bottle) was opened and allowed to flush for a period of time to ensure
108 samples were not contaminated with exterior residual water. Bottles (0.5-L, HDPE,
109 acid-cleaned with 2% nitric acid) were then filled from the bottom and allowed to
110 flush for 2-3 times their volume. Bottles were sealed, their caps taped, and stored in
111 the dark until sampled for ^{129}I analysis.

112

113 Samples were prepared for ^{129}I analysis using an adapted version of a commonly-
114 used solvent extraction procedure [16, 17, 18]. Briefly, 0.5 mg of iodine carrier
115 (Woodward Iodine Corporation) with a very low ^{129}I content ($^{129}\text{I}/^{127}\text{I} \sim 2 \times 10^{-14}$)

116 was added to a 250-mL aliquot of each sample. Dissolved inorganic iodine was
117 reduced to iodide by addition of sodium sulfite and hydroxylamine hydrochloride
118 and then oxidized to molecular iodine by addition of nitric acid and sodium nitrite.
119 The molecular iodine was extracted into chloroform and then back-extracted into a
120 reducing solution of aqueous sodium sulfite and potassium hydroxide. Silver iodide
121 was precipitated by addition of silver nitrate and the precipitate was rinsed and
122 dried before being mechanically mixed with niobium powder and loaded into a
123 target holder for analysis. The $^{129}\text{I}/^{127}\text{I}$ isotope ratios of the prepared targets were
124 measured by accelerator mass spectrometry (AMS) at the Lawrence Livermore
125 National Laboratory Center for AMS [19].

126

127 **Results & Discussion**

128 The results of $^{129}\text{I}/^{127}\text{I}$ measurement of the surface water samples collected during
129 the May 2011 crossing of the Pacific by the OOCL Tokyo are presented in Table 1. A
130 consistently increasing trend from west to east is observed, although the total range
131 in measured values is only a factor of two. All of the measured values fall within the
132 lower end of the range given for shallow seawater by Snyder [20]. The reason for
133 the increasing trend is not completely understood. As these samples were collected
134 two months following the Fukushima accident, there is likely some component of
135 ^{129}I resulting from atmospheric deposition. However, if this were the dominant
136 recent input of ^{129}I , then a peak would be expected closer to the point source.
137 Indeed, a local maxima is observed at Station 18, which is geographically the closest
138 station to Fukushima. The fact that the highest values of $^{129}\text{I}/^{127}\text{I}$ were observed at

139 the greatest distance (Stations 39, 42, 45) from Fukushima implies that there is
140 another input of ^{129}I into the Pacific.

141

142 The results of ^{129}I measurements of surface samples from the June 2011 KOK cruise
143 are presented in Figure 1. For comparison to corresponding ^{137}Cs measurements
144 from [15], the ^{129}I data in this figure are expressed in units of $\mu\text{Bq}/\text{m}^3$ by converting
145 the measured $^{129}\text{I}/^{127}\text{I}$ ratio using the literature value of iodine concentration in
146 seawater of 0.47 nM [21]. The ^{129}I data show the expected general trend of
147 increasing concentration with proximity to the Fukushima nuclear facility. As
148 discussed in [15], the confluence of the Kuroshio and Oyashiro currents create an
149 eddy that effectively constrained a bulk of the ^{129}I from migrating south into the
150 Kuroshiro. There is general agreement between the measured ^{129}I concentrations
151 and the ^{137}Cs concentrations presented in [15], however the $^{129}\text{I}/^{137}\text{Cs}$ ratio is not
152 constant for all stations.

153

154 Figure 2 shows a plot of the $^{129}\text{I}/^{137}\text{Cs}$ activity ratio as a function of the total ^{137}Cs
155 concentration measured in the surface waters from the June 2011 KOK cruise. At
156 low ^{137}Cs concentrations, the $^{129}\text{I}/^{137}\text{Cs}$ ratio asymptotically approaches a value
157 reflecting pre-Fukushima value of fallout in the Pacific. As the ^{137}Cs concentration
158 increases, this ratio approaches a value that reflects the average ratio in Fukushima
159 effluent. Based on this observation, the data were fit to a two end-member mixing
160 equation:

$$\left[\frac{^{129}\text{I}}{^{137}\text{Cs}} \right]_{\text{Meas}} = \left(\frac{[^{137}\text{Cs}]_{\text{Meas}}}{[^{137}\text{Cs}]_{\text{Meas}} + [^{137}\text{Cs}]_{\text{Bkg}}} \right) \cdot \left[\frac{^{129}\text{I}}{^{137}\text{Cs}} \right]_{\text{Fuk}} + \left(\frac{[^{137}\text{Cs}]_{\text{Bkg}}}{[^{137}\text{Cs}]_{\text{Meas}} + [^{137}\text{Cs}]_{\text{Bkg}}} \right) \cdot \left[\frac{^{129}\text{I}}{^{137}\text{Cs}} \right]_{\text{Bkg}} \quad (1)$$

161 where: $\left[\frac{^{129}\text{I}}{^{137}\text{Cs}} \right]$ represents the $^{129}\text{I}/^{137}\text{Cs}$ activity ratio, $[^{137}\text{Cs}]$ represents the ^{137}Cs
162 concentration, and the subscripts: Meas, Fuk, Bkg refer to the values measured from
163 the June 2011 KOK cruise, the derived average value in Fukushima effluent, and the
164 derived average value in pre-Fukushima Pacific Ocean seawater respectively. From
165 this fit we derive an average $^{129}\text{I}/^{137}\text{Cs}$ activity ratio of 0.443×10^{-6} in Fukushima
166 effluent and a ratio of 58.9×10^{-6} for pre-Fukushima Pacific Ocean water.

167

168 It is instructive to compare the $^{129}\text{I}/^{137}\text{Cs}$ activity ratio derived from this data set to
169 the comparable ratio from Chernobyl. For example, [13] reports a value of
170 0.275×10^{-6} for the $^{129}\text{I}/^{137}\text{Cs}$ activity ratio calculated in the active zone of the 4th
171 block of Chernobyl nuclear power plant at the time of that accident. This value is
172 38% lower than our derived value for Fukushima effluent. For further comparison,
173 we examined gamma spectroscopy measurements of ^{131}I and ^{137}Cs in near-shore
174 seawater samples as reported by the Tokyo Electric Power Company (TEPCO)
175 beginning on March 23, 2011 [22]. Figure 3 shows the measured $^{131}\text{I}/^{137}\text{Cs}$ activity
176 ratio as a function of days after the Tōhoku earthquake. Fitting these data to an
177 exponential decay function yields an initial (i.e., at the time of the earthquake) ratio
178 of 18.2. The corresponding value from [13] is 10.4 which is 43% lower than our
179 derived value for Fukushima effluent. This is consistent with the relative difference
180 between the $^{129}\text{I}/^{137}\text{Cs}$ ratios discussed above.

181

182 The results of ^{129}I measurements in depth profile samples collected from five
183 stations during the June 2011 KOK cruise are shown in Figure 4 along with
184 corresponding basic hydrographic data (salinity and temperature). As expected, the
185 three near-field stations (21, 27, 29) show the highest concentration of ^{129}I
186 throughout the depth profile. Stations 21 and 27 show a peak in the ^{129}I
187 concentration at the surface with an exponential decrease with depth. In contrast,
188 stations 14 and 29 show a peak in ^{129}I concentration below the surface, but still
189 within the mixed layer. The ^{129}I concentration within the mixed layer of station 5
190 seems to be fairly well homogenized. When augmented with depth profile data from
191 the rest of the sampling stations, these data will form the basis for estimating the
192 total release of ^{129}I activity from Fukushima as well lay the foundation for
193 interpreting hydrodynamic transport and mixing processes.

194

195 **Conclusion**

196 Our work provides a characterization of the ^{129}I that was released into the Pacific
197 Ocean resulting from the Fukushima nuclear accident. Observed concentrations of
198 ^{129}I in near-shore surface waters were nearly three order of magnitude above the
199 pre-Fukushima background based on our measurements. This input signal should
200 be sufficiently high to enable future studies that exploit ^{129}I as a tracer for ocean
201 movement and mixing processes. Furthermore, we provide an initial estimate of the
202 average $^{129}\text{I}/^{137}\text{Cs}$ activity ratio in Fukushima effluent. Deviations from this average
203 ratio were observed that may reflect inputs of both atmospherically-deposited and

204 directly-discharged radioactivity in the Pacific Ocean. Further studies of the
205 $^{129}\text{I}/^{137}\text{Cs}$ ratio in a broader range of samples are required to estimate the relative
206 contribution of these two inputs. Finally, the observed depth profiles of ^{129}I are
207 mainly consistent with ^{137}Cs , however some anomalies were observed that warrant
208 additional investigation.

209

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218

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280

281 **Tables**

282 Table 1. $^{129}\text{I}/^{127}\text{I}$ ratios measured in surface water samples collected during the May
 283 2011 crossing of the Pacific Ocean by the OOCL Tokyo. Values in parentheses
 284 represent 1-sigma uncertainty associated with the analytical measurements.
 285

Station	Coordinates	UTC Date/Time	$^{129}\text{I}/^{127}\text{I}$ ($\times 10^{-11}$)
1	22°22' N, 115°57' E	5/16/11 11:56	2.68(0.11)
3	24°19' N, 118°58' E	5/16/11 22:15	2.50(0.09)
6	26°25' N, 122°45' E	5/17/11 10:00	2.61(0.10)
9	30°35' N, 130°07' E	5/18/11 07:37	2.67(0.10)
12	32°49' N, 137°31' E	5/19/11 02:44	2.87(0.10)
15	35°30' N, 143°54' E	5/19/11 19:17	2.84(0.11)
18	38°24' N, 150°27' E	5/20/11 11:55	3.96(0.14)
21	41°25' N, 159°12' E	5/21/11 09:09	2.16(0.09)
24	43°19' N, 170°11' E	5/22/11 09:32	2.97(0.11)
27	43°16' N, 178°47' W	5/23/11 08:41	3.14(0.12)
30	43°08' N, 169°03' W	5/24/11 07:45	2.92(0.10)
33	42°59' N, 159°45' W	5/25/11 08:03	3.44(0.13)
36	42°10' N, 150°41' W	5/26/11 08:20	3.48(0.12)
39	40°45' N, 142°25' W	5/27/11 06:55	4.31(0.15)
42	38°39' N, 134°08' W	5/28/11 07:34	4.86(0.17)
45	36°18' N, 127°10' W	5/29/11 05:35	5.16(0.17)

286

287 **Figure Captions**

288 Figure 1. Measured concentration of ^{129}I in surface waters off the eastern coast of
289 Japan. Samples were collected during June of 2011 from the R/V Ka`imikai-o-
290 Kanaloa.

291

292 Figure 2. Plot showing the $^{129}\text{I}/^{137}\text{Cs}$ activity ratio as a function of ^{137}Cs
293 concentration measured in surface water samples collected from the June 2011
294 cruise of the R/V Ka`imikai-o-Kanaloa. The data were fitted to a two end-member
295 mixing function given in Equation 1.

296

297 Figure 3. Plot of the measured $^{131}\text{I}/^{137}\text{Cs}$ activity ratio measured at a sampling
298 station located approximately 30 m north of the discharge canal of units 5 and 6 of
299 the Fukushima Daiichi nuclear power plant. Data were taken from press releases
300 issues by TEPCO [22]. Line shows a fit of the data to an exponential function
301 corresponding to radioactive decay extrapolated back to the date of the Tōhoku
302 earthquake.

303

304 Figure 4. Plot of salinity, temperature, and ^{129}I concentration as a function of depth
305 at five stations from the June 2011 cruise of the R/V Ka`imikai-o-Kanaloa: a) Station
306 5 [36°N, 147°E]; b) Station 14 [36°N, 144°E]; c) Station 21 [37.5°N, 142.5°E]; d)
307 Station 27 [36.5°N, 141.5°E]; Station 29 [36.5°N, 142°E].

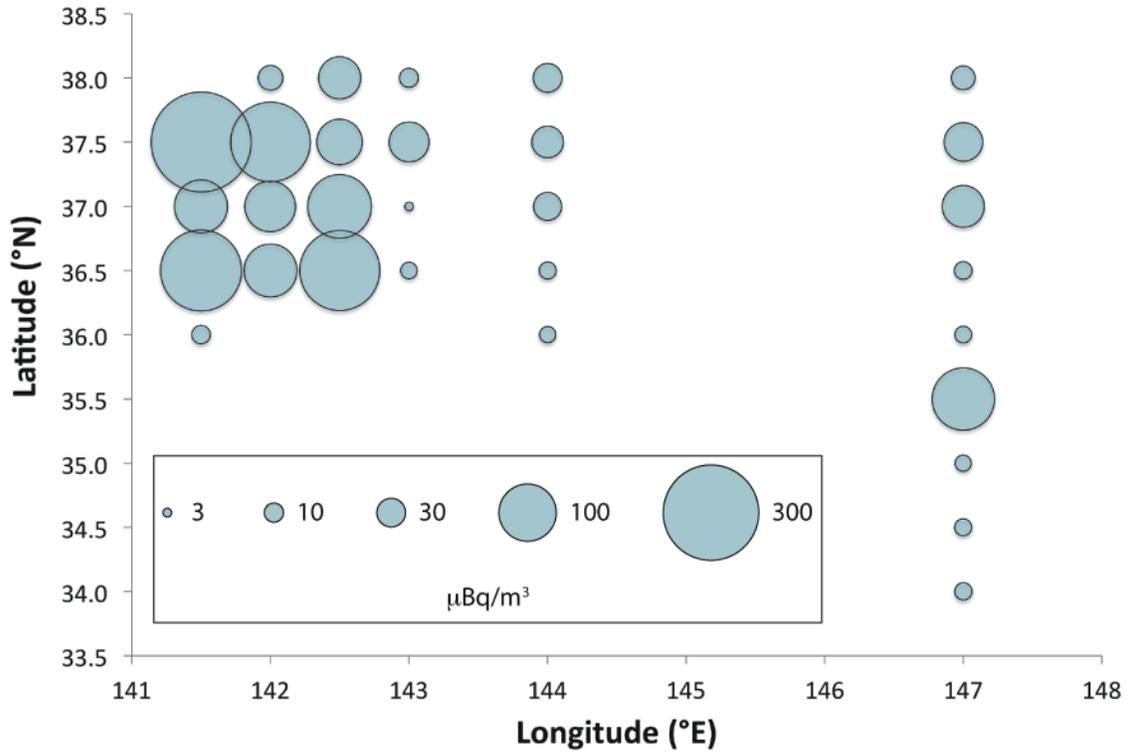
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310 **Figures**

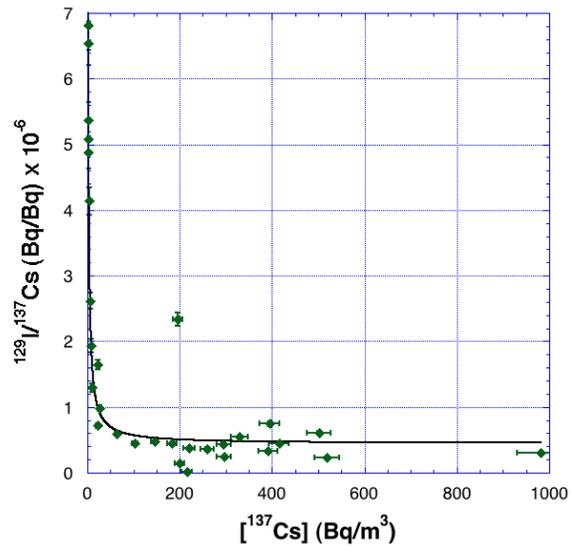
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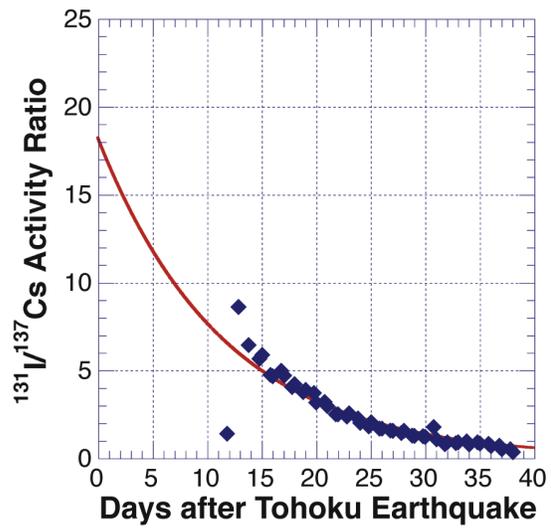
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315 Figure 2
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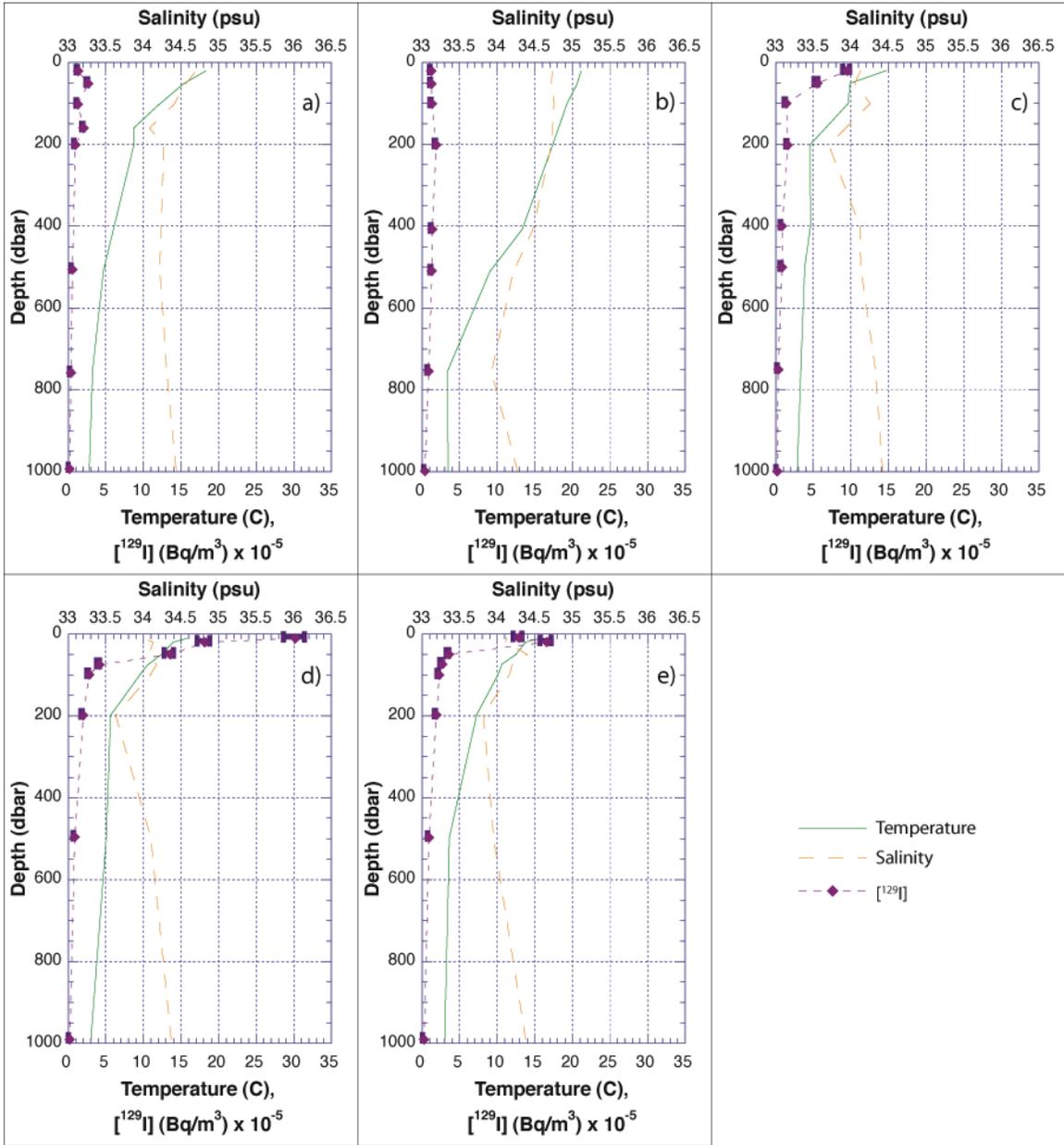
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319 Figure 3
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321
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323 Figure 4
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