Distribution of radioactive ¹³⁷Cs and ¹³⁴Cs in river water and bottom sand for major rivers at Minami-Soma City in Fukushima

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A nuclear power plant accident, which occurred on March 11, 2011, caused severe radioactive contamination in Fukushima. We initiated an environmental radioactive survey in Minami-Soma City the following October. The city is located approximately 10–40 km north of the TEPCO's Fukushima Daiichi Nuclear Power Plant. The study reported here involves an environmental radioactive survey conducted along three rivers and their tributary streams. Water and bottom sands were collected from four to six sampling locations along each river. Water samples (1 L) were dried on thin Teflon sheets, and gamma-ray measurements were performed with a well-type Ge detector. Sand samples were homogenized after drying, and approximately 50 g were measured for gamma-rays with a low-background coaxial-type Ge detector. The Cs concentrations ($^{137}Cs+^{134}Cs$) were determined to be 10,000–20,000 Bq/kg in the bottom sand collected from the upper reaches of the rivers, whereas the concentrations measured for samples collected near coastal down reaches were measured at about 200 Bq/kg. The Cs concentrations measured in river water were about 0.8 Bq/L in the upper reaches of the rivers and 0.3 Bq/L in the lower reaches, indicating that the Cs concentration in water is quite low. It is necessary to study the behavior of the contaminants in the upper reaches of the river system, and determine if they will move to the down stream area hereafter.

Key Words : Minami-Soma, river water, bottom sand, Cs concentration

1. Introduction

A nuclear reactor accident occurred on March 11, 2011, causing catastrophic radioactive contamination ¹⁾ in the prefecture of Fukushima. Short and long-lived fission products were released into the air and sea in the early stages of the accident. The release also resulted in external and internal exposures to residents. Kamada et al.2) estimated that external and internal exposure impacted the organ doses of thyroid gland for 15 residents in litate Village and Kawamata Town areas, as determined by urinary bioassay method. Impacts to agriculture are believed to be serious due to the potential for radioactive contamination in various foods, which could result in further exposure to people living both in and out of the contamination area. Endo et al.³⁾ measured the transfer coefficient of soil to rice in paddy fields. Such coefficients depend on environmental conditions including the quality of soil and water used in the field. About one half year following the accident, major fission products measured in the environment are primarily ¹³⁷Cs, ¹³⁴Cs, and ⁹⁰Sr. It is reported that the concentration level ⁴⁾ of ⁹⁰Sr is about 1/1000 of the ¹³⁷Cs level.

Minami-Soma City is located approximately 10 -40 km to the north of the TEPCO's Fukushima Daiichi Nuclear Power Plant. The city's eastern sea coast area was severely damaged by the tsunami while the western area of the city was subjected to high levels of contamination. Most of the rivers in Minami-Soma city originate in this high contamination area.

We have initiated an environmental radioactive survey along four rivers and their tributary streams in Minami-Soma City, with work beginning in October of 2011. The purpose of this work is to clarify the radioactive contamination along rivers which are closely related to residents living in the city.

Materials and Methods Samples

There are four major rivers in Minami-Soma City: the Mano, Niida, Ohta, and Odaka Rivers. With the exception of the Odaka River, all of these rivers were included in this study. Locations of these rivers are shown in Fig.1. In addition, the following tributary streams were studied: Mizumashi River to



Fig.1 Three major rivers in Minami-Soma City and sampling locations of river water and bottom sand.

the Niida River, and three tributary streams the Dainichi River (the Kami-mano and the Urugai to the Mano River). The Ohta River has no tributary streams. Sampling was performed from the river mouth to upper stream portions of each river. Sampling points were collected from four to six locations along each river, and from two to three points for the tributary streams. About one-liter of water and bottom sands of about 300 grams (g) to one kilogram (kg) to the depth about 3 cm were collected from the same points for each river. Locations of sampling points are shown in Fig.1. Most sampling points were located around bridges based on convenience and ease of sampling.

2.2 Sample preparation

Bottom sand samples were dried at 100° C for about one day, mixed, and then sand size was selected with a sieve. Sand samples less than 2 mm in diameter were encased in U9 containers (50 mm dia. \times 25 mm height). Each sample weighed approximately 40–50 g. The gamma-ray spectrum was measured with a coaxial type, low background Ge detector (Seiko EG&G, GEM-110225).

Water samples were measured in one liter volume with a volumetric flask, and then dried on a Teflon sheet- lined steel tray of 250 mm \times 250 mm \times 20 mm. Samples were dried with a hot plate kept at 120°. Thereafter, the Teflon sheet was folded to a size of 10 mm dia. \times 40 mm height, and gamma-ray measurements were performed with a low-background, well-type Ge detector (Seiko EG&G,GWL 120230-S). Typical gamma-ray spectra measured for sand sample and water samples at the sampling point O5 are shown in Figs. 2 and 3, respectively.



Fig.2 Typical spectrum of bottom sand of the Ohta River. The sampling location is O5.



Fig.3 Typical spectrum of dry-up water sample of the Ohta River. The sampling location is O5.

3 Results and Discussions

3.1 Bottom sands

Radioactive concentrations of ¹³⁷Cs, ¹³⁴Cs, and ⁴⁰K measured in samples of bottom sand collected from three major rivers and tributary streams are provided in Table 1-3. The results are also shown in Figs. 4-6. It is clear that ¹³⁷Cs and ¹³⁴Cs concentrations are high in the upper streams of the Ohta and Niida Rivers, and then rapidly decrease in the lower reaches of the river. The concentration is about 12,000–25,000 Bq/kg for upper stream samples of O5 and N5, but only 100–200 Bq/kg for down stream. samples of O1 and N1. It is estimated that high concentrations of upper course points O5, N8, and N5 can be attributed to direct deposition resulting from the reactor accident, and could also be coming from the high contamination area of the litate village area.

The concentrations measured in the Mano River show a somewhat different distribution when compared to other rivers, with concentrations of ¹³⁷Cs and ¹³⁴Cs measured at one order lower than the other rivers. The upper stream sample of M6 shows 158 Bq/kg and a maximum concentration of

Table 1 Bottom sand samples of the Ohta River.

Sampling location	¹³⁷ Cs(1	Bq/	′kg)	¹³⁴ Cs(Bq	/kg)	¹³⁷ Cs+	134	Cs(Bq/kg)	⁴⁰ K(H	3q/	′kg)
O5 Yaigomesaka bridge	15396	±	1089	9148	±	647	24544	±	1267	726	±	63
O4 Kaminouchi bridge	8826	±	624	5210	±	368	14036	±	724	969	±	83
O3 Masuda bridge	1970	±	139	1150	±	81	3120	±	161	1033	±	88
O2 Takagawa bridge	1435	±	102	843	\pm	60	2278	±	118	940	±	80
O1 Tokiwa bridge	76	±	6	42	\pm	3	118	±	6.7	612	±	53

Table 2 Bottom sand samples of the Niida River and Mizunashi tributary stream.

Sampling location	¹³⁷ Cs(B	q/kg	g)	¹³⁴ Cs(Bq	/kg)	¹³⁷ Cs+	¹³⁴	Cs(Bq/kg)	⁴⁰ K(1	Bq/	kg)
N5 Nakagawara bridge	7547	± 5.	34	4517	±	320	12064	±	623	868	±	75
N4 Confluence point	1285 =	± 9	1	763	±	54	2048	±	106	658	±	57
N3 Shinsakurai bridge	881 =	± 6	3	579	±	41	1460	±	75	600	±	54
N2 Sakegawa bridge	826 =	± 5	9	411	±	29	1237	±	66	787	±	69
N1 River mouth	145	± 1	0	79	±	6	224	±	12	303	±	13
[Mizunashi river]												
N8 Takanokura dam	6654 =	± 4'	71	3983	±	382	10637	±	606	766	±	66
N7 Sakashita bridge	1532 =	± 1	09	907	±	64	2439	±	126	1014	±	87
N6 Confluence point	2108 =	± 14	49	1249	±	88	3357	±	173	718	±	62

Table3 Bottom sand samples of the Mano River and its tributaries: the Urugai, Kami-mano and Dainichi Rivers.

Sampling location	¹³⁷ Cs(Bq/	kg)	$^{134}Cs(1)$	Βą	/kg)	¹³⁷ Cs+	¹³⁴	Cs(Bq/kg)	⁴⁰ K(H	3q/1	kg)
M6 Mano dam	105 ±	8	53	±	4	158	±	9	683	±	58
M5 Furukawa bridge	526 ±	37	246	±	19	772	±	42	637	±	54
M4 Kagitori bridge	1165 ±	82	592	±	42	1757	±	92	671	±	57
M3 Kurumagawa bridge	958 ±	68	481	±	34	1439	±	76	749	±	63
M2 Sakurada bridge	182 ±	13	92	±	7	274	±	15	608	±	57
M1 River mouth	328 ±	23	169	±	12	497	±	26	247	±	21
[Urugai river]											
M7 Shiozaki bus stop	2573 ±	182	1435	±	102	4008	±	209	402	±	36
M8 Annoko bridge	3714 ±	263	2072	±	147	5786	±	301	438	±	39
{Kami-mano river]											
M9 Kamahai bridge	1232 ±	87	687	±	49	1919	±	100	630	±	54
M10 Zentoku bridge	1255 ±	89	707	±	50	1962	±	102	623	±	54
[Dainichi river]											
M11 Kakurew bridge	$800 \pm$	57	443	±	31	1243	±	65	593	±	51
M12 Kuramae bridge	606 ±	43	340	±	24	946	±	49	507	±	44

Table 4 Water samples along the Ohta River.

Sampling location	¹³⁷ Cs (B	q/L)	¹³⁴ Cs	s (Bq/L)	¹³⁷ Cs+	⁻¹³⁴ Cs (Bq/L)
O5 Yaigomesaka bridge	0.460 ±	0.023	0.350	± 0.022	0.810	± 0.032
O4 Kaminouchi bridge	0.300 ±	0.018	0.248	± 0.013	0.548	± 0.022
O3 Masuda bridge	0.240 ±	0.012	0.207	± 0.014	0.447	± 0.018
O2 Takagawa bridge	0.255 ±	0.015	0.223	± 0.017	0.478	± 0.023
O1 Tokiwa bridge	0.436 ±	0.018	0.276	± 0.017	0.712	± 0.025

Table 5 Water samples along the Niida River and Mizumashi tributary stream.

Sampling location	¹³⁷ Cs (Bq/L)			¹³⁴ Cs	s (B	lq/L)	$^{137}Cs+^{134}Cs$ (Bq/L)				
N5 Nakagawara bridge	0.432	±	0.024	0.383	±	0.027	0.815	±	0.036		
N4 Confluence point	0.114	±	0.010	0.091	±	0.007	0.205	±	0.012		
N2 Sakegawa bridge	0.098	±	0.007	0.046	±	0.008	0.144	±	0.011		
N1 River mouse	0.172	±	0.010	0.157	±	0.012	0.329	±	0.016		
[Mizunashi river]											
N8 Takanokura dam	0.395	±	0.022	0.328	±	0.023	0.723	±	0.032		
N7 Sakashita bridge	0.145	±	0.011	0.143	±	0.013	0.288	±	0.017		
N6 Confluence point	0.129	±	0.008	0.110	±	0.010	0.239	±	0.013		

Sampling location	¹³⁷ Cs	(Bg	/L)	¹³⁴ Cs	s (B	Bq/L)	¹³⁷ Cs-	⊦ ¹³⁴ (Cs (Bq/L)
M6 Mano dam	0.146	±	0.008	0.092	±	0.009	0.238	±	0.012
M5 Furukawa bridge	0.109	\pm	0.007	0.053	±	0.016	0.162	±	0.017
M4 Kagitori bridge	0.166	\pm	0.007	0.098	±	0.014	0.264	±	0.016
M3 Kurumagawa bridge	0.068	\pm	0.005	0.037	±	0.007	0.105	±	0.009
M2 Sakurada bridge	0.132	\pm	0.008	0.071	±	0.008	0.203	±	0.012
M1 River mouse	0.410	\pm	0.014	0.282	±	0.017	0.692	±	0.022
[Urugai river]									
M7 Shiozaki bus stop	0.290	±	0.011	0.162	±	0.009	0.452	±	0.014
M8 Annoko bridge	0.248	±	0.011	0.212	±	0.014	0.46	±	0.018
[Kami-mano river]									
M9 Kamahai bridge	0.639	±	0.023	0.551	±	0.023	1.190	±	0.033
M10 Zentoku bridge	0.531	±	0.021	0.396	±	0.020	0.927	±	0.029
[Dainichi river]									
M11 Kakurew bridge	0.084	±	0.008	0.058	±	0.007	0.142	±	0.011
M12 Kuramae bridge	0.183	±	0.031	0.106	±	0.011	0.289	±	0.033

Table 6 Water samples of the Mano River and branches of the Urugai, Kami-mano and Dainichi Rivers.



Fig.4 Bottom sand samples of the Ohta River.



Fig. 5 Bottom sand samples of the Niida River.



Fig.6 Bottom sand samples of the Mano River.



Fig.7 Water samples along the Ohta River.



Fig. 8 Water samples along the Niida River.



Fig.9 Water samples along the Mano River.

about 1600 Bq/kg around M4 and M3 (located at about halfway down the Mano River). The concentrations measured in tributary stream M7 and M8 of 4000–6000 Bq/kg are higher than those measured in the upper stream of M6. The lower distribution of contaminants along the Mano river could be due to streams not being fed from the high contamination area of Iitate village. Further surveys may be warranted along the Mano River.

The concentrations of 40 K measured at 500–800 Bq/kg, as provided in Tables 1-3, are rather uniformly distributed along each river. Correlations between the concentrations of 40 K and 137 Cs or 134 Cs are not observed.

3.2 River water

The concentrations of ¹³⁷Cs and ¹³⁴Cs measured in river water are provided in Tables 4-6. The results are also shown in Figs. 7-9. The concentrations in river water were less than 1 Bq/L for all samples, indicating that Cesium is attached to soil and does not easily migrate in water. Distributions of water concentrations show that the concentration is generally high in the upper reaches of streams, and then gradually decreases in the lower portions of streams. Such trends are in agreement with measurements of bottom sands in the river. The increase in concentrations measured at river mouths is believed to come from sea water mixing. This is further supported by high concentrations of salt measured in water samples O1, N1, and M1. The distribution of contaminants measured along the Mano River is different from that of other rivers. The Cs concentrations of the Mano River are almost uniform, in contrast to the measurements collected from the other rivers.

4. Conclusion

The Cs concentrations (¹³⁷Cs+¹³⁴Cs) were measured at 10,000–20,000 Bq/kg in the bottom sands collected from upper reaches of the rivers, whereas the concentrations measured from the lower reaches near the sea coast were measured at about 200 Bq/kg. The Cs concentrations measured in river water were about 0.8 Bq/L in the upper reaches of the river, and 0.3 Bq/L in the lower reaches, indicating that the Cs concentrations in water are actually quite low. In the case of the Mano River, Cs concentrations measured in bottom sands and river water showed a different distribution from the other rivers. It is necessary to follow the behavior of the contamination at the upstream of the river to determine how it will migrate to the downstream area hereafter.

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