REVIEW OF INTERNATIONAL SOURCE TERM RELEASE HISTORIES FOR THE ESTABLISHMENT OF RADIOLOGICAL EMERGENCY PREPAREDNESS FRAMEWORK OF NUCLEAR POWER PLANT

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Since TMI-2 accident happened in 1979, research on severe accident started. Historical source term researches are reviewed first. There also has been efforts to make tools which can simulate severe accidents which can be happen in nuclear power plants realistically. Comparisons are made among three real source term release accidents occurred historically in nuclear power plants in terms of major released types of radionuclides, release magnitude, and released places. These aspects are reviewed and discussed in detailed manner in the main paper. Insights obtained during this study will be helpful to guiding correct directions for severe accident simulation tool development. Harmony among regulatory framework, tool development and analysis work will be needed in design of nuclear power plant and planning of emergency response preparedness.

I. INTRODUCTION

For the radiological emergency preparedness of nuclear power plant, it is needed to improve the understanding on the severe accident behaviors and source term analysis techniques in the nuclear power plant. For the establishment of correct and effective emergency preparedness, severe accident analysis and source term characterization is very important. For the establishment of appropriate and effective emergency preparedness, severe accident analysis and source term characterization is very important. To establish the framework on the source term analysis technique, the following works are performed in this project: (1) Review of source term research history in USA, (2) Review of experimental research on source term issues in other countries, (3) Review of source term research history in Korea, and (4) Documental search on fission product aerosol release amount from real accidents happened in nuclear power plant, such as TMI 2 (1979), Chernobyl (1986), and Fukushima Daiichi (2011) accidents. The search results on source terms in this project will be helpful to establish research direction in Korea. For the radiological emergency preparedness of nuclear power plant, it is needed to improve the understanding on the severe accident behaviors and source term release characteristics in the nuclear power plant. For more detailed information, you may refer to the following two reports, Kim T.W. et al., KAERI/AR-1099/2015 Review of International and Domestic Source Term Research Histories for the Establishment of Radiological Emergency Preparedness Framework of Nuclear Power Plant

There is a long history of applying radiological source terms to the reactor risk study, siting criteria development and radiological emergency preparedness of the light water reactors: TID-14844, NUREG-1465 (Accident Source Terms), WASH-1400, NUREG-1150, etc. Recently, the SOARCA project (US NRC, 2012) in U.S. NRC (Nuclear Regulation Commission) has treated long-term and short-term SBO accident sequences for Surry (Large dry containment PWR) and Peach Bottom (MARK I BWR) plants and presented the reduced release amounts of radiological source term with the current-state-of-the-art knowledge of radiological transport in the severe accident environment by MELCOR code (US NRC, 2005). ORIGEN code is usually used to estimate initial inventory of fission product and activation product. To estimate environmental impact MACCS2 code is used. RASCAL program is used to plan and to simulate emergency drill.

Each country is developing his own severe accident codes for the thermal hydraulics and source term analysis in severe accident of nuclear power plants. MAAP code is used in US nuclear industries for the analysis of severe accidents. ASTEC code is developed in Europe. THALES/ART code is developed in Japan. SAMPSON code is under development in Japan by IAE.

Since the Three Mile Island (TMI) (1979), Chernobyl (1986), Fukushima Daiichi (March 11, 2011) accidents, the assessment of radiological source term effects on the environment has been a key concern of nuclear safety. Source terms are estimated by reverse or inverse method using the monitoring data from the real severe accident of TMI-2, Chernobyl, and Fukushima

Daiichi accidents. IAEA 2003, 2006 reports and UNSCEAR-2008 report summarized the estimated environmental source terms and human health impacts for Chernobyl accident. UNSCEAR-2013, WHO 2012, 2013 reports summarized the estimated environmental source terms and human health impacts from Fukushima accidents. Environmental source terms from these three accidents are reviewed and summarized in this paper.

II. ACCIDENT SEQUENCES

II.A TMI-2 Accident

TMI-2 accident occurred at Pensylvenia, USA, in 1979. Even though reactor core was melted and relocated to lower plenum region in reactor vessel, there was no reactor vessel failure. Intact reactor vessel and containment made no much fission product aerosols are released to the environment.

II.B Chernobyl Accident

Chernobyl accident occurred on April 26, 1986 in former Soviet Union. The accident at unit 4 of the Chernobyl nuclear power plant took place shortly after midnight on 26 April 1986. Radioactive materials released to atmosphere during 10 days. Fig. 1 shows daily release rate to the atmosphere of radioactive material in Chernobyl Accident (decay corrected to 6 May 1986) (IAEA, 2006). Among the 3300, 3100, 190 and 290 PBq of Te-132, I-131, Cs-134, and Cs-137 initial inventory, about 35%, 57%, 25% and 29% are released to atmosphere. Even though it is only one core, larger amount of nuclides are released to the environment due to the graphite fire during 10 days.

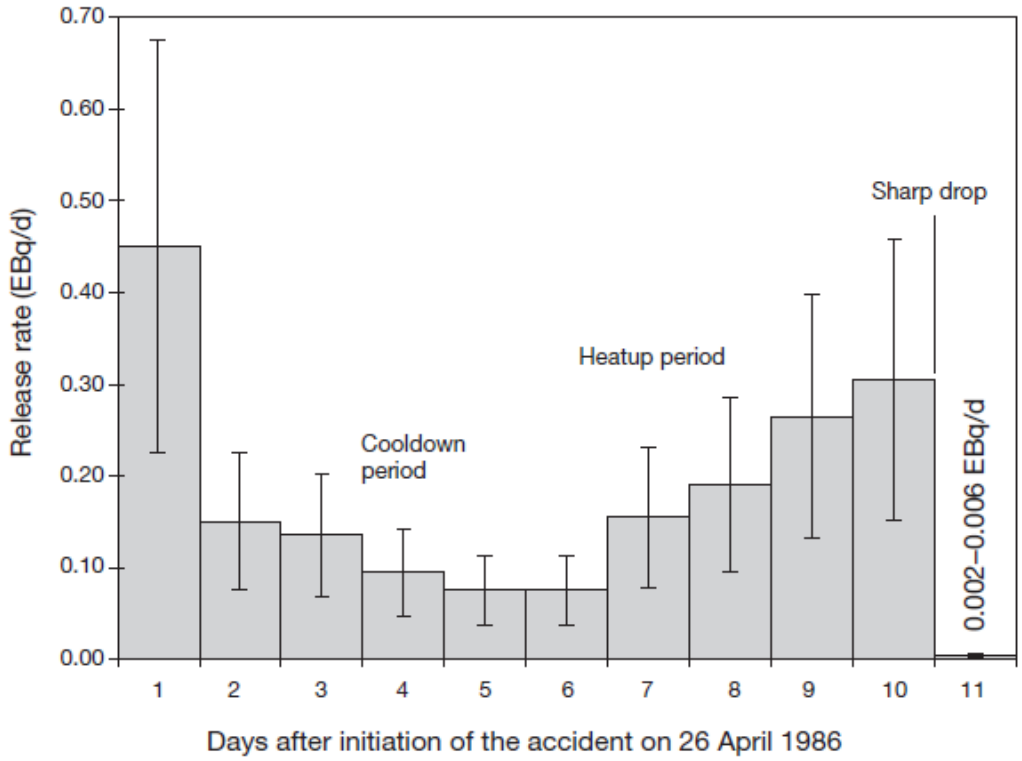

Fig.1 Daily release rate to the atmosphere of radioactive material in Chernobyl Accident (decay corrected to 6 May 1986)
Source : IAEA (2006)

Most of the radionuclides for which there were large releases have short physical half-lives, and the radionuclides with long half-lives were mostly released in small amounts. In the early period after the accident, the radionuclide of most radiological concern was ^{131}I; later, the emphasis shifted to ^{137}Cs.

By 2005 most of the radionuclides released by the accident had already decayed below levels of concern. Interest over the next few decades will continue to be on ^{137}Cs and, to a lesser extent, ^{90}Sr; the latter remains more important in the near zone of the Chernobyl nuclear power plant. Over the longer term (hundreds to thousands of years), the only radionuclides anticipated to be of interest are the plutonium isotopes. The only radionuclide expected to increase in its levels in the coming years is ^{241}Am, which arises from the decay of ^{241}Pu; it takes about 100 years for the maximum amount of ^{241}Am to form from ^{241}Pu.

II.C Fukushima Daiichi Accident

Great Tohoku earthquake occurred at 14:37 JST on March 11, 2011. Reactor trip occurs at Fukushima Daiichi Units 1, 2, and 3. However, with loss of electrical powers at these units, there is no way to cooldown decay heat generated in reactor cores. Therefore, fission products released from high temperature fuel start to release to environment at 20, 40, and 80 hours after the reactor trip at Units 1, 3, and 2, respectively. Fission product gamma ray release was detected in monitoring cars at the site. Monitoring post at site cannot be operated due to the loss of electrical power.

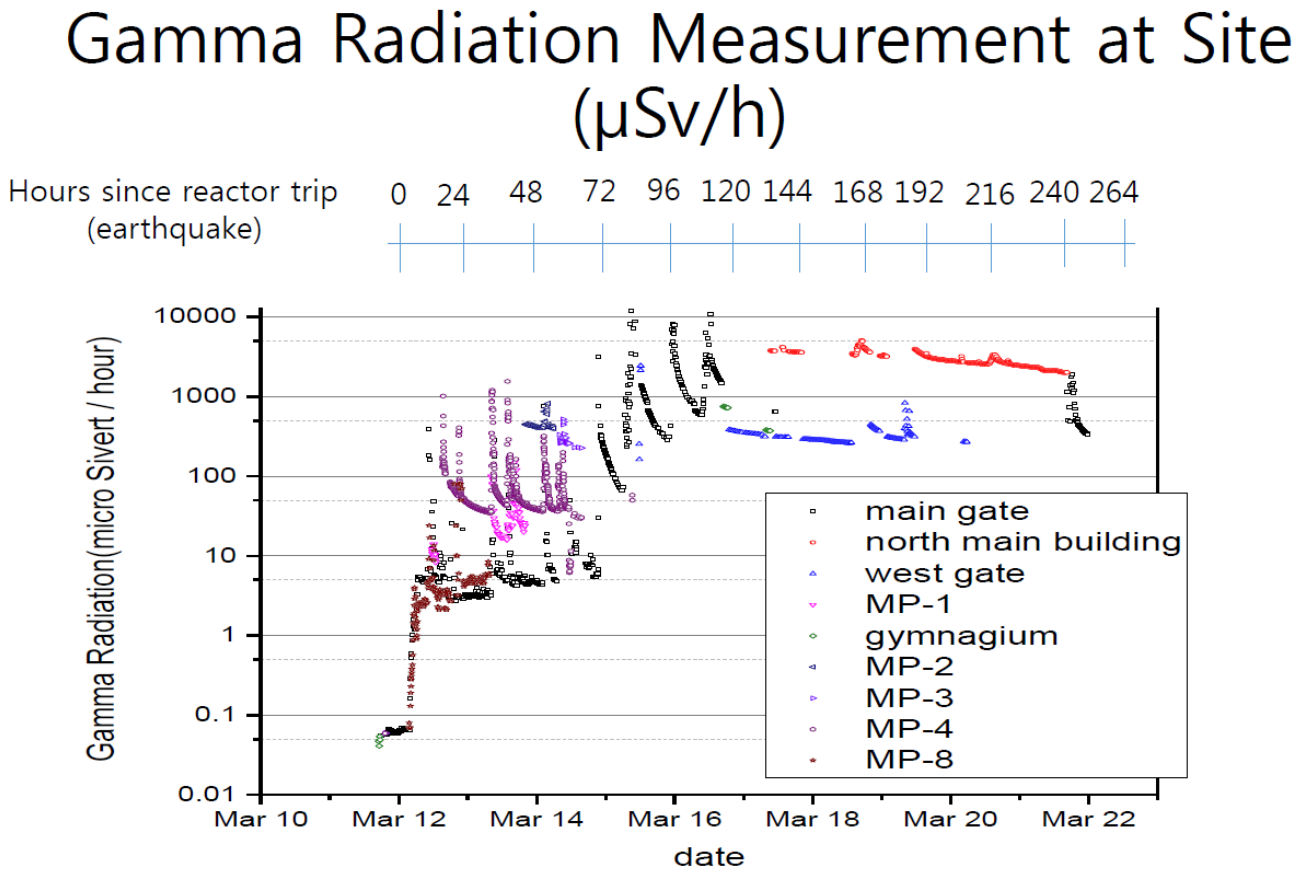

Fig.2 Gamma radiation monitoring results in Fukushima Daiichi site by monitoring post and cars (Kim et al., 2016)

A budget analysis indicated that approximately 13% of I-131 and 22% of Ce-137 were deposited over land in Japan, and the rest was deposited over the ocean or transported out to the model domain (700 km x 700 km). Radioactivity budgets are sensitive to temporal emission patterns Accurate estimation of emissions to the air is important for estimation of the atmosphere behavior of radionuclides and their subsequent behaviors in land water, soil, vegetation, and the ocean. (Morino)

III. COMPARISON OF RELEASE TO ATMOSPHERE

Initial core inventory is usually calculated by ORIGEN-2 code. Initial core inventory and release fraction to atmosphere is compared in Table 1. Initial core inventory data is well summarized in Guntay et al. IAEA (2003, 2005) and UNSCEAR

(2008) reports estimated the atmospheric source terms on the Chernobyl accident. Initial inventory and release fraction data of Fukushima is well summarized in the book edited by Povinec et al. (2016).

III.A Behavior of Noble Gases (Kr, Xe)

Initial inventory of Fukushima is about 2 times higher than those of Chernobyl and TMI-2. This is because Fukushima is composed of three units while Chernobyl and TMI-2 composed of only one unit. Therefore, about 2 times higher activity is released in Fukushima compared with Chernobyl. It is assumed that 100% of noble gases are released to atmosphere in both accidents.

In the TMI-2 accident, however, among 25 PBq of Kr-85 and 3412 PBq of Xe-133, only 9% of initial core inventory (2.26 PBq of Kr-85 and 307 PBq of Xe-133) is released to atmosphere. The other 91 % is retained in containment basement.

II.B Behavior of Volatile Elements (Cs, I, Te)

In the TMI-2 accident, very small amount of I-131 (0.52 TBq) and I-133 (0.1 TBq) is released to atmosphere. (Gudiksen, 1990). In the Chernobyl accident, 25 – 57 % of volatile element are released to atmosphere. 57% (1760 PBq) of I-131 and 29% (85 PBq) of Cs-137 inventory is released to atmosphere. In the Fukushima accident, 1 – 8% of inventory is released to atmosphere. 2.6% (159 PBq) of I-131 and 2.2% (15 PBq) of Cs-137 is estimated to be released to atmosphere. MELCOR code is used to estimate atmospheric release in Fukushima accident (NERH, 2012).

II.C Behavior of Intermediately Volatile Elements (Sr, Ba, Ru)

In the Chernobyl accident, 3.4 – 5.0 % of intermediately volatile elements are released to atmosphere. 5% (115 PBq) of Sr-89 and 5% (10 PBq) of Sr-90 are released to atmosphere. In the Fukushima accident, 0.03% (1.96 PBq) of Sr-89 and 0.03% (0.139 PBq) of Sr-90 are estimated to be released to the atmosphere. In the TMI-2 accident, 0.1% of Sr-90 and 0.7% of Sb-125 inventory is released to the auxiliary building.

II.D Behavior of Refractory Elements (Zr, Mo, Ce) and Actinides (Np, Pu, Cm)

In the Chernobyl accident, 1.5% of refractory element and actinides are released to the atmosphere in average. Meanwhile, in the Fukushima accident, 1E-7 – 1E-6 fractions of initial inventory of actinides are released to the atmosphere in average. The reason of higher fraction of release in Chernobyl than that of Fukushima is believed that 10 days long graphite fire occurs in Chernobyl core while in Fukushima it is a slow pressurizing sequence in RPV and PCV.

Table 1. Comparison of atmospheric release between Chernobyl and Fukushima accidents

Radionuclide	Half-life		Chernobyl Inventory (PBq)	Chernobyl Activity Released (PBq)	Chernobyl Released Fraction	Ratio Fukushima to Chernobyl Inventory	Fukushima Inventory (PBq)	Fukushima Released to Atmosphere (PBq)	Fukushima Released Fraction to Atmosphere	Ratio Fukushima to Chernobyl Release
			(A0)	(A)	(A1=A/A0)	(A2=B/A0)	(B)	©	(D=C/B)	(E=C/A)
Inert gases										
Kr-85	10.72	a	33	33	100%	2.54	83.7	83.7	100%	254%
Xe-133	5.25	d	7300	6500	89%	1.64	12000	12000	100%	185%
Volatile elements										
Te-129m	33.6	d		240			189	3.33	1.8%	1.4%
Te-132	3.26	d	3300	1150	35%	2.63	8690	88.4	1.0%	7.7%
I-131	8.04	d	3100	1760	57%	1.94	6010	159	2.6%	9.0%
I-133	20.8	h		910			527	42.2	8.0%	4.6%
Cs-134	2.06	a	190	47	25%	3.78	719	17.5	2.4%	37%
Cs-136	13.1	d	110	36	33%	1.98	218			
Cs-137	30	a	290	85	29%	2.41	700	15.3	2.2%	18%
Elements with intermediate volatility										
Sr-89	50.5	d	2300	115	5.0%	2.58	5930	1.96	0.03%	1.7%
Sr-90	29.12	a	200	10	5.0%	2.61	522	0.139	0.03%	1.4%
Ru-103	39.3	d	5000	168	3.4%					
Ru-106	368	d	2000	73	3.7%					
Ba-140	12.7	d	5300	240	4.5%					
Refractory elements (including fuel particles)										
Zr-95	64	d	5800	84	1.4%					
Mo-99	2.75	d	7300	72	1.0%	1.56	11400	6.70E-06	5.88E-10	9.3E-08
Ce-141	32.5	d	5600	84	1.5%					
Ce-144	284	d	3200	50	1.6%	1.85	5920	1.15E-02	1.94E-06	2.3E-04
Np-239	2.35	d	36000	400	1.1%					
Pu-238	87.74	a	1	0.015	1.5%	14.70	14.7	1.88E-05	1.28E-06	1.3E-03
Pu-239	24065	a	0.85	0.013	1.5%	3.08	2.62	3.23E-06	1.23E-06	2.5E-04
Pu-240	6537	a	1.2	0.018	1.5%	2.73	3.27	3.13E-06	9.57E-07	1.7E-04
Pu-241	14.4	a	170	2.6	1.5%			1.25E-03		4.8E-04
Pu-242	376000	a		0.00004						
Cm-242	18.1	a	250	0.4	0.2%	1.13	283	1.02E-04	3.60E-07	2.6E-04
Total (except noble gases)			80113	5527	6.9%	0.51	41129	328	7.97E-03	5.9E-02

III. COMPARISON OF RELEASE TO STAGNANT WATER

Nishihara et al. (2012) estimated fission product release to stagnant water in reactor building, turbine building, radwaste treatment building, trench and central radwaste facility, etc. for Fukushima Daiichi Units 1, 2, and 3. TEPCO measured radioactivity in stagnant water in each building about 10 times during March 24 through May 27, 2011. Resulting release ratio from core to stagnant water is shown in Table 2. They are compared with TMI-2 results (Akers, 1990). Most of them are exist in in-vessel and containment basement.

In the Fukushima accident, 32% of I-131 is released to stagnant water of various buildings. 20% of Cs-134 and Cs-137 is released to stagnant water in multiple buildings. 1.2% of Sr-89 and 1.6% of Sr-90 is released to stagnant water of buildings.

Unit 2 has highest release to stagnant water among 3 units while Unit 1 has lowest.

Table 2. Release fraction to Stagnant Water in Fukushima Units 1, 2, 3

	1F1	1F2	1F3	Total	TMI-2[1)]	TMI-2[2)]
I131	6.9%	52%	27%	32%	14%	55%
Cs134	6.8%	33%	17%	21%		
Cs136	7.0%	27%	14%	18%		
Cs137	6.2%	34%	17%	20%	41%	55%
Ba140		1.0%	0.7%	0.7%		
La140	1.5E-05	0.5%	0.1%	0.2%		
H3	5.9%	57%	67%	46%	57%	
Sr89	5.1E-06	1.6%	1.5%	1.2%		
Sr90	1.1E-05	2.4%	2.2%	1.6%	1.8%	3.2%
Tc99m		1.0%	0.5%	0.6%		
Sb125		0.04%			0.5%	1.6%
Ru106					6.0E-05	0.5%

1) McIsaac (1988)
2) Akers (1990)

In the TMI-2 accident, high volatile radionuclides such as Cs-137, I-129, and Kr-85 are retained in containment structures and water. 5% of Cs-137 and 7% of I-129 initial inventory are retained in auxiliary building.

Less amount of medium volatile radionuclides are retained in containment, RCS, and auxiliary building compared with the case of high volatile radionuclides. 0.1% of Sr-90 and 0.7% of Sb-125 retained in auxiliary building. Low volatile materials and actinides are all retained in in-vessel.

Table 3. Compartmental deposition fraction in Fukushima accident

	Low volatile radionuclide			Medium volatile radionuclide			High volatile radionuclide		
	Ce-144	Eu-154	Eu-155	Sr-90	Ru-106	Sb-125	Cs-137	I-129	Kr-85
Ex-vessel									
Containment atmosphere, basement, and tanks	0.01			2.1	0.5	0.7	47	47	54
Reactor coolant system				1.0		0.2	3	1	
Auxiliary building				0.1		0.7	5	7	
In-vessel									
Upper reactor plenum									
Upper core debris	20	19	19	19	16	24	4.3	5.3	6
Upper crust region	1.4	2	1.6	0.73	4.6	8.3	0.41	0.27	
Consolidated region	24	32	22	8.3	11.2	10	0.77	2.1	
Lower crust	5.9	7.9	5.1	4.5	29.7	43.4	1.4	3.5	
Intact fuel rods	30	30	30	30	30	30	30	30	30
Upper core support assembly	3.4	4.5	4.5	3.9	0.23	0.22	0.46	0.12	
Lower core support assembly	4.7	6.3	6.3	5.3	0.32	0.3	0.63	0.16	
Lower head - reactor vessel	16	21	21	18	1.1	1	2.1	0.54	
Total	105	123	110	93	94	119	95	97	90

Source : Akers (1990)

VI. OCEANIC RELEASE

137Cs Budget Estimates

Global fallout as of 1970	290 ± 30 PBq a
Estimated N. Pacific pre-Fukushima	69 PBq b
Fukushima direct discharge	3.5-15 PBq c
Fukushima atmospheric release	10 - 16 PBq d
Fukushima deposition on land	2 – 2.9 PBq e
Total Fukushima in ocean	14 – 31 PBq
Chernobyl global release in 1986	85 PBq f
(10-20% of Chernobyl fallout in oceans)	9-17 PBq g

a: Aoyama, M., Hirose, K., Igarashi, Y., J. ENVIRON. MONITOR., 8, 431-438, 2006
b: Aoyama unpublished data estimated 3-D distribution of 137Cs
c: Tsumune et al., 2011, Rypina et al., 2012, Charette et al., 2012
d: Chino et al., 2011, Morino et al, 2011, ISRN 2011, Aoyama et al. in prep.
e: Aoyama et al. in prep.
f: IAEA, Proceeding of an International Conference, Vienna, 8-12 Apr. 1996
g: Buesseler, 2012

Due to the Chernobyl accident, Cs-137 concentration of black sea was increased. Among total release of 85 PBq of Cs-137, about 10-20% (9-17 PBq) are estimated to be fell out to sea.

Tsumune (2012) reported that large direct discharge to sea is made during from March 26 to April 6, 2011 in Fukushima site. He estimated that about 3.5 PBq of Cs-137 is discharged to sea during that period. Oceanic concentrations of Cs-137 are measured by TEPCO, JAEA and many other organizations and reported in technical papers and many websites including TEPCO website.

Bailly du Bois (2012) estimated that oceanic source term of Cs-137 as 27 +/- 15 PBq (12 – 41 PBq) up to July 18, 2011. Most of the marine source terms are deposited on to ocean surface which was released to atmosphere by the explosions or discharges to atmosphere by venting and leakages. There is an estimation that more than 80% of airborne release are deposited on to the ocean, 19% are deposited on the terrestrial region of Japan, and other 1% are flowed to other countries. Oceanic input from Fukushima accident is estimated to be 14 – 31 PBq.

Before the Fukushima accident, distribution and inventory of ^{137}Cs which originated from atmospheric weapons tests had been studied in the Pacific Ocean since the late 1950s and the ^{137}Cs inventory in the North Pacific Ocean was 290 ± 30 PBq in January 1970 and it decreased to 69 PBq in 2011 because due to decay and inter-basin transport from the North Pacific Ocean to Indian Ocean and the South Pacific Ocean. Fukushima accident contributed 137Cs inventory in the North Pacific Ocean by 22–27 % (12-15 PBq) (Aoyama, 2016).

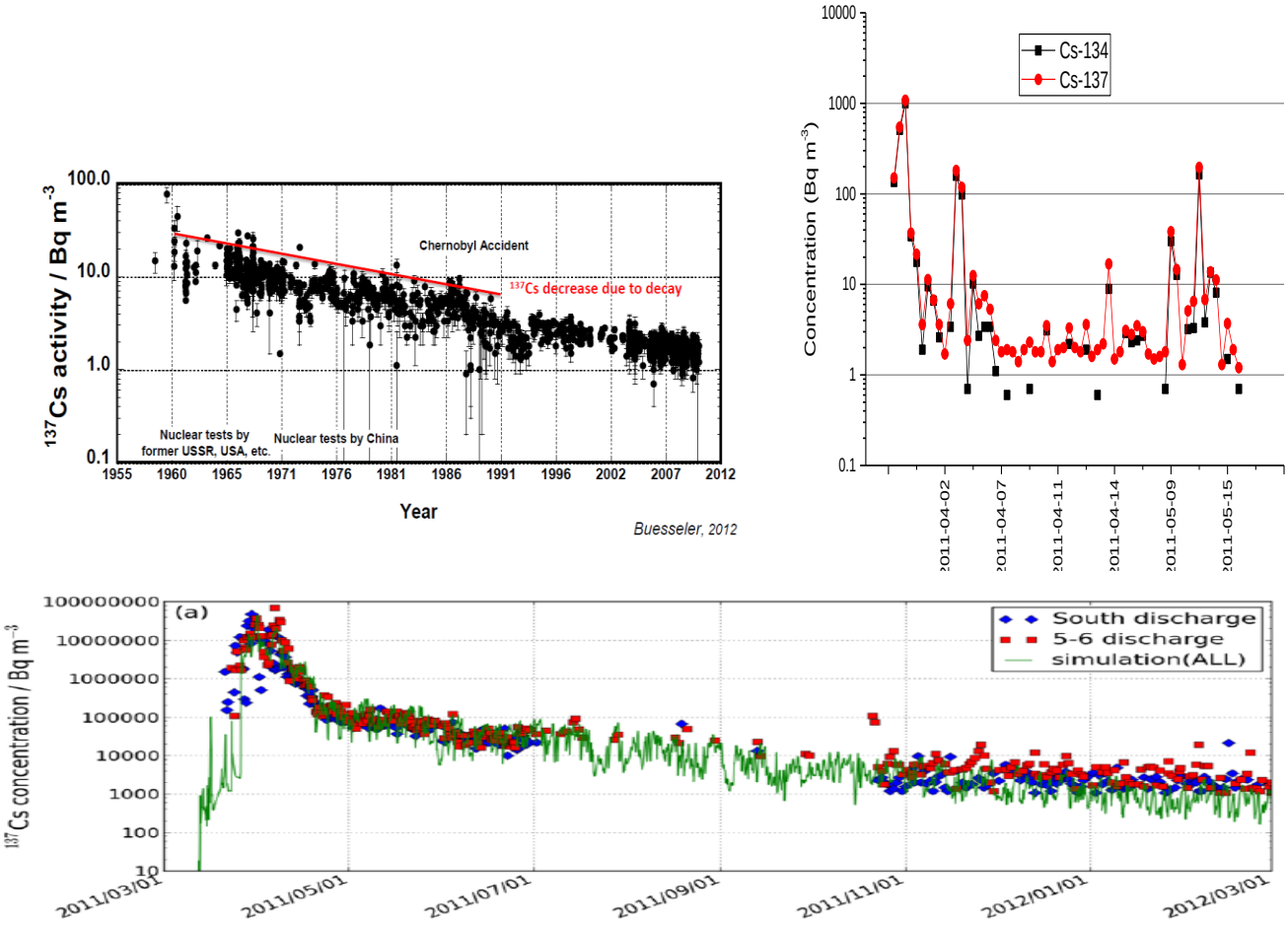

Fig.3 Cs-137 activity concentration in western North Pacific Ocean
(Left) Long term trend of Cs-137 in surface water in the western North Pacific Ocean: Pre-Fukushima (Buesseler, 2012)
(Right) Aril/May 2011, After Fukushima accident (Aoyama, 2013)
(Bottom) Measured Cs-137 activities at the north and south discharge canals near the 1F NPP (1 year) (Tsumune, 2013)

In the western North Pacific Ocean, ^{137}Cs activity in surface water was 10–100 Bq m^{-3} in the late 1950s and the 1960s, then it decreased gradually and the ^{137}Cs activity in surface water decreased to around a few Bq m^{-3} as shown in Fig. 1. In 2000s just before the FNPP1 accident, the ^{137}Cs activity in surface water ranged from 1 to 2 Bq m^{-3}. The maximum concentration of Cs-134 and Cs-137 measured at North Pacific Ocean were 1,000 and 1,080 at April 1, 2011. (Aoyama et al., 2013)

The ^{137}Cs activity ranged from around 1 to 1,000 Bq m^{-3} with activity ratios of $^{134}Cs/^{137}Cs$ close to 1 which is a signature of radiocaesium originated from the FNPP1 accident. At east of the International Date Line north of 40° N in the Pacific Ocean in April 2011, ^{134}Cs activity in the surface water ranged from 2 to 12 Bq m^{-3}. (Aoyama et al., 2013)

Fig.4 Cs-137 activity in the Pacific Ocean in the 2000s (unit : Bq m^{-3}) (Aoyama, 2013)

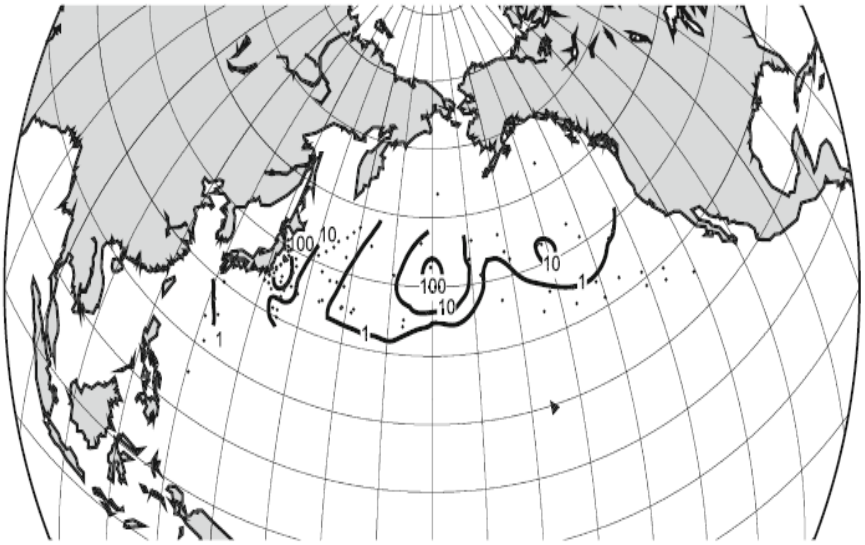

Fig.5 Cs-134 activity in the North Pacific Ocean after the FNPP1 Accident in Aprial/May 2011 (unit : Bq m^{-3})

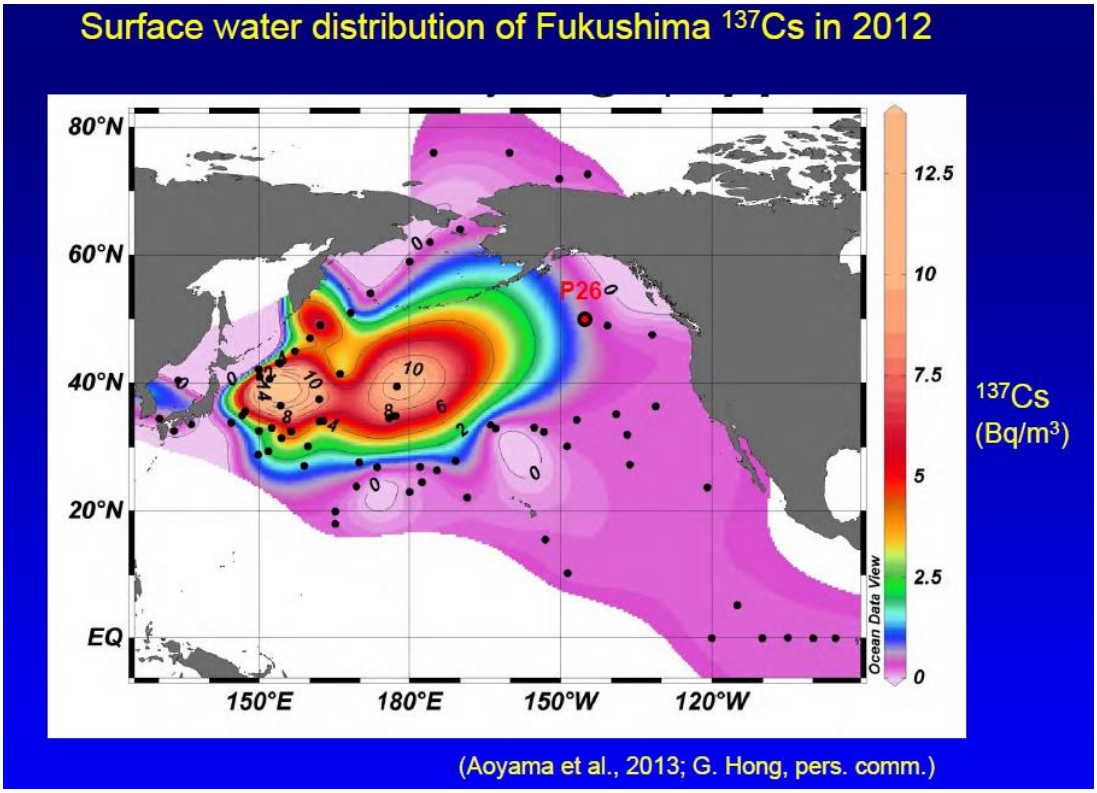

Fig.6 Surface water distribution of Fukushima Cs-137 in 2012 (Aoyama et al., 2013)

V. CONCLUSIONS

Historical source term researches are reviewed first. There also has been efforts to make tools which can simulate severe accidents which can be happen in nuclear power plants realistically. Comparisons are made among three real source term release accidents occurred historically in nuclear power plants. Insights obtained during this study will be helpful to guiding

correct directions of such a severe accident simulation tool development. Harmony between tool development and analysis work will be needed in design of nuclear power plant and planning of emergency response preparedness. Integrity of reactor vessel and containment in TMI-2 accident give results in very small amount of volatile radionuclide are released to the environment. Graphite fire in Chernobyl accident and long-term station blackout (SBO) in Fukushima accident give results in much different kind and magnitude of radionuclides released to the environment. Accurate assessment of radioactivity release to environment is very important in subsequent human health impact assessment. An agreement between the reverse/inverse method with monitoring data sets and the accident progression method using severe accident analysis codes will results in accurate assessment of radioactivity release to environment. There are still much debates on the accurate amount of land deposition and oceanic input from the Fukushima accident. Cesium-137 moves from west North Pacific to east North Pacific gradually. Cesium-134 decays out during the movement.

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