

1           **Contribution Ratios of Natural Radionuclides to**  
2           **Ambient Dose Rate in Air After the Fukushima Daiichi**  
3           **Nuclear Power Plant Accident**

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5           Kazumasa Inoue <sup>1</sup>, Hiroshi Tsuruoka <sup>1</sup>, Tan Le Van <sup>1</sup> and Masahiro Fukushi <sup>1,\*</sup>

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7           <sup>1</sup> *Graduate School of Human Health Sciences, Tokyo Metropolitan University,*

8

*7-2-10 Higashiogu, Arakawa-ku, Tokyo 116-8551, Japan*

9           **Abstract**

10          It is important that the contribution ratio of natural radioactivity to ambient dose rate in  
11          air is clarified after the accident at the Fukushima Daiichi Nuclear Power Plant. In this  
12          study, ambient dose rates in air were observed at 34 places in eastern Japan and the  
13          contribution ratios were clarified. The mean contribution ratio of the natural  
14          radionuclides was 71% (range: 0 – 100%). In most places, the natural radionuclides made  
15          a larger contribution to the ambient dose rate in air. (80/100 words)

16          **Keywords**

17          Fukushima Daiichi Nuclear Power Plant; Eastern Japan; Ambient dose rate in air; Natural  
18          radionuclides; Artificial radionuclides; Mobile survey

19

20        **Introduction**

21    Large amounts of artificial radionuclides such as  $^{131}\text{I}$ ,  $^{134}\text{Cs}$  and  $^{137}\text{Cs}$  were released to the  
22    environment in the March 2011 accident at the Fukushima Daiichi Nuclear Power Plant  
23    (F1-NPP) of the Tokyo Electric Power Company (TEPCO) [1]. UNSCEAR has estimated  
24    that 100-500 PBq of  $^{131}\text{I}$  and 6-20 PBq of  $^{137}\text{Cs}$  were released from the reactor buildings  
25    in March 2011 [2]. These amounts from F1-NPP were about 10% and 20% for  $^{131}\text{I}$  and  
26     $^{137}\text{Cs}$ , respectively, of those estimated for the Chernobyl accident. While large amounts  
27    of the released artificial radionuclides were mainly deposited in areas northeast of F1-  
28    NPP such as Futaba Town, Okuma Town, Iidate Village, and Namie Town [3], these  
29    radionuclides were diffused and deposited all over eastern Japan depending on the wind  
30    direction and precipitation field [4]. Regularly updated ambient dose rates in air obtained  
31    from monitoring posts located in each prefecture have been publicized on the website of  
32    the Nuclear Regulation Authority since 2011 [5]. The distribution of ambient dose rates  
33    in air within 80 km from the F1-NPP has also been publicized by researchers from the  
34    Japan Atomic Energy Agency [6].

35        While many studies about ambient dose rates in air near the F1-NPP (i.e., the  
36    Pacific Ocean side of Honshu) have been reported, there are only a few reports, including  
37    simulation studies, about the rates on the Sea of Japan side [7, 8] (Fig.1). Additionally,  
38    the contribution ratios of artificial radionuclides ( $^{134}\text{Cs}+^{137}\text{Cs}$ ) and natural radionuclides  
39    ( $^{40}\text{K}$ ,  $^{238}\text{U}$  series and  $^{232}\text{Th}$  series) to ambient dose rate in air have not been clearly  
40    explained. In fact, the ambient dose rate in eastern Japan is not uniform; there are areas  
41    on both the Pacific and Sea of Japan sides with higher ambient dose rates depending on  
42    the basement geology [9, 10]. In the present study, a car-borne survey and fixed-point  
43    observations were carried out on both the Pacific and Sea of Japan sides and the  
44    contribution ratios of natural radionuclides were analyzed.

45

46        **Materials and Methods**

47    Ambient dose rates in air ( $\text{nGy h}^{-1}$ ) were measured during September 10-15, 2014 and  
48    November 1-3, 2014 in eastern Japan on both Pacific and Sea of Japan coasts and in the  
49    interior (Fig.1). A car-borne survey technique is a common method for fast assessment of  
50    dose rate in a large area. The present car-borne survey was carried out using a 3-in  $\times$  3-in  
51    NaI(Tl) scintillation spectrometer with a global positioning system (EMF Japan Co.,  
52    Osaka, Japan). Latitude and longitude at each measurement point were measured at the  
53    same time as the count rates were recorded. Measurements were performed every 30 s.  
54    The survey route mainly selected expressways and major highways such as national  
55    routes. The survey route was 3,412 km long. Shielding by the car body was estimated by  
56    making measurements inside and outside the car at the 34 numbered locations (Figs. 1a  
57    and 1b). Counting time inside and outside the car was set to 2 min. For the outside  
58    measurements, a scintillation spectrometer was positioned 1 m above the ground surface.  
59    A preliminary experiment was performed and the shielding factor of 1.53 was found from  
60    the correlation between count rates inside and outside the car. This factor was used for  
61    correction of the measured value so as to better represent the outside count rate.  
62    Additionally, the pulse height distributions were obtained outside the car from a total  
63    recording period of 10 min at the 34 locations. Obtained data were unfolded using a  $22 \times$   
64     $22$  response matrix [11] and the ambient dose rates in air were calculated. The dose  
65    conversion factor ( $\text{nGy h}^{-1}/\text{cps}$ ) was found to be 0.093 by correlation between total count  
66    rate and ambient dose rate in air.

67        All the obtained data from the car-borne survey were plotted on distribution maps  
68    using generic mapping tools (University of Hawaii, HI, USA). For more detailed analysis,  
69    all the obtained pulse height distributions outside the car were analyzed using a response  
70    matrix [11] as described above and the separated ambient dose rates from the natural  
71    radionuclides ( $^{40}\text{K}$ ,  $^{238}\text{U}$  series and  $^{232}\text{Th}$  series) and the artificial radionuclides ( $^{134}\text{Cs}$  and  
72     $^{137}\text{Cs}$ ).

73

74        **Results and Discussion**

75        Fig. 2a shows a distribution map of the ambient dose rates in air obtained by the car-  
76 borne survey in eastern Japan ( $n = 5,674$ ). Higher ambient dose rates in air were observed  
77 in areas west and southwest of the F1-NPP; these data were in agreement with previously  
78 reported values [6]. The mean and range of ambient dose rates in air in this survey were  
79  $129 \text{ nGy h}^{-1}$  and  $11\text{-}7489 \text{ nGy h}^{-1}$ , respectively. On the Sea of Japan side, lower ambient  
80 dose rates in air were observed compared to those of the Pacific Ocean side. Especially,  
81 the lowest values were observed in Akita Prefecture; the mean and range of absorbed  
82 dose rates were  $24 \text{ nGy h}^{-1}$  and  $11 - 59 \text{ nGy h}^{-1}$ , respectively.

83        The distribution maps with enlarged scales of ambient dose rate in air are shown  
84 in Figs. 2b and 2c which exhibit the distribution of very high observed dose rates near the  
85 F1-NPP. The highest ambient dose rate in air ( $7489 \text{ nGy h}^{-1}$ ) was observed in Okuma  
86 Town at a location 2.3 km west of the F1-NPP ( $37.42342^\circ\text{N}$ ,  $141.00407^\circ\text{E}$  in Fig. 2c).  
87 This point was located on National Route (NR) 6 that had been in a regulated access zone  
88 (NR 6 distance within the zone: 14 km) until September 15, 2014. The mean and range of  
89 ambient dose rates in air ( $n = 59$ ; measured on November 1, 2014) at this point were  $2812$   
90  $\text{ nGy h}^{-1}$  and  $359\text{-}7489 \text{ nGy h}^{-1}$ , respectively. Here, if a car with the windows closed  
91 passed along NR 6 at a speed of  $40 \text{ km h}^{-1}$ , the effective dose to everyone in the car was  
92 calculated as  $0.45 \text{ }\mu\text{Sv}$ . The dose conversion factor used was  $0.748 \text{ Sv Gy}^{-1}$  [12] for  
93 conversion to the effective dose. According to the UNSCEAR [13], the dose conversion  
94 factor is defined to be  $0.7 \text{ Sv Gy}^{-1}$ . However, this factor is available for 0.5 MeV of  
95 gamma ray, it cannot use to the energy of gamma ray in the environment (tens of keV –  
96  $2.614 \text{ MeV}$ ). Thus, Moriuchi et al. [12] reported factor was used in this study.

97        According to the Team in Charge of Assisting the Lives of Disaster Victims,  
98 Government of Japan [14], the mean and range of ambient dose rates in air on this same  
99 NR 6 obtained with a NaI(Tl) survey meter between July 2 and August 12, 2014 were  
100  $4679 \text{ nGy h}^{-1}$  and  $414 - 19652 \text{ nGy h}^{-1}$ , respectively. The effective dose to a person  
101 traveling this route by car at a speed of  $40 \text{ km h}^{-1}$  was estimated from the concentration of

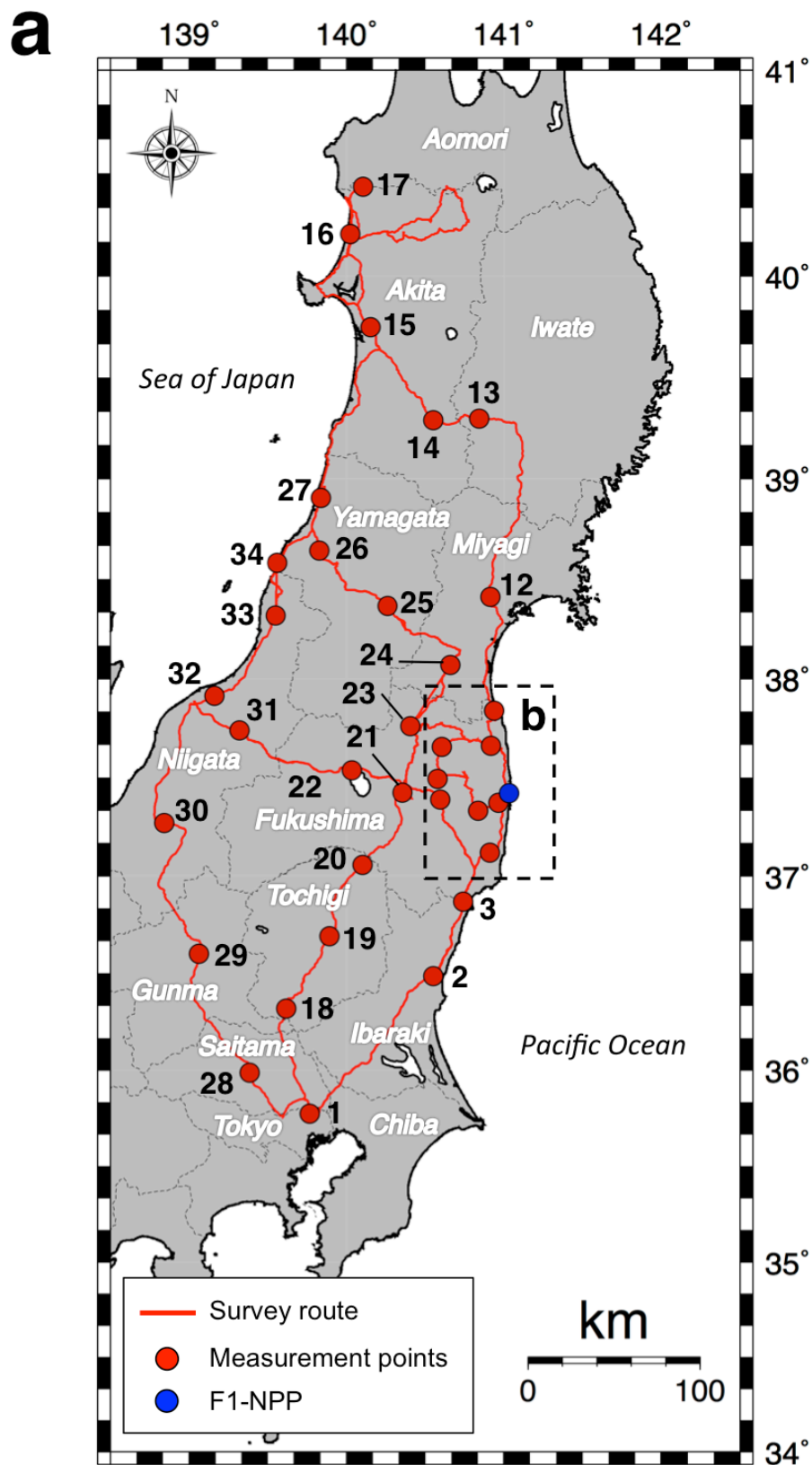
102 radioactivity collected on a dust sampler [14] and was 0.79  $\mu\text{Sv}$  for the shielding factor of  
103 1.53 obtained in this preliminary study. A decontamination project has been carried out at  
104 Tomioka and Okuma Towns since June 2013 [15]. The present discrepancy in values  
105 (0.45  $\mu\text{Sv}$  vs. 0.79  $\mu\text{Sv}$ ) could be attributed to the success of this decontamination project.

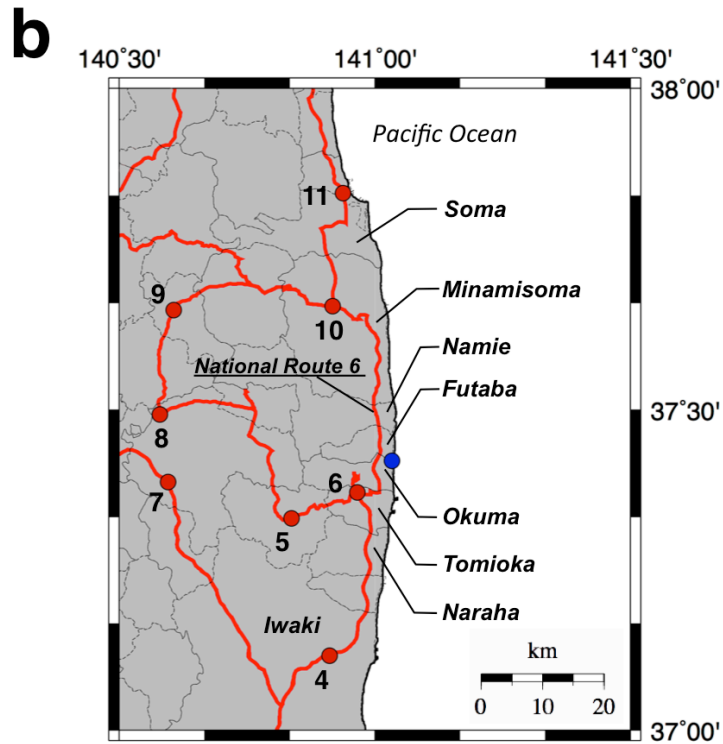
106 Ambient dose rates in air obtained from all radionuclides, natural radionuclides  
107 and artificial radionuclides at 34 locations are shown in Table 1. The mean dose rates  
108 from all, natural and artificial radionuclides were 93  $\text{nGy h}^{-1}$ , 35  $\text{nGy h}^{-1}$  and 57  $\text{nGy h}^{-1}$ ,  
109 respectively. The mean of the contribution ratio of natural radionuclides was 71%. While  
110 higher dose rates in air were observed at #9 (i.e., near the F1-NPP), #32 and #33 (i.e., Sea  
111 of Japan side) compared to the mean dose rate in Japan (51  $\text{nGy h}^{-1}$ ) [9], the contribution  
112 ratios of natural radionuclides were significantly different. Most ambient dose rates in air  
113 observed on the Sea of Japan side are influenced by natural radionuclides and they vary  
114 with the basement geology. Abe et al. [9] observed higher dose rates in air in the northern  
115 area of Niigata Prefecture (94.0 – 130.6  $\text{nGy h}^{-1}$ ) before the accident, and their results  
116 showed the same tendency as the presently obtained results. These areas have mainly  
117 granite formations. The statistical dose rate of this kind of granite has been reported to be  
118  $79 \pm 25 \text{ nGy h}^{-1}$  ( $n = 143$ ) and higher compared to other rocks such as basalt ( $20 \pm 12$   
119  $\text{nGy h}^{-1}$ ;  $n = 49$ ) [10]. Additionally, higher dose rates in air from natural radionuclides  
120 were also observed near the F1-NPP (#6, #8, #20 and #21) and Sea of Japan side (#26,  
121 #27, #31 and #34) for the same reason as given above. The geological map of granite in  
122 eastern Japan is shown in Fig. 3 [16], and the obtained higher dose rates in air from  
123 natural radionuclides were correlated with that distribution. From the results of this study,  
124 it was possible to deduce that when the ambient dose rates in air before the accident were  
125 assumed as the ambient dose rates in air from natural radionuclides as shown in Table 1,  
126 the ambient dose rates in air after the accident had not changed on the Sea of Japan side.  
127 On the contrary, ambient dose rates had increased 0.3 (#21) to 29.2 (#6) times near the  
128 F1-NPP.

129 **Table 1** Ambient dose rates in air and contribution ratios of natural radionuclides in eastern Japan in fall 2014

#*	Shielding factor	Ambient dose rate in air (nGy h <sup>-1</sup> )			Contribution ratio of natural radionuclides (%)	#*	Shielding factor	Ambient dose rate in air (nGy h <sup>-1</sup> )			Contribution ratio of natural radionuclides (%)
		All	Artificial radio-nuclides	Natural radio-nuclides				All	Artificial radio-nuclides	Natural radio-nuclides	
1	1.52	31	0	31	99	18	1.37	43	7	35	83
2	1.37	42	6	36	85	19	1.45	65	35	30	46
3	1.65	82	47	35	43	20	1.54	80	31	50	62
4	1.41	91	63	28	30	21	1.45	69	15	54	78
5	1.64	88	53	35	40	22	1.52	46	4	42	91
6	1.60	1421	1374	47	3	23	1.39	88	64	24	28
7	1.78	58	22	36	63	24	1.54	28	12	16	57
8	1.32	100	50	49	50	25	1.51	38	16	22	59
9	1.46	67	41	26	38	26	1.48	51	2	49	96
10	1.36	90	63	27	30	27	1.43	47	0	47	99
11	1.32	31	13	18	58	28	1.59	37	5	32	87
12	1.54	21	8	13	62	29	1.63	32	2	30	92
13	1.19	21	5	17	79	30	1.55	31	1	30	96
14	1.42	28	2	26	92	31	1.73	51	1	50	97
15	1.07	23	2	21	90	32	1.66	72	0	72	100
16	1.43	28	3	25	88	33	1.40	74	0	74	99
17	1.49	27	3	24	89	34	1.47	50	2	48	96

130 \*Measurement point locations (#) are shown in Fig. 1



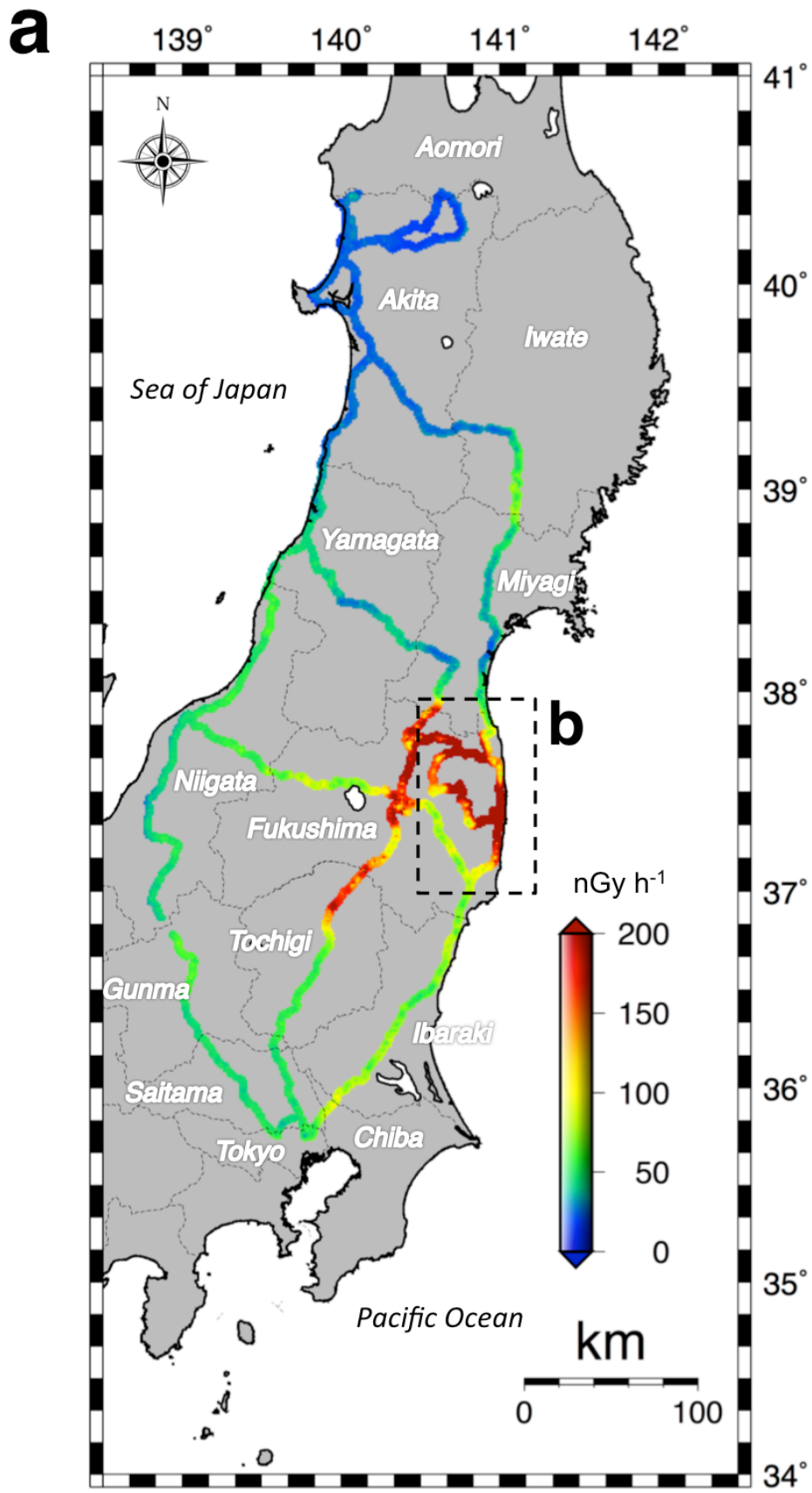


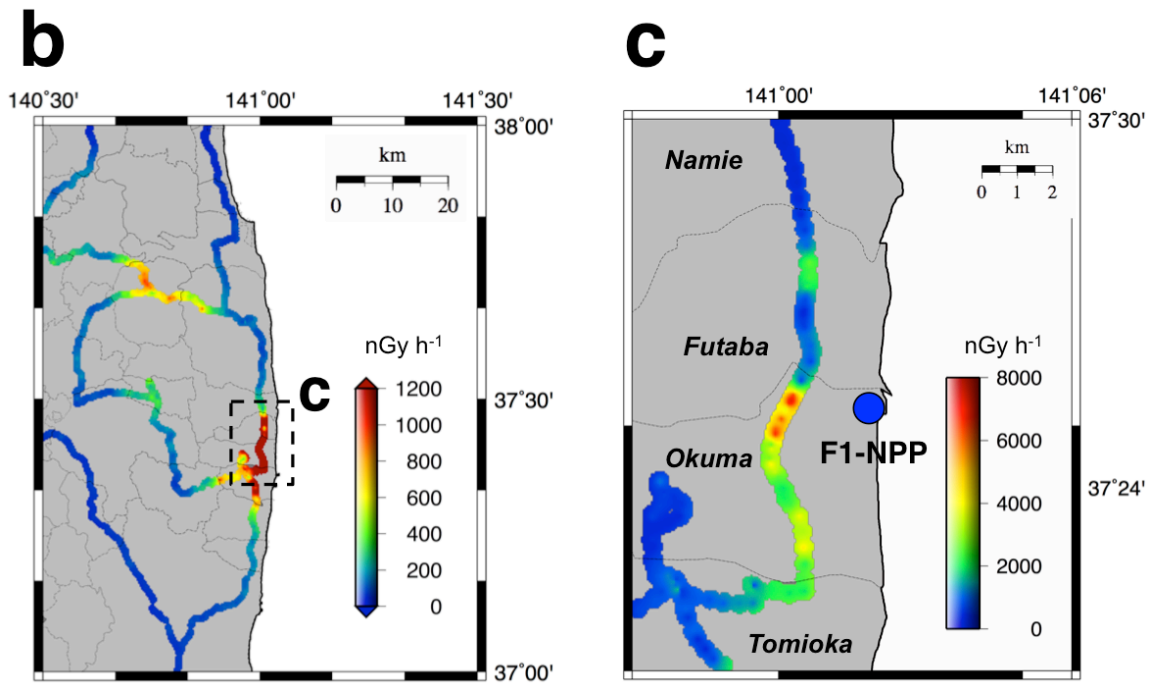
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133 **Fig. 1** Location of the Fukushima Daiichi Nuclear Power Plant, the survey route for  
134 measuring ambient dose rate in air, and the 34 points for the fixed-measurement  
135 observations.

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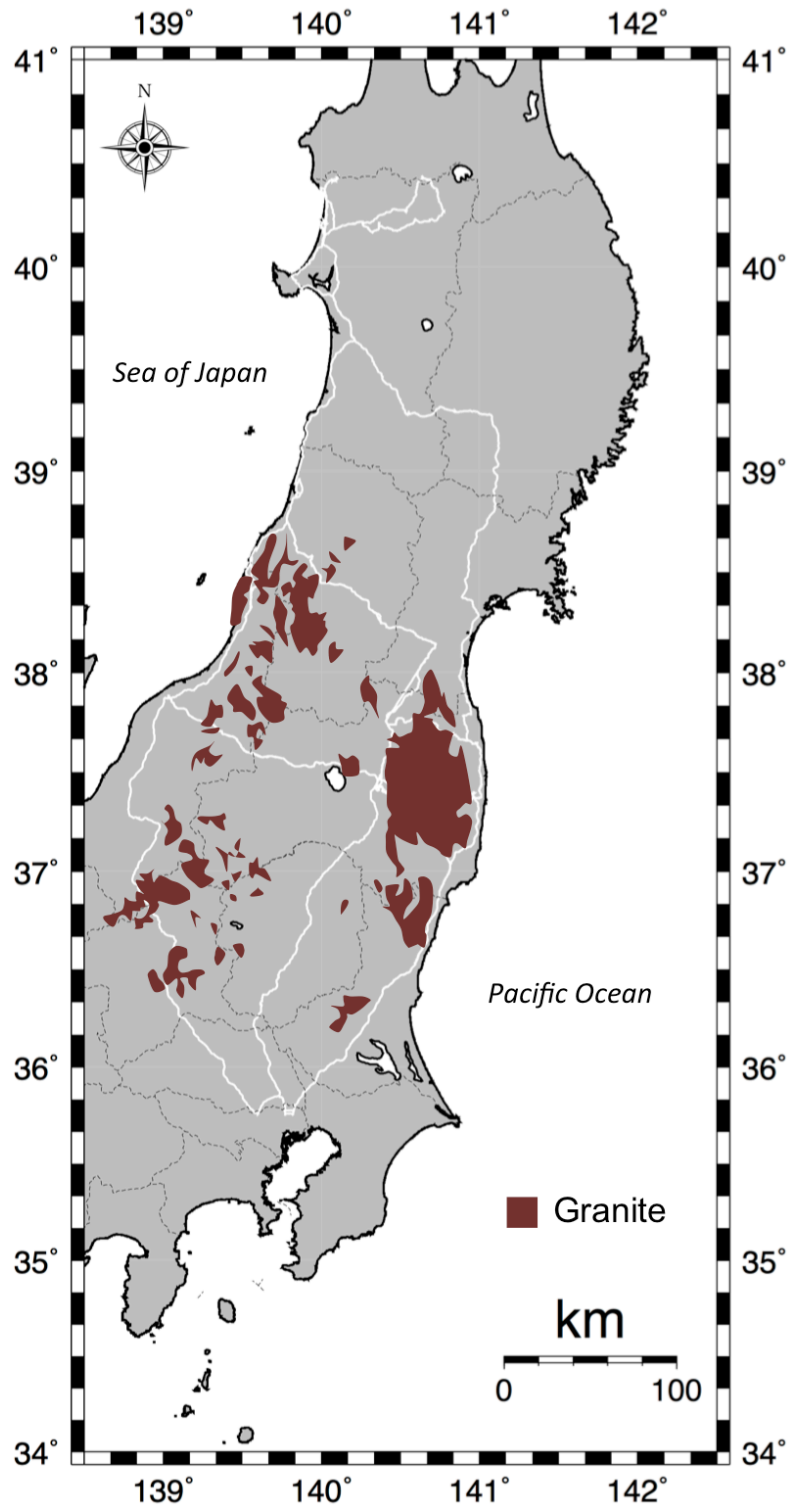




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139 **Fig. 2** The distribution maps of ambient dose rate in air in eastern Japan in fall 2014 with  
140 different magnifications and scale ranges (a, b and c).

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142

143 **Fig. 3** The geological map showing granite formations in eastern Japan [15].

144

145        **Conclusions**

146    A 3,412 km car-borne survey in eastern Japan including an area around F1-NPP (i.e., the  
147    Pacific Ocean side) and the Sea of Japan side was performed. This survey was quite  
148    extensive and considered the underlying geology. While a higher ambient dose rate of  
149    1421 nGy h<sup>-1</sup> was observed in a southwestern area 8 km from F1-NPP (#6), with the  
150    exception of this site, most of the dose rates represented contributions from natural  
151    radionuclides. From the results on the Sea of Japan side, it could be concluded that the  
152    main contribution to the ambient dose rates was influenced by the natural radionuclides  
153    with no evidence of a change in their values that would correlate to the F1-NPP accident.

154        **Acknowledgements**

155    This work was funded by the strategic research fund of Tokyo Metropolitan University.

156        **References**

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