

**Graduate School of Frontier Science, The University of Tokyo**

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**Master's Thesis**

**Spatial and Temporal Distribution of Radio-Cesium in  
the Sediment of Tokyo Bay and Surrounding Rivers.**

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## Abstract

After the Fukushima Daiichi Nuclear Power Plant (FDNPP) incident on March 11, 2011, a large amount of radioisotope material was released to the environment. The contamination affected not only the areas around the reactor itself, but also a relatively large area of eastern mainland of Japan (Oura and Ebihara, 2012). It was an opportunity to use the radio-caesium associated sediment as a tracer in Tokyo Bay and connecting Arakawa, Edogawa and Sumidagawa Rivers for studying the contaminated sediment distribution and their fate. Surface sediment samples were taken for the distribution pattern and core samples were taken to observe the vertical profile for seasonal variation of diffusion and sedimentation. Fine sediments are mostly assumed for the adsorption of radio-caesium though there are some contradictions; Particle size effect on the adsorption of radio-caesium was observed. Suspended solid (SS) concentration and water discharge was observed and calculated to obtain the relationship between land use and SS flux and also impact of water quality parameters of salinity and turbidity on the distribution of radio-caesium was observed in this study. The results showed that the sediment transport in Arakawa River was relatively larger due to river topography and water discharge pattern. However in Edogawa River, it was smaller and it usually happened only after large rainfall event and typhoon effect. In Arakawa River, sedimentation was dominant in all seasons ( $R^2$  value was 0.698 for sedimentation whereas  $R^2$  value was 0.256 for diffusion). In Edogawa River, diffusion was dominant in summer of wet season ( $R^2$  value was 0.964 for diffusion whereas  $R^2$  value was 0.654 for sedimentation). In Sumidagawa River, sedimentation and diffusion were almost same level. It was observed that, total caesium concentration ( $^{134}\text{Cs}+^{137}\text{Cs}$ ) was high where the silt contents were high for example, in Arakawa River the silt content was highest 76% in station-7, the total caesium concentration ( $^{134}\text{Cs}+^{137}\text{Cs}$ ) was highest 449 Bq/kg. Similarly in Edogawa River the silt content was highest 79% in station-15, the total caesium concentration ( $^{134}\text{Cs}+^{137}\text{Cs}$ ) was highest 1856 Bq/kg. Most of the contaminated sediment moved to the Tokyo Bay from Arakawa River. In Edogawa and Sumidagawa River still contaminated sediment remained in the upstream region. Moreover the supply of contaminated sediment was limited from upper most stream region and most of the contaminated sediment buried in between 4-18 cm depth from the sediment bed in those three rivers which was enough to prevent further exposure of the contaminated sediment to Tokyo Bay.

Key Words: Radio-caesium, Distribution, Vertical profile, Suspended Solid (SS), Diffusion, Sedimentation.

## **Declaration**

This thesis is a presentation of my original research work. Wherever contributions of others are involved, every effort is made to indicate them clearly, with due reference to the literature, and acknowledgment of colleagues and teachers.

Signature

Name : A.T.M. TOUFIQ MAHMUD

Date : 2014/01/27

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# Chapter 1: Introduction

## 1.1 Background

Tokyo Bay is a very important water body in Southern Kanto region of Japan. The population of Tokyo Metropolitan area is about 13.19 million as of 2011 (Tokyo Metropolitan Government Website). Aside from being commercially important, Tokyo Bay has also great importance in terms of environmental perspective. Many rivers drain into the Tokyo Bay. The Arakawa, Tamagawa and Sumidagawa rivers drain into Tokyo Bay near Tokyo. The Edogawa River drains into Tokyo Bay in between Tokyo and Chiba Prefecture. After the Fukushima incident in March, 2011, the radio-caesium materials were spread out in atmosphere and in land-sediment. Radio-caesium can be used as a good tracer to determine the sediment age (i.e., distinguish the older sediment from newer sediment). Further this radio-caesium associated sediment can be used for the sediment transport indicator in Tokyo Bay and connected rivers.

Rivers play an important role in erosion as they are the primary carrier of transferring erosion products to the ocean. Understanding sediment transport in rivers and estuaries and their amount will improve the perception of many basic processes such as biogeochemical cycling of pollutants and nutrients, soil formation and their erosion (Allen, 2008).

On the shorter time scale, humans may act as geomorphic agents by constructing dams and reservoirs, and changing land use by deforestation and mining (Hooke, 2000; Syvitski et al., 2005; Wilkinson and McElroy, 2007).

Sediment tracing technology was begun in the early 1960s with research on sediment patterns, which include erosional, depositional rates and mass sediment accumulation. The developing field of sediment-tracing technology provides a method to measure sediment erosion, estimate sediment age and calculate sediment deposition and accumulation rates in marine, fluvial, riverine, and lacustrine environments (Corcoran M.K. and Kelley J.R, 2006).

Radionuclide is added to the sediment by the deposition of particulate with the associated radio nuclides or in some circumstances, by movements of radio nuclide through the interface into the sediments.

According to D.E Walling and Q. He (1993), the fallout radionuclide tracers, and more particularly cesium-137 ( $^{137}\text{Cs}$ ), can provide a valuable means of assembling detailed information on average rates and patterns of floodplain sedimentations over the past 35 years. In essence, the basis for using  $^{137}\text{Cs}$  for this purpose is that radio-cesium is rapidly and strongly adsorbed by the fine fractions of soil and sediments. Accumulation in flood plain sediments as a result of inputs from two primary sources one is direct deposition from the fallout and another is deposition of sediment associated  $^{137}\text{Cs}$  during the process of sediment accretion.

The sediment associated radio-cesium can be mobilized from the upstream by erosion and transported to downstream as an integral part of the suspended sediment load of the river. In this study Suspended Sediment Concentration (SSC) examined to make relationship with the water level and to estimate the suspended sediment flux as well as radio-cesium flux through the SSC. Determining the suspended sediment source also a feature of this study.

According to Oura and Ebihara, 2012, after the Fukushima Daiichi Nuclear Power Plant (FDNPP) incident on Friday 11 March 2011, a large amount of radioisotope material was released to the environment. The contamination affected not only the areas around the reactor itself, but also a relatively large area of eastern mainland of Japan with some specific areas became heavily contaminated.

In order to understand the distribution of the contaminated sediment and their fate, the  $^{137}\text{Cs}$  and  $^{134}\text{Cs}$  associated sediment as tracers, in Tokyo Bay and its three connecting rivers. Through this study, the spatial and temporal distribution of radio-cesium in the sediment of Tokyo Bay and Arakawa River, Edogawa River and Sumidagawa River which are mixed with Tokyo Bay will be investigated as well as transportation mechanisms and fate of the contaminated sediment.

The vertical profile of the radio-cesium concentration result from the core samples can be utilized to estimate the total mass of contaminated sediment and total mass of radio-cesium in the study area. Moreover, the vertical profile data can be used to calculate the diffusion coefficient and the sedimentation speed in the three studied rivers.

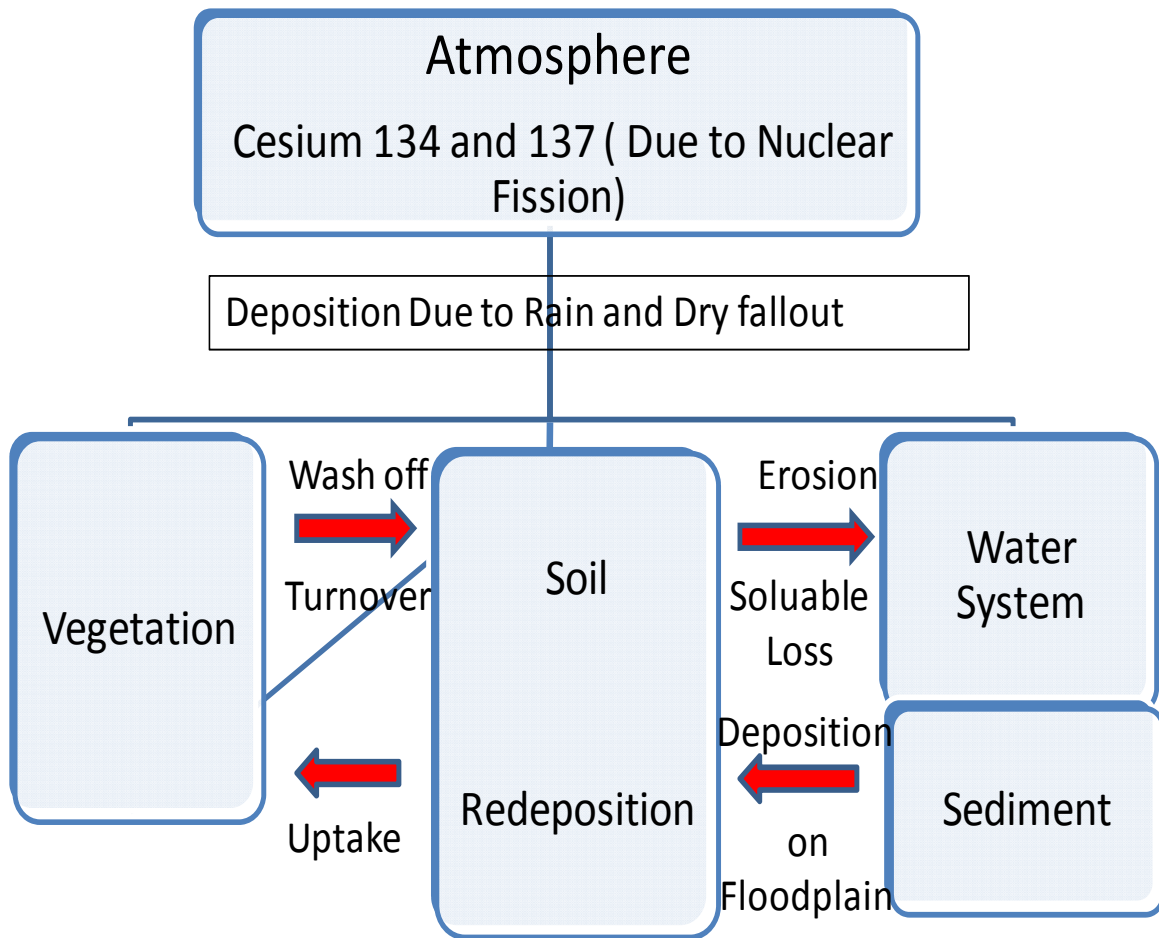


Figure 1 : Diagram of the Cesium Cycle in the Landscape, adopted from Ritchie and McHenry (1990).

Relationship of the distribution of radio-cesium with the sediment particle size, water quality parameters as salinity and turbidity were also has been taken in consideration in this study. Besides, the relationship of water level and suspended sediment concentration (SSC) in Arakawa and Edogawa River were also observed. After each large rainfall event the sample were taken and the concentration change of radio-cesium in the SSC with time was observed.

The water level data of Arakawa and Edogawa River is available from the River department of Japan. By using the water level and Discharge curve that is HQ curve, it was possible to calculate the water discharge and thus radio-cesium discharge through suspended load in those rivers.

## **1.2 Literature Review**

### **1.2.1 Environmental Radionuclides**

Natural and artificial radionuclides, collectively referred to as environmental radionuclides, are those radionuclides that are commonly occurring and widely distributed in the environment or on the landscape and are measurable (Walling 2003). Environmental radionuclides are used extensively to measure sediment movement and accumulation in closed systems or in shallow quiescent environment, such as lakes, estuaries, shallow marine, and river floodplains.  $^{134}\text{Cs}$  and  $^{137}\text{Cs}$  is the product of nuclear testing or accident and therefore they are artificial radionuclide and in combination they also known as radio-caesium.

### **1.2.2 Radio-caesium as a Tracer**

The basis for using  $^{134}\text{Cs}$  and  $^{137}\text{Cs}$  for deriving sedimentation rates is that it is rapidly and strongly adsorbed by soil, preferentially by clay-sized particles (Tamura 1964). The  $^{137}\text{Cs}$  presence in soils is from direct deposition from the atmosphere, from vegetation, turnover from vegetation, re-deposition of eroded particles, and deposition from water on floodplains (Walling and He 1993).

The presence of  $^{134}\text{Cs}$  and  $^{137}\text{Cs}$  in water is from direct deposition on water surfaces and input of  $^{134}\text{Cs}$  and  $^{137}\text{Cs}$  adsorbed on eroded soil particles. The  $^{134}\text{Cs}$  and  $^{137}\text{Cs}$  fallout deposited on vegetation may be adsorbed or absorbed. Most adsorbed  $^{134}\text{Cs}$  and  $^{137}\text{Cs}$  is washed from vegetation and moved to the soil. Absorbed  $^{134}\text{Cs}$  and  $^{137}\text{Cs}$  is released to soils when vegetations dies and decays (Ritchie and McHenry 1990). The amount of  $^{134}\text{Cs}$  and  $^{137}\text{Cs}$  contributing to aquatic or terrestrial environments is also dependent on precipitation rates (Mitchell et al. 1983).

River floodplains represent important sediment sinks for sorting suspended sediment and contaminants from upstream. The  $^{134}\text{Cs}$  and  $^{137}\text{Cs}$  radioisotopes are an established sediment tracer used to study floodplain deposits and sediment sources (Owens et al. 1999; Walling and He. 1997).

The  $^{137}\text{Cs}$  radioisotope, with a half-life of 30.2 years (He and Walling 1996), possesses specific properties, which contribute to using it as a viable sediment tracer. According to Ritchie and McHenry (1990), these properties are as follows:

- a. There are no natural sources of  $^{137}\text{Cs}$ .
- b. Fallout is strongly related to local precipitation patterns with total fallout varying linearly with rainfall within latitudinal zones.
- c. Temporal and spatial distribution of radioactive fallout on the earth's surface has been extensively measured and documented.
- d.  $^{137}\text{Cs}$  is strongly adsorbed on clay and organic particles and is essentially non exchangeable.
- e. Adsorption on soils and sediment is rapid with distribution in undisturbed soil profiles showing an exponential decrease with soil depth.

Ritchie and McHenry (1990) sum up the considerations for using  $^{137}\text{Cs}$  as a sediment tracer as follows:

- $^{137}\text{Cs}$  is preferentially adsorbed on clay particles. In areas where sand is prevalent, there will be a decrease in the amount of  $^{137}\text{Cs}$  present.
- There is a time lag between the time of atmospheric deposition of  $^{137}\text{Cs}$  and the time of deposition in the soil.
- $^{137}\text{Cs}$  is not useful in sediments that have been disturbed by bioturbation or tillage or in areas where the sedimentation rate is  $< 1$  cm per year.

### **1.2.3 Particle size effect on radio-caesium adsorption**

In most environments, these fallout radionuclides are rapidly and strongly adsorbed by soils and sediments and an understanding of their distribution between different size fractions is important for their use as tracers (Walling and He, 1996).

Walling and He (1996) reported some results of sediment particle size and radio-caesium adsorption which are contradictory. For example they noted, in studying the distribution of  $^{238}\text{Pu}$ ,  $^{239,240}\text{Pu}$  and  $^{137}\text{Cs}$  in various particle size fractions of Lake Michigan sediments, Albert and Muller (1979) found that concentrations of these radionuclides were almost constant across a range of particle size classes. Conversely, Walling and Woodward (1992) found that for soils collected from the Jackmoor Brook catchment, Devon, UK, the concentrations of  $^{137}\text{Cs}$  in the



finer fractions ( $< 8 \mu\text{m}$ ) were several times higher than those in the coarser fractions. Similarly, Livens and Baxter (1988b) reported a study of the distribution of various radionuclides in different size fractions of soils, in which soil samples were separated into several size fractions ranging from  $< 2 \mu\text{m}$  to  $250\text{-}2000 \mu\text{m}$ , and concentrations of  $^{137}\text{Cs}$  in the fractions were shown to decrease with an increase in particle size.

In most cases, radionuclide activities in suspended sediment will be greater than those in equivalent source materials, due to the selective mobilization of fines (Walling and He, 1996).

#### **1.2.4 Other Related Researches**

One research group is currently engaged in ongoing research on the sedimentary environment in the Mekong River delta to assess the deltaic environment and its conservation status (Saito et al., 2012). During a survey of the deltaic sediment in October 2011, they conducted a geomorphologic study and also took core samples. It was surprising to detect Cs isotopes released from the FDNPP. Next year they made the survey again. The study focuses on the Cs isotopes in the core in order to study the seasonal variations of this deltaic environment. Deltaic sediments of the Mekong River delta sampled from a tidal beach in Vietnam during the wet season (late October 2011) showed strong  $^{137}\text{Cs}$  and  $^{134}\text{Cs}$  activities reflecting radionuclides released from the Fukushima Dai-ichi nuclear power plant (FDNPP). However samples from the same site taken about three months later during the dry season (early February 2012) showed weak activities. This finding indicates that soil from the Mekong drainage basin was deposited along the delta front in the wet season and then removed in the dry season during the winter monsoon.

Zheng et al., (2012) made a study on the distribution of Pu isotopes in surface sediments in the Pacific 30 km off Fukushima after the Fukushima DNPP accident. Activities of  $^{239+240}\text{Pu}$  and  $^{241}\text{Pu}$ , and atom ratios of  $^{240}\text{Pu}/^{239}\text{Pu}$  in surface sediments collected in July- August, 2011 were analyzed to make a quick assessment on the environmental impact of those possible Pu contaminations. The Pu contamination from the Fukushima DNPP accident was not observed in marine sediments outside the 30 km zone.

### **1.3 Research Objectives**

From the background of this study, the research objectives can be summarized as follows

- To clarify the transportation mechanisms and fate of the contaminated sediment. Whereas the most of the contaminated sediment remained in the land area or moved to river bed or Tokyo Bay basin.
- Fine sediments are mostly attractive for the adsorption of radio-caesium though there are some contradictions; to clarify the impact of the particle size effect on the adsorption of radio-caesium.
- To observe the seasonal variation of diffusion and sedimentation in vertical direction by using the vertical profile results of the core samples.
- To know the relationship between land use and sediment flux, impact of water quality parameters of salinity and turbidity on the distribution of radio-caesium.

## Chapter 2: Study Area

### 2.1 Tokyo Bay

Tokyo Bay is located in the Southern of Kanto region of Japan and spans the coasts of Tokyo, Kanagawa prefecture and Chiba prefecture. The Tokyo Bay covers the areas about 922 square kilometers from the Kape Kannon of the West of Miura peninsula to Kape Futtsu on the East of Bōssō Peninsula but if the Uraga Channel through which Tokyo Bay is connected with the Pacific Ocean is taken in consideration then the area covers about 1320 square kilometers. The average depth of Tokyo Bay is about 40 meters and it increases towards the Pacific Ocean. Many rivers drain into the Tokyo Bay. The Arakawa, Tamagawa and Sumidagawa rivers drain into Tokyo Bay near Tokyo. The Edogawa River drains into Tokyo Bay in between Tokyo and Chiba Prefecture. The Obitsu and Yōrō rivers drain into Tokyo Bay in Chiba Prefecture. Sediments samples are collected from the Tokyo Bay to know the Radio-caesium distribution. From Tokyo Bay only surface sediment were collected in five stations (Table-1 and Figure: 2). The samples were collected almost every month from Tokyo Bay.

**Table 1 : Sampling locations in Tokyo Bay**

<b>Study Area</b>	<b>Station No</b>	<b>Locations</b>
Tokyo Bay	Station-8	N 35°33' 16", E 139°54' 20"
	Station-13	N35°29' 18" , E 139°54' 24"
	Station-97	N35°29' 16" , E 139°49' 07"
	Station-98	N35°33' 59" , E139°51' 21"
	Station-99	N35°37' 45", E140°00' 31"

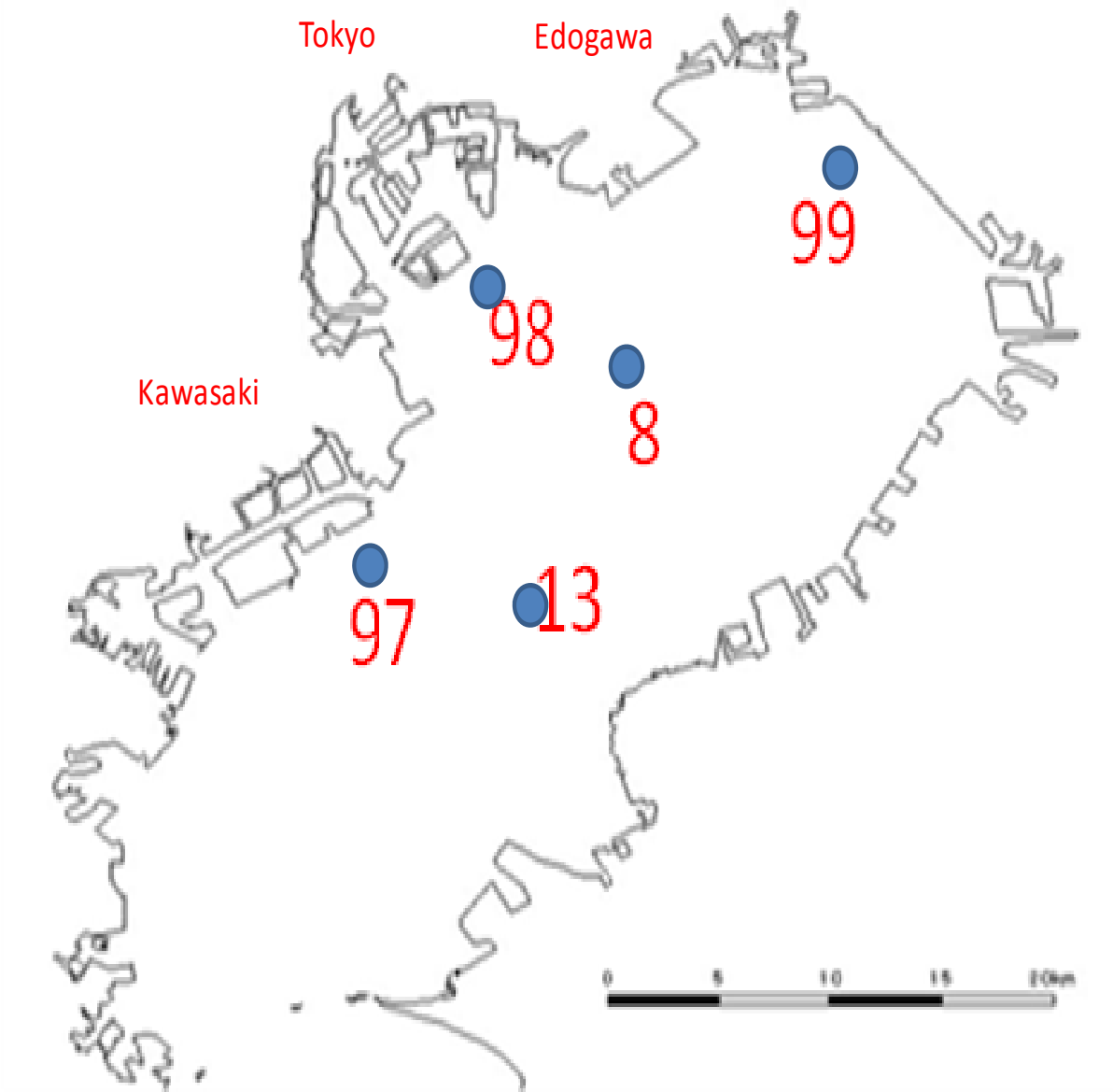


Figure 2 : Sampling sites in the Tokyo Bay

## 2.2 Arakawa River

The Arakawa River is one of the main rivers in the southern Kanto region, flowing through Tokyo. It originates in Mount Kobushigatake (2,475 m.a.s.l) in the Kanto Mountains and flows to Tokyo Bay. The length of its mainstream is 173 km. and its drainage basin covers 2,940 km<sup>2</sup> in Saitama prefecture and Tokyo Metropolis. There are seven sampling sites in Arakawa River (Table-2 and Figure: 3). The station-6 is considered as the river mouth where the river empties into the Tokyo Bay. Surface sediment samples were taken from all seven stations in the Arakawa River for measuring the spatial and temporal distribution of radio-caesium and from the station 4 and station 7 the core samples were taken to get the vertical profile of caesium distribution in the vertical direction of the sediment.

Table 2 : Sampling locations in Arakawa River

Study Area	Station No	Locations
Arakawa River	Station-1	N 35°43' 05.7",E 139°50' 34.4"
	Station-2	N 35°42' 19.9",E 139°51' 16.5"
	Station-3	N 35°41' 15.3",E 139°51' 13.6"
	Station-4	N 35°40' 18.6",E 139°50' 47.7"
	Station-5	N 35°39' 20.1",E 139°50' 40.1"
	Station-6	N 35°38' 26.3",E 139°50' 36.6"
	Station-7	N 35°36' 50.4",E 139°50' 24.4"

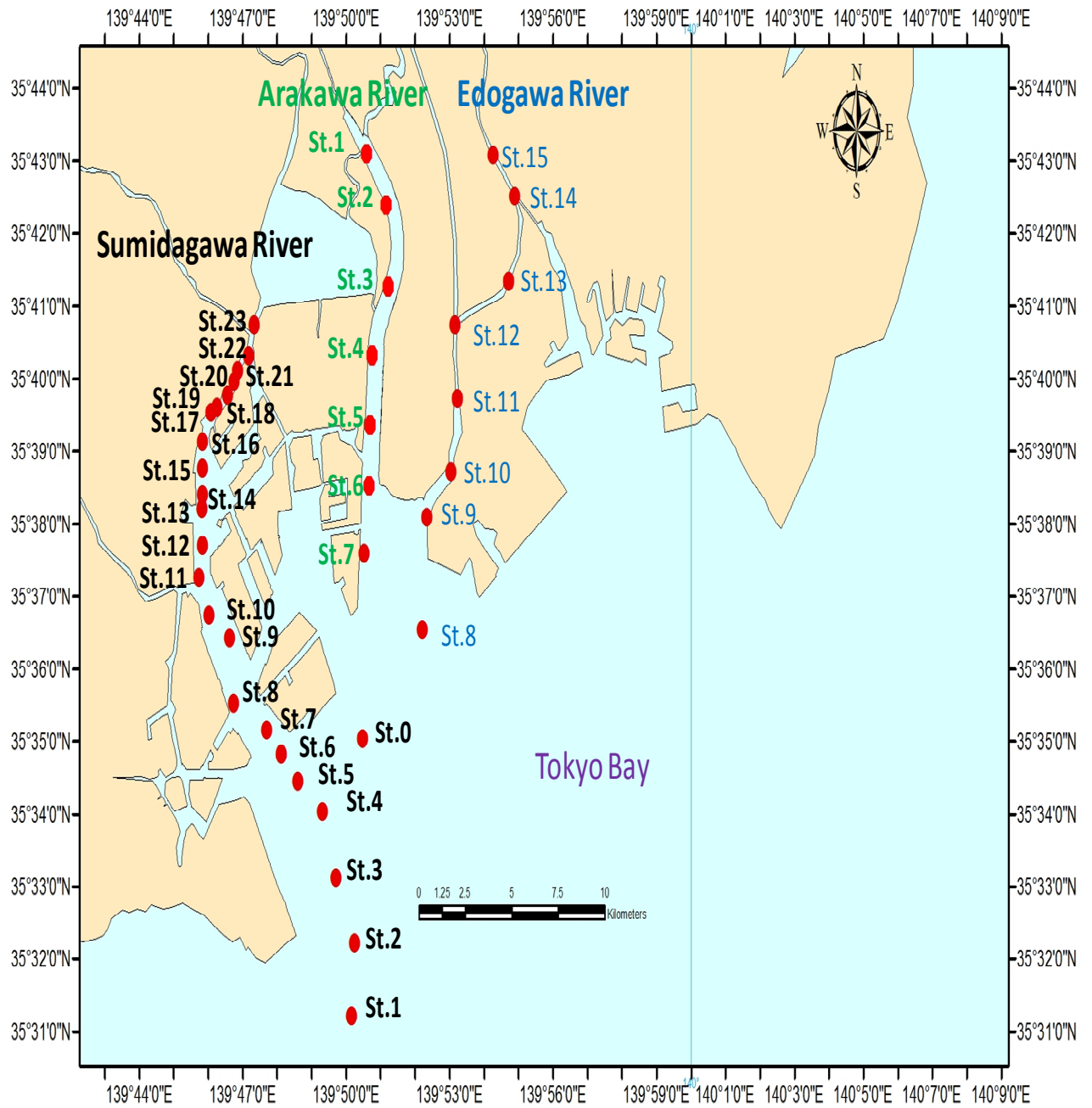


Figure 3 : Arakawa, Edogawa and Sumidagawa River sampling sites

### 2.3 Edogawa River

The Edogawa River is another river that flows through Tokyo with a 200 km<sup>2</sup> drainage basin and a 55 km. mainstream, the Edogawa river flows from the eastern part of Tokyo into Tokyo Bay. In the past, this river was a part of the Tone River, however, in the 17<sup>th</sup> century; diversion works made the river independent like the present condition. On the other hand, channels connecting the Arakawa and Edogawa rivers, as well as the flat topography in their lower reaches made the exact separation of the two basins impossible. There are ten sampling sites in Edogawa River (Table-3 and Figure: 3). The station 9 is considered as the river mouth where the river empties into the Tokyo Bay. Surface sediment samples were taken from all ten stations in the Edogawa River for measuring the spatial and temporal distribution of radio-caesium and from the station 9 and station 14 the core samples were taken to get the vertical profile of caesium distribution in the vertical direction of the sediment.

**Table 3 : Sampling Locations in Edogawa River**

Study Area	Station No	Locations
Edogawa River	Station-8	N 35°36' 34.1",E 139°52' 12.8"
	Station-9	N 35°38' 05.5",E 139°52' 20.4"
	Station-10	N 35°38' 43.2",E 139°53' 01.8"
	Station-11	N 35°39' 41.8",E 139°53' 14"
	Station-12	N 35°40' 44.1",E 139°53' 10.5"
	Station-13	N 35°41' 21.3",E 139°54' 43.5"
	Station-14	N 35°42' 30.5",E 139°54' 54.4"
	Station-15	N 35°43' 06.6",E 139°54' 15.6"
	Station-16	N 35°37' 35.9",E 139°50' 32.2"
	Station-17	N 35°41' 54.9",E 139°55' 07.2"

### **2.3 Sumidagawa River**

Sumidagawa River is another river which flows through Tokyo. It branches from the Arakawa River at Iwabuchi and empties into Tokyo Bay. Its tributaries include the Kanda and Shakujii rivers. River Length is 23.5 km, drainage area 690 km<sup>2</sup>. There are twenty three sampling sites in Sumidagawa River (Table-4 and Figure-3). This is an estuary areas and station 8 can be considered as the river mouth where the river empties into the Tokyo Bay. Surface sediment samples were taken from all twenty four stations in the Edogawa River for measuring the spatial and temporal distribution of radio-caesium and from the station 17 and station 20 the core samples were taken to get the vertical profile of caesium distribution in the vertical direction of the sediment.



Table 4 : Sampling Locations in the Sumidagawa River Estuary

Study Area	Station No	Locations
Sumidagawa River	Station-0	N 35°35' 02.3",E 139°50' 28.9"
	Station-1	N 35°31' 12.8",E 139°50' 07.8"
	Station-2	N 35°32' 13.9",E 139°50' 14.7"
	Station-3	N 35°33' 07.4",E 139°49' 42.8"
	Station-4	N 35°34' 02.1",E 139°49' 18.8"
	Station-5	N 35°34' 27.4",E 139°48' 36.3"
	Station-6	N 35°34' 49.7",E 139°48' 06.9"
	Station-7	N 35°35' 10.8",E 139°47' 42.4"
	Station-8	N 35°35' 31.1",E 139°46' 45.9"
	Station-9	N 35°36' 26.5",E 139°46' 37.7"
	Station-10	N 35°36' 44.2",E 139°46' 00.1"
	Station-11	N 35°37' 14.5",E 139°45' 44.4"
	Station-12	N 35°37' 41.7",E 139°45' 49.8"
	Station-13	N 35°38' 12.4",E 139°45' 49.5"
	Station-14	N 35°38' 23.8",E 139°45' 49.7"
	Station-15	N 35°38' 45.8",E 139°45' 50.1"
	Station-16	N 35°39' 08.2",E 139°45' 50.5"
	Station-17	N 35°39' 30.7",E 139°46' 04.9"
	Station-18	N 35°39' 36.4",E 139°46' 17.3"
	Station-19	N 35°39' 45.9",E 139°46' 33.4"
	Station-20	N 35°39' 56.3",E 139°46' 45.4"
	Station-21	N 35°40' 06.5",E 139°46' 52.3"
	Station-22	N 35°40' 20.5",E 139°47' 10.3"
	Station-23	N 35°40' 42.7",E 139°47' 20.4"

## 2.4 The land Use and Cover of Arakawa, Edogawa and Sumidagawa Rivers

The Arakawa River is one of the main rivers in the southern Kanto region, flowing through Tokyo. It originates in Mount Kobushi (2,475 m.a.s.l) in the Kanto Mountains and flows to Tokyo Bay. The length of its mainstream is 173 km. and its drainage basin covers 2,940 km<sup>2</sup> in Saitama prefecture and Tokyo Metropolis.

The Edogawa River is another river that flows through Tokyo with a 200 km<sup>2</sup> drainage basin and a 55 km. mainstream, the Edogawa river flows from the eastern part of Tokyo into Tokyo Bay. In the past, this river was a part of the Tone River, however, in the 17<sup>th</sup> century; diversion works made the river independent like the present condition. On the other hand, channels connecting the Arakawa and Edogawa rivers, as well as the flat topography in their lower reaches made the exact separation of the two basins impossible.

The land use or cover in the Arakawa and Edogawa River basin is characterized by forested areas in the upper reach with some agricultural areas along the rivers. In the middle reach, agriculture is dominant in case of land use. In the 17<sup>th</sup> century, the lower catchment was designated as an agricultural zone to supply food to Tokyo. However, in the past 50 years, rapid urban development of Tokyo resulted in the conversion of this zone into a residential and industrial one. According to River Department (2000a), the Arakawa and Edogawa river basin has the highest population density that reaches 3,164 persons/km<sup>2</sup> and 17,000 person/km<sup>2</sup> respectively. (Master Thesis, "Land Use/Cover and Surface Water Quality at Multiple Spatial Scales in the Kanto Region"- Andy Darmawan, p-13-14)

Sumidagawa is another river which flows through Tokyo. It branches from the Arakawa River at Iwabuchi and flows into Bay. Its tributaries include the Kanda and Shakujii rivers.

The river which is now known as the "Sumida River" was previously the path of the Arakawa, however towards the end of the Meiji era, work was carried out to divert the main flow of the Arakawa to prevent flooding. River Length is 23.5 km, drainage area 690 km<sup>2</sup>, Population about 3 million. Tokyo Metropolitan Government operates or maintains this river. The entire river is a tidal river, which is influenced by tides (<http://ja.wikipedia.org/wiki/sumidagawa>).

As it is known that, the land use pattern in any river basin has the impact on the sediment run off or water runoff towards the receiving water bodies. Thus the water shade pattern information of

Arakawa, Edogawa and Sumidagawa river basin has been showed (Figure-4). Edogawa River's water shade is a sub water shade of Tone River water shade and Sumidagawa River's water shade is a sub water shade of Arakawa river water shade. Table-5 included the water shade land use information of Arakawa River and Table-6 included the water shade information of Arakawa, Edogawa and Sumidagawa Rivers.



Figure 4 : River catchment map of Arakawa, Tonegawa River and Prepared for World Water Assessment Program (WWAP) by AFDEC, 2002.

**Table 5 : Arakawa River Water Shade Information (1985).**

<b>Land Use Type</b>	<b>Percentages (%)</b>	<b>Main Cities</b>	<b>Population (million)</b>
Forest	48.2	Tokyo, Omiya, Urawa, Kawagoe, Chichibu.	About 9 million
Paddy Field	5.1		
Agriculture	6.5		
Water surface	4		
Urban	26.5		
Others	9.7		

**Table 6: Information about Arakawa, Edogawa and Sumidagawa River**

<b>River Name</b>	<b>Catchment (km<sup>2</sup>)</b>	<b>Stream length (km)</b>	<b>Related Prefecture</b>	<b>Population (Million)</b>	<b>Population Density (person/km<sup>2</sup>)</b>
Arakawa	2940	173	Saitama prefecture and Tokyo Metropolis	9.3	3,164
Edogawa	200	55	Ibaraki, Chiba, Saitama and Eastern Part of Tokyo	3.4	17,000
Sumidagawa	690	23.5	Tokyo Metropolis	3	

## **Chapter 3: Materials and Methods**

### **3.1 Sample collection**

Samples were collected from all the sampling sites discussed in chapter 2, study area section. For the surface sediment, from water- sediment interface 0-2 cm depth sediment was collected by using the standard surface sediment sampler. And for the core samples, from water- sediment interface up to 38 cm depth's sediment was collected by using tube core samplers with diameter of 4 cm and having with cutting heads, 5 m rope, detachable grip 60 cm, extension rod 100 cm, stainless ball and rubber piston. There are 12 layers in each core sample, width of the each core layer was 2 cm up to layer 5 and after that the width of each layer was 4 cm Thus the total depth of the each core is 38 cm from water sediment interface.

Samples were collected from above stated five stations of Tokyo Bay almost every month from May 2012 to October 2013. From Tokyo Bay only surface sediment were collected. In case of other study sites in Arakawa, Edogawa and Sumidagawa rivers, both surface sediment and core samples were collected around three months interval.

The locations for core sampling in Arakawa River are station 7, located in the 3.59 km downstream from the river mouth and in station 4, located in the 3.25 km upstream area from the river mouth. In case of Edogawa River the locations for core sampling are station 9 which is considered as the river mouth and in station 14, located in the 9.81 km upstream area from the river mouth. In case of Sumidagawa River the core samples were taken from the station 17 and station 20 are located in the upstream area.

Moreover from the core sampling locations 4 liters water samples also collected each and every sampling time in order to get the suspended solid concentration (SSC) and along with Cesium concentration in the suspended solid. The exact timing of sampling in these rivers is June, 2012; September, 2012; November, 2012; March, 2013; July, 2013 and September, 2013. Thus these entire samples can reflect the temporal or seasonal variations as well as spatial distributions of the radio-caesium in the Tokyo Bay and all three rivers.

Besides, water samples of Edogawa River from Noda bridge point after each large rainfall event were collected to measure the suspended solid concentration and radio-caesium concentration and

thus the relationship of water level and the SSC (suspended solid concentration) was made to estimate the yearly total suspended solid flux and thus cesium flux. The schematic diagram of sample collection and handling has been shown in figure-5.

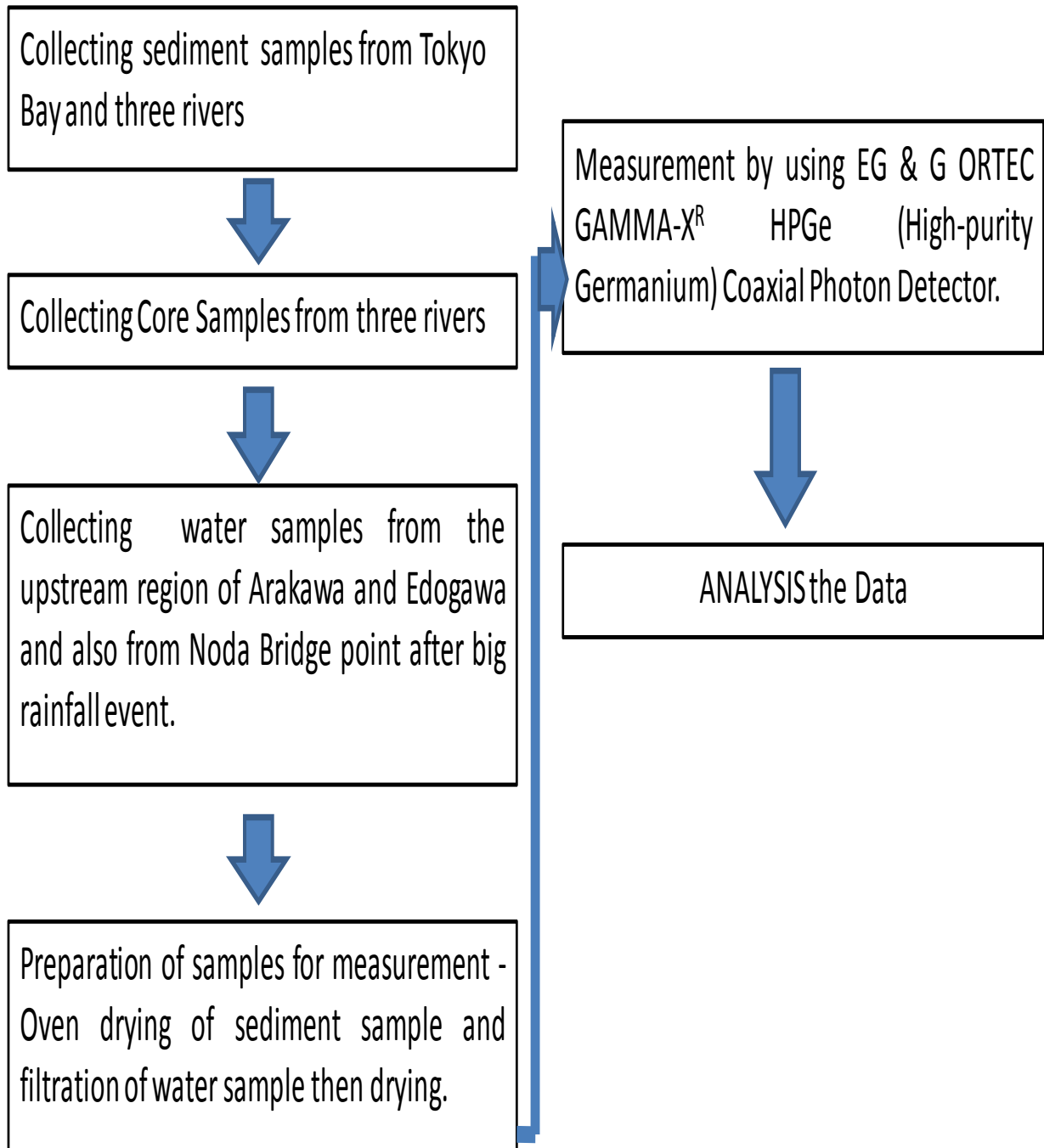


Figure 5 : Schematic Diagram of Sample Collection and Handling

### 3.2 Laboratory Procedure

The sediment samples were brought to the laboratory in wet condition and keep them in the refrigerator in cool condition. The sediment samples were then oven dried for 20-24 hours and then kept in the polyethylene bag (Ziploc) for radio cesium activity determination. The sediment sample weight varies from 5 to 180 gm. The radio cesium concentration was measured as a unit of Becquerel per kilogram (Bq/kg) in dry weight. The radio cesium were measured by using EG & G ORTEC GAMMA-X<sup>R</sup> HPGe (High-purity Germanium) Coaxial Photon Detector along with Seiko EG & G Spectrum Navigator as a software tool (Figure-6). The efficiencies of the detection system were calibrated using standard samples measured by IAEA. The samples were placed on the top of the detector sensor and counted for minimum 3600 s, providing a precision of c.  $\pm 5\%$  at 90% level of confidence for the  $\gamma$ -spectrometry measurements. The activity of <sup>137</sup>Cs and <sup>134</sup>Cs in the samples was obtained from the counts at the 662 keV and 605 keV peak respectively on the measured  $\gamma$ -spectrum.



Figure 6 : EG & G ORTEC GAMMA-XR HPGe (High-purity Germanium) Coaxial Photon Detector

The particle size of the sediment samples were analyzed by using a laser diffraction particle size analyzer (SALD-3000S) produced by Shimadzu Co. Japan, with the compatible software (Wing SALD-3000S) (Figure-7). The particles were mainly classified into three type clay, silt and sand in this study and their classification system followed as Wentworth Scale (Wentworth, 1922, Table-7). Particle size was expressed in micro-meter. In table-8 the Type and size has been shown.

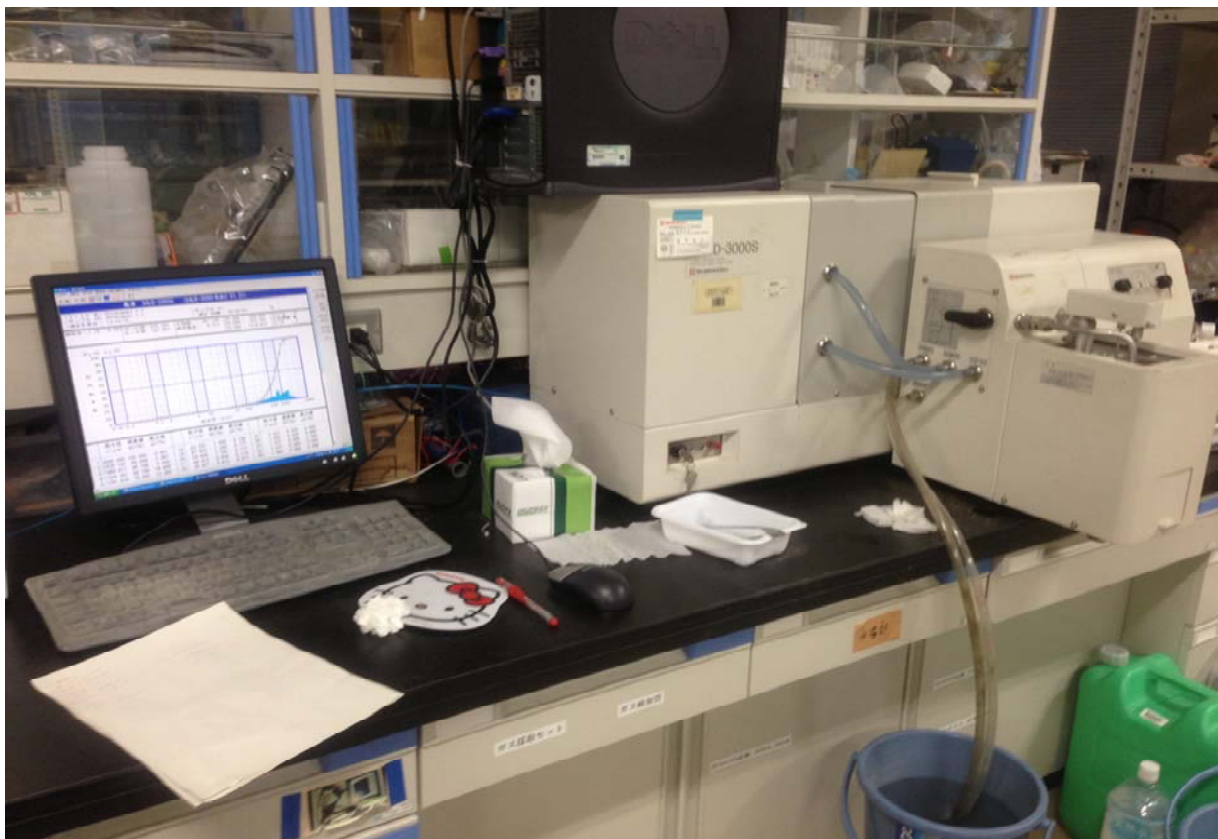


Figure 7 : SALD-3000S produced by Shimadzu Co. Japan, with the compatible software (Wing SALD-3000S).



Table 7 : Wentworth (1922) Grain Size Classification.

Millimeters (mm)	Micrometers ( $\mu\text{m}$ )	Phi ( $\phi$ )	Wentworth size class
4096		-12.0	Boulder
256		-8.0	Cobble
64		-6.0	Pebble
4		-2.0	Granule
2.00		-1.0	Very coarse sand
1.00		0.0	Coarse sand
1/2	500	1.0	Medium sand
1/4	250	2.0	Fine sand
1/8	125	3.0	Very fine sand
1/16	63	4.0	Coarse silt
1/32	31	5.0	Medium silt
1/64	15.6	6.0	Fine silt
1/128	7.8	7.0	Very fine silt
1/256	3.9	8.0	Clay
0.00006	0.06	14.0	

Table 8 : The Types and Sizes of Sediment.

Type of Particles	Size Range (mm)	Size Range ( $\mu\text{m}$ )
Sand	0.06-2.0	60-200
Silt	0.0039-0.06	3.9-60
Clay	<0.0039	<3.9

### 3.3 Numerical Modeling for Diffusion Coefficient and Advection

Governing equation for diffusion (equation-1)

$$\frac{\partial c}{\partial t} = k_z \frac{\partial^2 C}{\partial z^2} \dots\dots\dots 1$$

Analytical form of the Eq.1 expressed as Eq.2 including the decay constant for diffusion, only in vertical direction.

$$\frac{C_i^{t+1} - C_i^t}{\Delta t} = k_z \frac{C_{i-1}^t - 2C_i^t + 2C_{i+1}^t}{\Delta z^2} - \lambda \Delta t C^t \dots\dots\dots 2$$

where,  $K_z$ = Diffusion coefficient ( $m^2/$  month),  $C$ = Concentration of Cesium,  $\Delta t$ = change of Time,  $\Delta z$  = Depth and  $\lambda$  = Decay constant of  $^{137}\text{Cs}$  in Eq-1.

The physical decay of a radionuclide can be calculated simply. The activity concentration  $C(t)$  of a radionuclide at time  $t$  is given by equation-2.

$$C(t) = C(0) \exp(-\lambda_p t) \dots\dots\dots 3$$

where,  $C(0)$  is the activity concentration at time  $t = 0$  and  $\lambda_p$  is the radioactive decay constant. The radioactive constant is related to the physical half-life  $T_p$  given by equation-3

$$T_p = \frac{\ln 2}{\lambda_p} \dots\dots\dots 4$$

where,  $\ln 2$  (= 0.693) is the natural logarithm of 2.

Re-arranging the Eq-2 the Eq-5 can be get to calculate the diffusion coefficient  $K_z$ , the equation became as

$$k_z = \frac{\Delta z^2}{\Delta t} \frac{C_i^{t+1} - C_i^t}{C_{i-1}^t - 2C_i^t + 2C_{i+1}^t} \dots\dots\dots 5$$

Governing equation for Advection (Eq-6).

$$\frac{\partial c}{\partial t} = -\frac{\partial uc}{\partial z} - \lambda(t) \dots\dots\dots 6$$

$$\frac{\partial c}{\partial t} = -u \frac{\partial C}{\partial z} - \lambda(t) \dots\dots\dots 7$$

Analytical form of the Eq.7 expressed as Eq.8 for advection or sedimentation rate only in vertical direction.

$$\frac{C_i^{t+1} - C_i^t}{\Delta t} = -u \frac{C_i^t - C_{i-1}^t}{\Delta z} - \lambda(t) \dots\dots\dots 8$$

where, u= advection rate (m/ month), C= Concentration of Cesium, Δt= change of Time, Δz = Depth and λ = Decay constant of <sup>134</sup>Cs and <sup>137</sup>Cs in equation-4.

The vertical profile was 38 cm long and the Δz was 2 cm, Δt was 744 hours or 1 month, three layers were used to calculate the diffusion and it was assumed that no cesium added from the water column and the result of first sample which was taken in September, 2012 was the initial concentration and then compared with other time concentration.

### 3.4 Data Processing and analysis

Both the γ-spectrometry measurements and particle size measurements data were saved in connected computers with the detecting machine and the data were collected through USB and brought the laboratory. Further processing such as converting, calculation was done for the results of the experiment. Obtained result data were then analyzed by using the Software like Microsoft Office Excel-2007, MATLAB R2009a. The maps of the study area were produced by using the Arc GIS-10.

### 3.5 Suspended Solid (SS) Flux experiment

Water samples of Edogawa River from Noda bridge point after each large rainfall event were collected to measure the suspended solid concentration and radio-caesium concentration. The relationship of water level and the SSC (suspended solid concentration) was made to estimate the yearly total suspended solid flux and thus cesium flux. The relationship of cesium concentration and the SSC (suspended solid concentration) was also observed to evaluate the concentration of cesium in the source sediment. The water samples were filtered in the experiment room using suction filtration machine (Figure-8) and used the Whatman Glass Microfiber filters GF/F, diameter is 90 mm and the pore size of the filter is as standard of 0.45- $\mu\text{m}$ .



Figure 8 : Suction Filtration Machine

## Chapter 4: Results

### 4.1 Radio-Cesium ( $^{134}\text{Cs}$ and $^{137}\text{Cs}$ ) distribution in Tokyo Bay

In case of the distribution of radio-caesium in the sediment of Tokyo Bay, the result shows that the concentration in all five stations ranging from 5 to 160 Bq/kg and 0.71 to 80 Bq/kg in case of  $^{137}\text{Cs}$  and in case of  $^{134}\text{Cs}$  respectively. Figure-9 shows the spatial distribution of radio-caesium in Tokyo Bay of October, 2012 and Figure-10 shows the time series of caesium concentration from May 2012 to October 2013. The highest concentration in station-8 was 18.53 Bq/kg and 50.36 Bq/kg for  $^{134}\text{Cs}$  and  $^{137}\text{Cs}$  respectively in May, 2012. It was 0.71 Bq/kg and 14.51 Bq/kg for  $^{134}\text{Cs}$  and  $^{137}\text{Cs}$  respectively in October, 2012 and in last sampling that was in October, 2013 the concentration was 4.54 Bq/kg and 17.02 Bq/kg for  $^{134}\text{Cs}$  and  $^{137}\text{Cs}$  respectively. The highest concentration in station-13 was 21.85 Bq/kg and 66.82 Bq/kg  $^{134}\text{Cs}$  and  $^{137}\text{Cs}$  respectively also in May, 2012 and in last sampling that was in October, 2013 the concentration was 2.75 Bq/kg and 10.33 Bq/kg for  $^{134}\text{Cs}$  and  $^{137}\text{Cs}$  respectively. In case of station-97 the concentration observed in May, 2012 was 5.58 Bq/kg and 22.75 Bq/kg for  $^{134}\text{Cs}$  and  $^{137}\text{Cs}$  respectively and in last sampling that was in October, 2013 the concentration was 4.58 Bq/kg and 12.88 Bq/kg for  $^{134}\text{Cs}$  and  $^{137}\text{Cs}$  respectively. The highest concentration in station-98 was 43.54 Bq/kg and 106.52 Bq/kg for  $^{134}\text{Cs}$  and  $^{137}\text{Cs}$  respectively in October, 2012 and in last sampling that was in October, 2013 the concentration was 13.56 Bq/kg and 41.69 Bq/kg for  $^{134}\text{Cs}$  and  $^{137}\text{Cs}$  respectively. In case of station-99 the concentration observed in December, 2012 was 79.52 Bq/kg and 155.85 Bq/kg for  $^{134}\text{Cs}$  and  $^{137}\text{Cs}$  respectively which highest in that location and in last sampling in October, 2013 the concentration was 26.76 Bq/kg and 66.82 Bq/kg for  $^{134}\text{Cs}$  and  $^{137}\text{Cs}$  respectively.

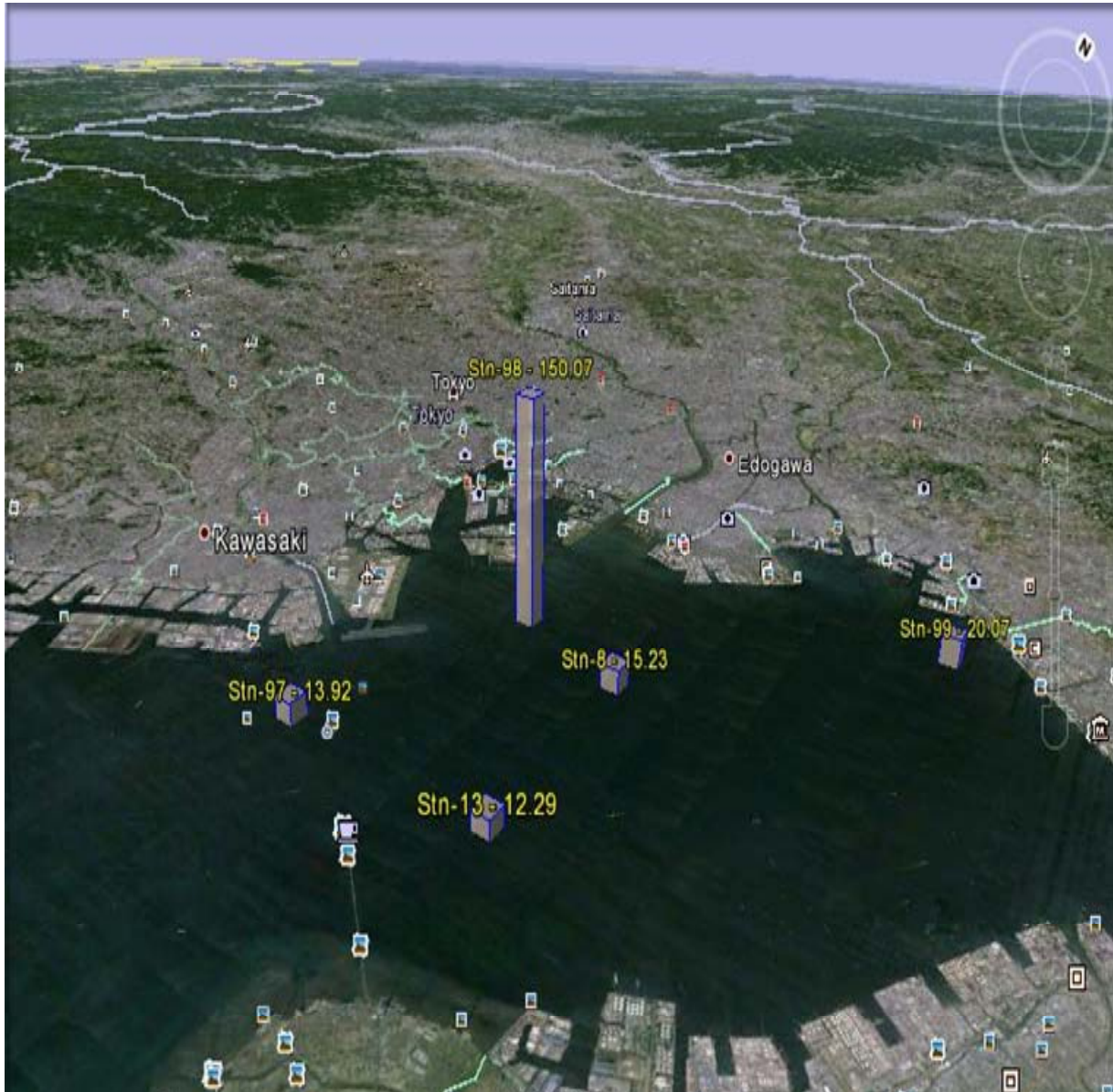


Figure 9 : Spatial Distribution of Radio-cesium in Tokyo Bay October, 2012.

# Cesium Distribution in Tokyo Bay



Figure 10 : Radio-cesium Distribution in the Tokyo Bay

#### 4.2 Radio- Cesium ( $^{134}\text{Cs}$ and $^{137}\text{Cs}$ ) distribution in Arakawa River

Figure-11 shows the spatial and temporal distribution of radio-caesium on surface sediment of sediment bed in Arakawa River from June-2012 to September-2013. In June ,2012 there was a peak of the caesium concentration at station 5 which is 1.5 km upstream from the river mouth and the concentration was 870 Bq/kg (Total Cesium  $^{134}\text{Cs}+^{137}\text{Cs}$ ). In September, 2012 there was two small peak at station-7 which is 3.59 km downstream from the river mouth that means within the Tokyo Bay (Total Cesium  $^{134}\text{Cs}+^{137}\text{Cs}$  was 695 Bq/kg) and in station 2 which is 7.27 km upstream from the river mouth (Total Cesium  $^{134}\text{Cs}+^{137}\text{Cs}$  was 412 Bq/kg). In November, 2012 there were two peaks at 1.5 km (Total Cesium  $^{134}\text{Cs}+^{137}\text{Cs}$  was 557 Bq/kg) and at 9 km (Total Cesium  $^{134}\text{Cs}+^{137}\text{Cs}$  was 490 Bq/kg) upstream area from the river mouth but from March the peaks disappear at 1.5 km (Total Cesium  $^{134}\text{Cs}+^{137}\text{Cs}$  was 75 Bq/kg) and at 9 km total Cesium ( $^{134}\text{Cs}+^{137}\text{Cs}$ ) was 112 Bq/kg and the next sampling time that in July, 2013 and the concentration was at 1.5 km (Total Cesium  $^{134}\text{Cs}+^{137}\text{Cs}$  was 226 Bq/kg) and at 9 km total Cesium ( $^{134}\text{Cs}+^{137}\text{Cs}$ ) was 71 Bq/kg. the last sampling time that was in September, 2013 there was three new peaks appeared among the one is large peak at station 4 and the concentration was 523 bq/kg ( $^{134}\text{Cs}+^{137}\text{Cs}$ ) and other two peaks appeared at station 2 the concentration was 302 bq/kg ( $^{134}\text{Cs}+^{137}\text{Cs}$ ) and station 6 or river mouth area, the concentration was 241 bq/kg ( $^{134}\text{Cs}+^{137}\text{Cs}$ ).



# Cesium Distribution in Arakawa River

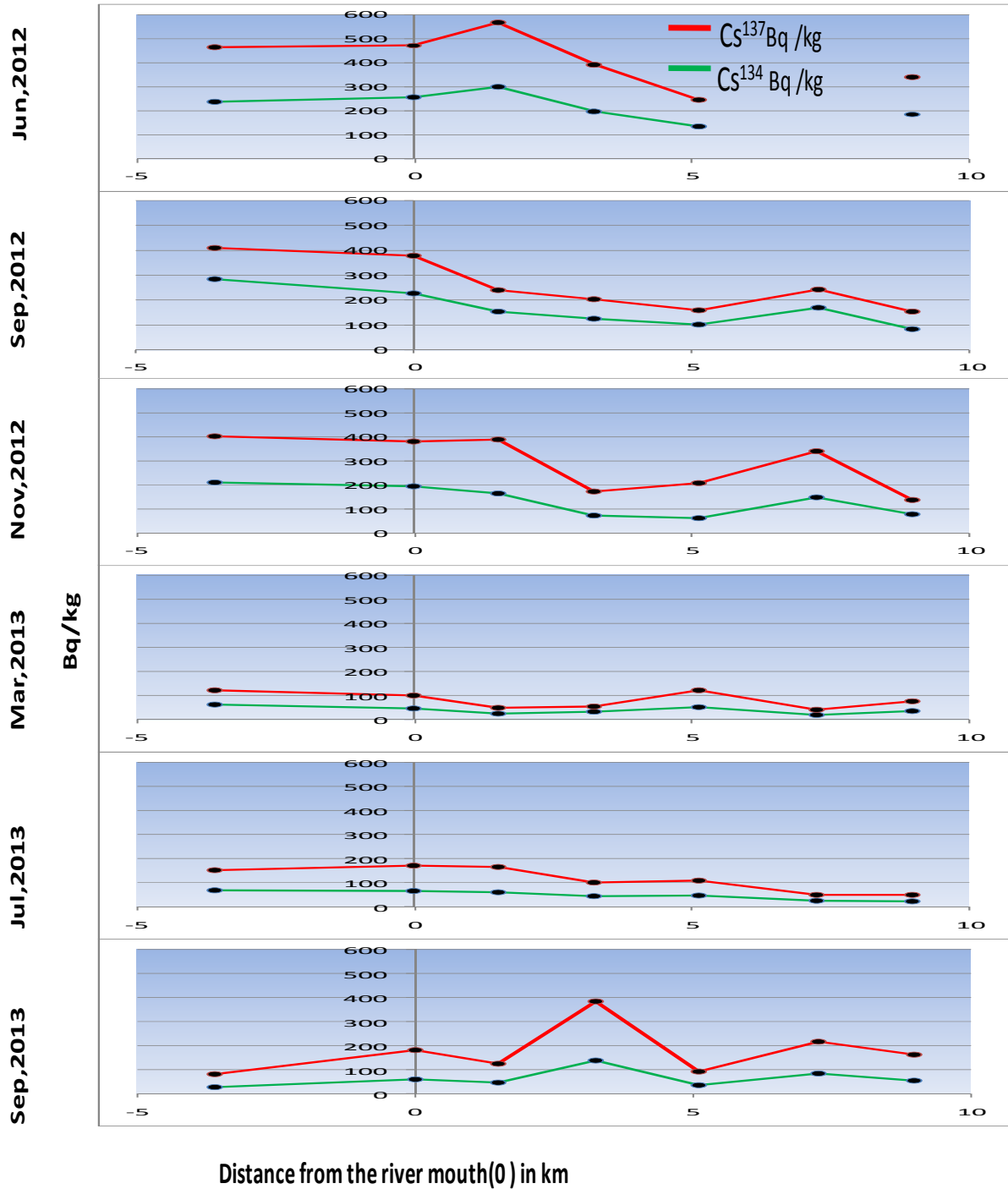


Figure 11 : Radio-Cesium Distribution Time series in Arakawa River

### 4.3 Radio- Cesium ( $Cs^{134}$ and $Cs^{137}$ ) distribution in Edogawa River

Figure-12 shows the spatial and temporal distribution of radio-caesium on surface sediment of sediment bed in Edogawa River from June-2012 to September-2013. In June, 2012 there was a peak of the caesium concentration at station 14 which is about 10 km upstream from the river mouth and the concentration was 290 Bq/kg (Total Caesium  $^{134}Cs+^{137}Cs$ ). In September, 2012 there was also same peak at station-14 and the concentration was 236 Bq/kg (Total Caesium  $^{134}Cs+^{137}Cs$ ). In November, 2012 there were two peaks at station 15, 11.33 km upstream from the river mouth (Total Caesium  $^{134}Cs+^{137}Cs$  was 1010 Bq/kg) and at station 14, about 10 km upstream from the river mouth it continued the higher concentration (Total Caesium  $^{134}Cs+^{137}Cs$  was 542 Bq/kg) and at station 12, 5 km upstream area from the river mouth the peak appeared newly and the concentration was 203 Bq/kg ( $^{134}Cs+^{137}Cs$ ). In March the peak disappear at station 12, (Total Caesium  $^{134}Cs+^{137}Cs$  was 83 Bq/kg) and at station 15 and 14 caesium concentration decreased but the peak was remained and total caesium concentration ( $^{134}Cs+^{137}Cs$ ) was 772 Bq/kg and 245 Bq/kg respectively. The next sampling time that in July, 2013 at station 15 and 14 caesium concentration decreased but the peak was remained as March, 2013 and total caesium concentration ( $^{134}Cs+^{137}Cs$ ) was 694 Bq/kg and 386 Bq/kg respectively. The last sampling time that was in September, 2013, at station 15 and 14 caesium concentration decreased but the peak was remained as July, 2013 and total caesium concentration ( $^{134}Cs+^{137}Cs$ ) was 688 Bq/kg and 188 Bq/kg respectively. But in the river mouth area the concentration increased and the amount was 170 Bq/kg ( $^{134}Cs+^{137}Cs$ ) though it was 130 Bq/kg in July, 2013.

## Cesium Distribution in Edogawa River

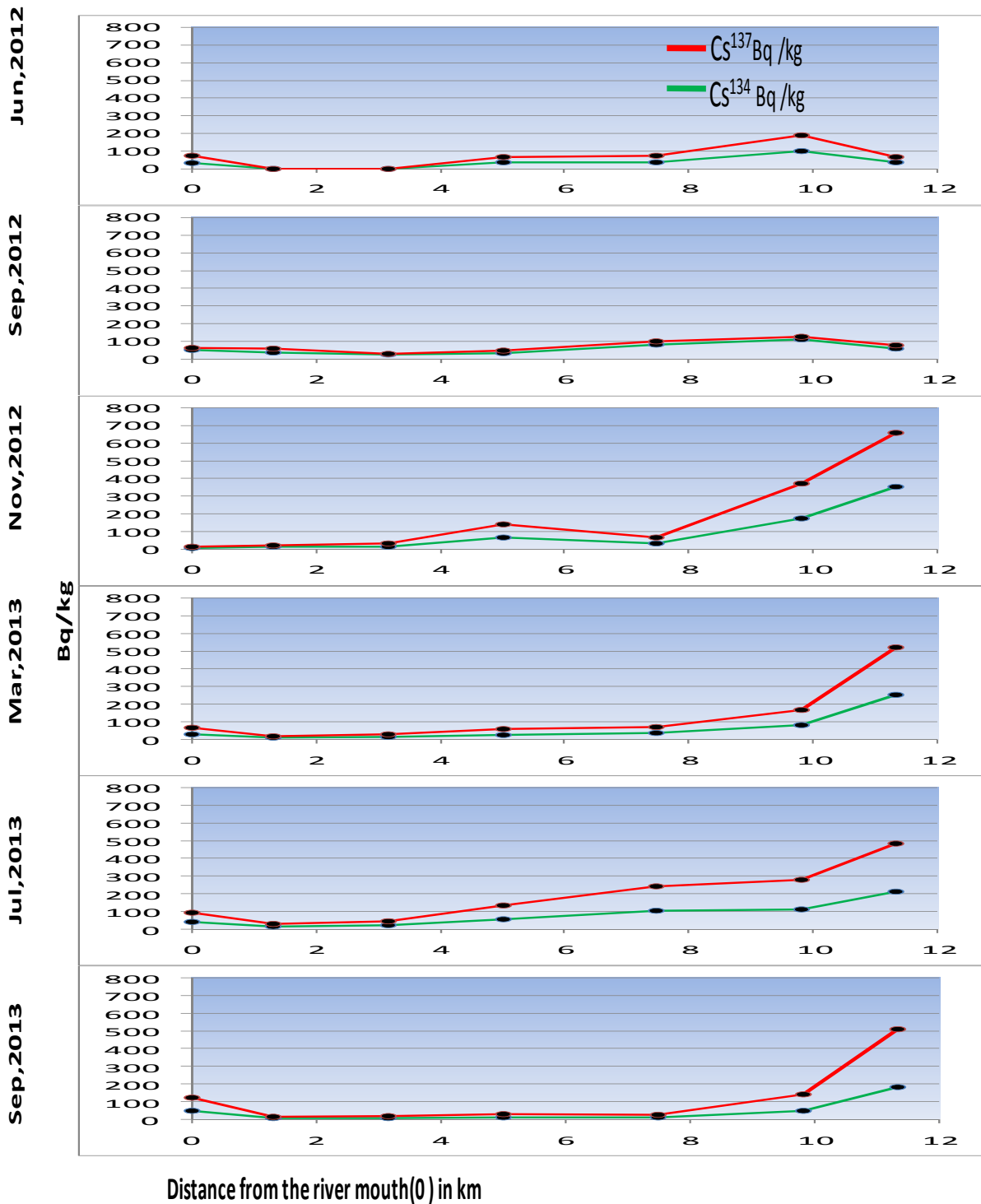


Figure 12 : Radio-Cesium Distribution Time series in Edogawa River

#### 4.4 Radio- Cesium ( $^{134}\text{Cs}+^{137}\text{Cs}$ ) distribution in Sumidagawa River

Figure-13 and Table: 9 show the spatial and temporal distribution of radio-caesium on surface sediment of sediment bed in Sumidagawa River and estuary from August-2012 to September-2013. First time in August, 2012 samples from 17 points were collected though the total sampling points were 24 in this river and estuary. The results shows that in the upstream area the concentration of caesium in the surface sediment is quite high and the total caesium concentration ( $^{134}\text{Cs}+^{137}\text{Cs}$ ) at station N21, station N20, station N19, station N17 and station N15 were 839 Bq/kg, 1683 Bq/kg, 1587 Bq/kg, 1003 Bq/kg and 395 Bq/kg respectively. From station N14 to station N0 the total caesium concentration ( $^{134}\text{Cs}+^{137}\text{Cs}$ ) was low almost in every station, less than 200 Bq/kg. Next in December, 2012 the samples were taken from all 24 stations, Station N0 to Station N23. The results shows that also in the upstream area the concentration of caesium in the surface sediment is quite high and the total caesium concentration ( $^{134}\text{Cs}+^{137}\text{Cs}$ ) at station N23, station N22, station N21, station N20, station N19, station N17 and station N15 were 737 Bq/kg, 1032 Bq/kg, 745 Bq/kg, 1175 Bq/kg, 600 Bq/kg, 292 Bq/kg and 526 Bq/kg respectively. The last sampling point in this area that is station N0 and located within the Tokyo Bay, the total caesium concentration ( $^{134}\text{Cs}+^{137}\text{Cs}$ ) was 234 Bq/kg. In March, 2013, The results shows that also in the upstream area the concentration of caesium in the surface sediment is quite high than the middle and lower stream area and the total caesium concentration ( $^{134}\text{Cs}+^{137}\text{Cs}$ ) at station N23, station N22, station N21, station N20, station N19, station N17 and station N15 were 469 Bq/kg, 298 Bq/kg, 342 Bq/kg, 265 Bq/kg, 260 Bq/kg, 150 Bq/kg and 120 Bq/kg respectively. The last sampling point in this area that is station N0 and located within the Tokyo Bay, the total caesium concentration ( $^{134}\text{Cs}+^{137}\text{Cs}$ ) was 170 Bq/kg. In last sampling that was in September, 2013 The results shows that also in the upstream area the concentration of caesium in the surface sediment is quite high than the middle and lower stream area and the total caesium concentration ( $^{134}\text{Cs}+^{137}\text{Cs}$ ) at station N23, station N22, station N21, station N20, station N19, station N17 and station N15 were 450 Bq/kg, 400 Bq/kg, 270 Bq/kg, 502 Bq/kg, 443 Bq/kg, 131 Bq/kg and 122 Bq/kg respectively. The last sampling point in this area that is station N0 and located within the Tokyo Bay, the total caesium concentration ( $^{134}\text{Cs}+^{137}\text{Cs}$ ) was 114 Bq/kg.

**Table 9 : The Result of Sumidagawa River Estuary**

<b>Total Cesium Concentration ( Cs134+ Cs137)</b>				
	August,2012	December,2012	March,2013	September,2013
N-0		234.46	169.57	113.98
N-1	47.47	84.87	60.44	42.83
N-2	144.82	71.95	58.14	38.33
N-3	145.01	78.65	134.12	25.53
N-4	215.38	119.85	56.78	32.60
N-5		135.39	57.30	17.58
N-6	116.11	53.35	37.44	29.07
N-7	189.57	76.68	22.26	25.36
N-8	205.55	47.34	35.50	9.59
N-9		5.71	7.17	3.47
N-10	110.83	38.12	22.05	4.30
N-11	5.65	13.51	20.18	19.74
N-12	222.75	52.05	33.94	48.91
N-13	109.90	160.82	45.91	54.18
N-14		18.84	12.59	31.11
N-15	394.73	526.93	119.92	122.78
N-16	54.03	525.41	54.09	10.92
N-17	1003.49	292.95	150.79	131.09
N-18		264.92	243.88	11.75
N-19	1586.65	600.22	260.29	443.77
N-20	1682.66	1175.24	265.53	502.18
N-21	838.91	745.03	342.90	270.24
N-22		1032.63	298.68	400.09
N-23		737.02	469.46	450.46

## Spatial and Temporal Distribution of Cesium in Sumidagawa River

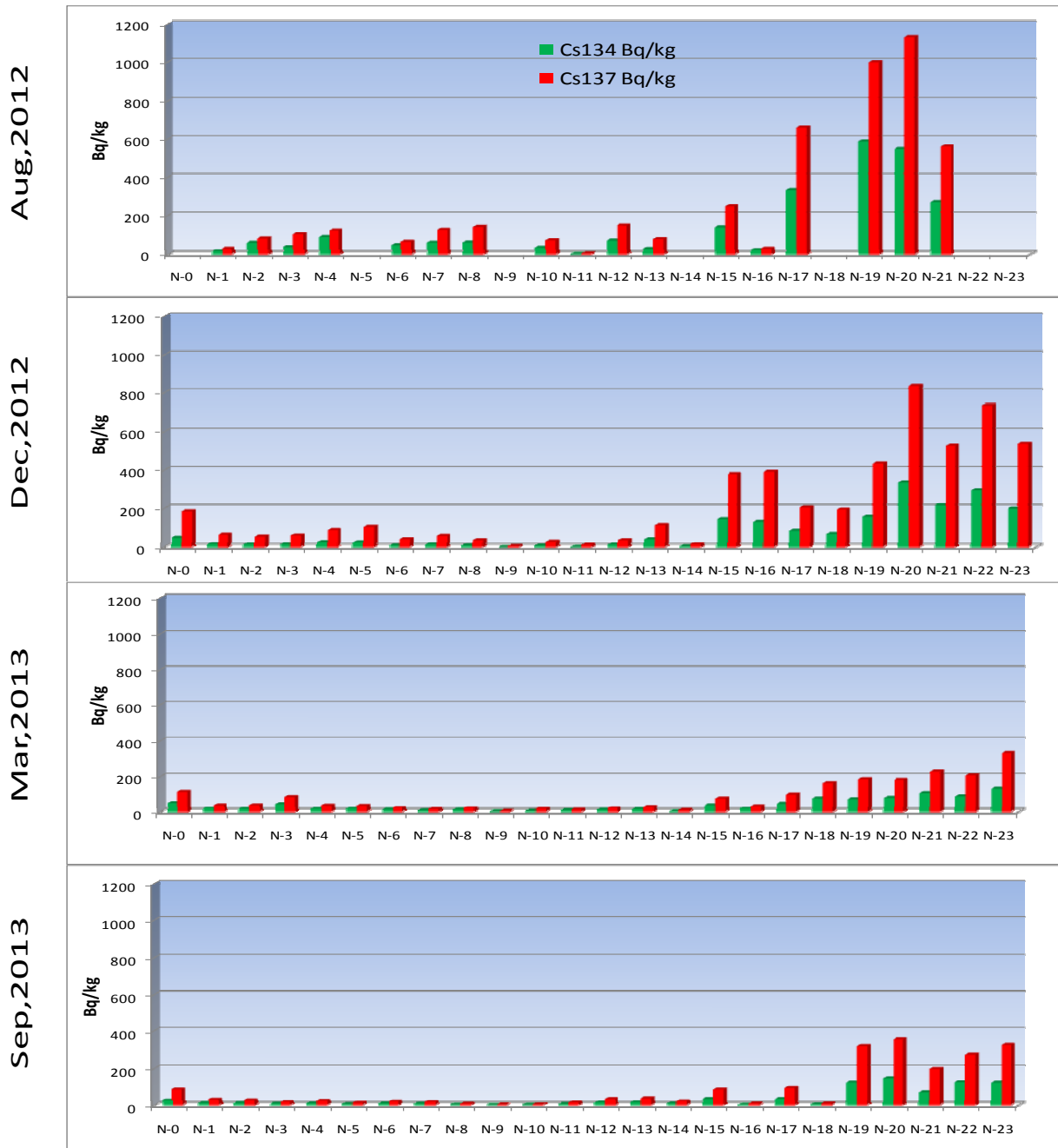


Figure 13 : Radio-Cesium distribution in Sumidagawa River and Estuary.

#### 4.5 Core Samples Result of Arakawa River

In Arakawa River from two locations, station-4 which is 3.25 km. upstream from the river mouth and another station-7 which is 3.59 km. downstream from the river mouth means within the Tokyo Bay, the core samples were taken to observe the vertical profile, movement and diffusion activity. The cores total length were 38 cm the cores were divided into the 12 layers first 5 layers width were 2 cm each and 6 to 12<sup>th</sup> layer the cores were cut into 4 cm width. Figure-14 shows the time series of core result from station-4 and the cores were taken four times September, 2012, November, 2012, March, 2013 and July, 2013. First time in September, 2012 there was a clear peak at the depth 8-10 cm from the sediment bed and the total Cesium concentration ( $^{134}\text{Cs}+^{137}\text{Cs}$ ) was 1683 Bq/kg. in November the peak moved to more deeper in 18-22 cm and the total Cesium concentration ( $^{134}\text{Cs}+^{137}\text{Cs}$ ) was 1179 Bq/kg, in March, 2013 the peak again moved to 8-10 cm depth and the total Cesium concentration ( $^{134}\text{Cs}+^{137}\text{Cs}$ ) was 1207 Bq/kg and in July, 2013 the peak moved to more upward 2-4 cm depth and the total Cesium concentration ( $^{134}\text{Cs}+^{137}\text{Cs}$ ) was 824 Bq/kg.

#### Station-4, Arakawa River

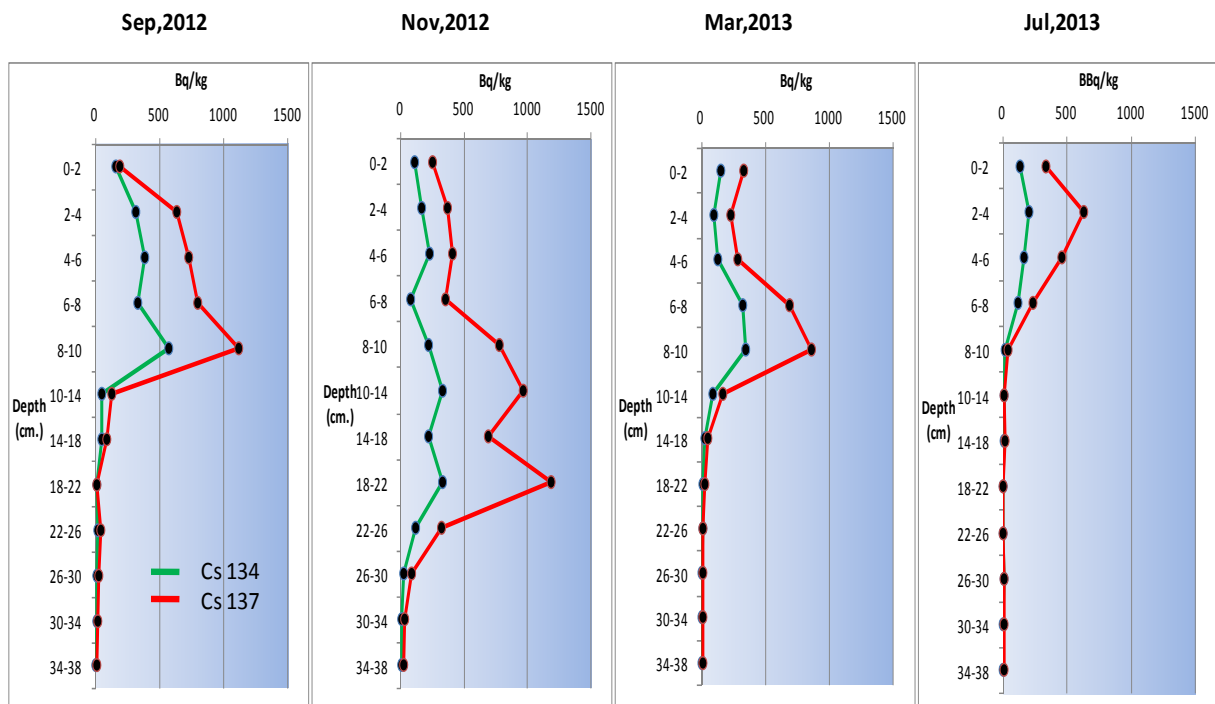


Figure 14 : Time series of Core samples result in station-4 Arakawa River.

Figure-15 shows the time series of core result from station-7 and the cores were taken three times September, 2012, November, 2012 and March, 2013. First time in September, 2012 there was a clear peak at the depth 2-4 cm from the sediment bed and the total Cesium concentration ( $^{134}\text{Cs}+^{137}\text{Cs}$ ) was 295 Bq/kg, in November, 2012 the peak was in the same depth 2-4 cm and the total Cesium concentration ( $^{134}\text{Cs}+^{137}\text{Cs}$ ) was 287 Bq/kg and in March, 2013 also the peak was in the same layer 2-4 cm depth and the total Cesium concentration ( $^{134}\text{Cs}+^{137}\text{Cs}$ ) was 239 Bq/kg.

### Station-7, Arakawa River

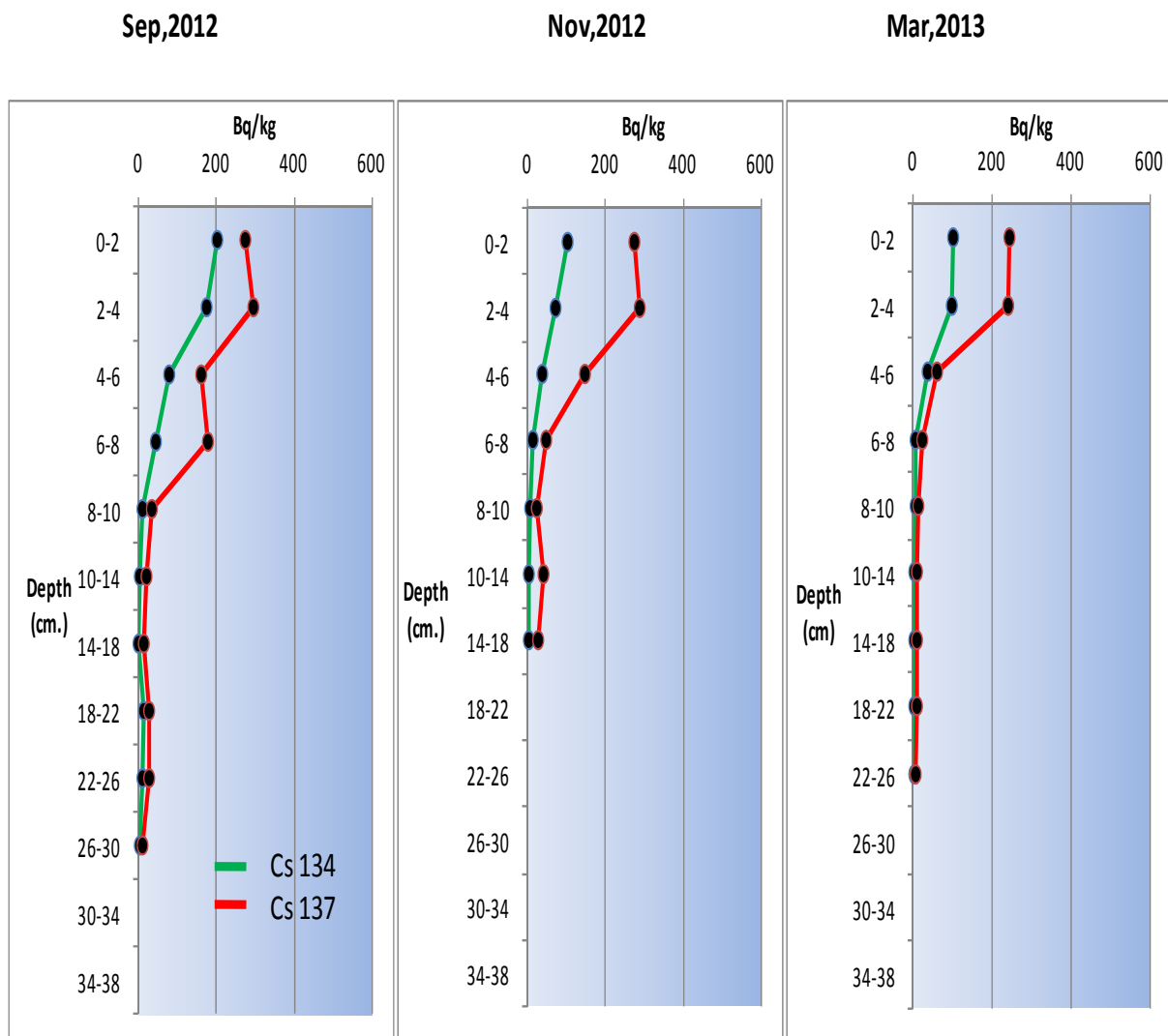


Figure 15 : Time series of Core samples result in station-7 Arakawa River.



#### 4.6 Core Samples Result of Edogawa River

In Edogawa River from two locations, station-9 which is assumed as the river mouth where it falls in the Tokyo Bay and another station-14 which is located 9.81 km. upstream from the river mouth, the core samples were taken to observe the vertical profile, movement and diffusion activity. The cores total length were 38 cm the cores were divided into the 12 layers first 5 layers width were 2 cm each and 6 to 12<sup>th</sup> layer the cores were cut into 4 cm width. Figure-16 shows the time series of core result of station-9 river mouth of Edogawa River. The cores were taken four times September, 2012, November, 2012, March, 2013 and July, 2013. First time in September, 2012 there were two clear peak at the depth 4-6 cm and 10-14 cm depth from the sediment bed and the total Cesium concentration ( $^{134}\text{Cs}+^{137}\text{Cs}$ ) was 405 Bq/kg and 533 Bq/kg respectively. In November, 2012 also there were two clear peak at the depth 8-10 cm and 14-18 cm depth from the sediment bed and the total Cesium concentration ( $^{134}\text{Cs}+^{137}\text{Cs}$ ) was 325 Bq/kg and 273 Bq/kg respectively, in March, 2013 there were also two clear peak in the same depth at the depth 4-6 cm and 10-14 cm depth from the sediment bed and the total Cesium concentration ( $^{134}\text{Cs}+^{137}\text{Cs}$ ) was 548 Bq/kg and 484 Bq/kg respectively and in July, 2013 again there were two clear peak at the depth 4-6 cm and 10-14 cm depth from the sediment bed and the total Cesium concentration ( $^{134}\text{Cs}+^{137}\text{Cs}$ ) was 258 Bq/kg and 263 Bq/kg respectively.

# Station-9, Edogawa River

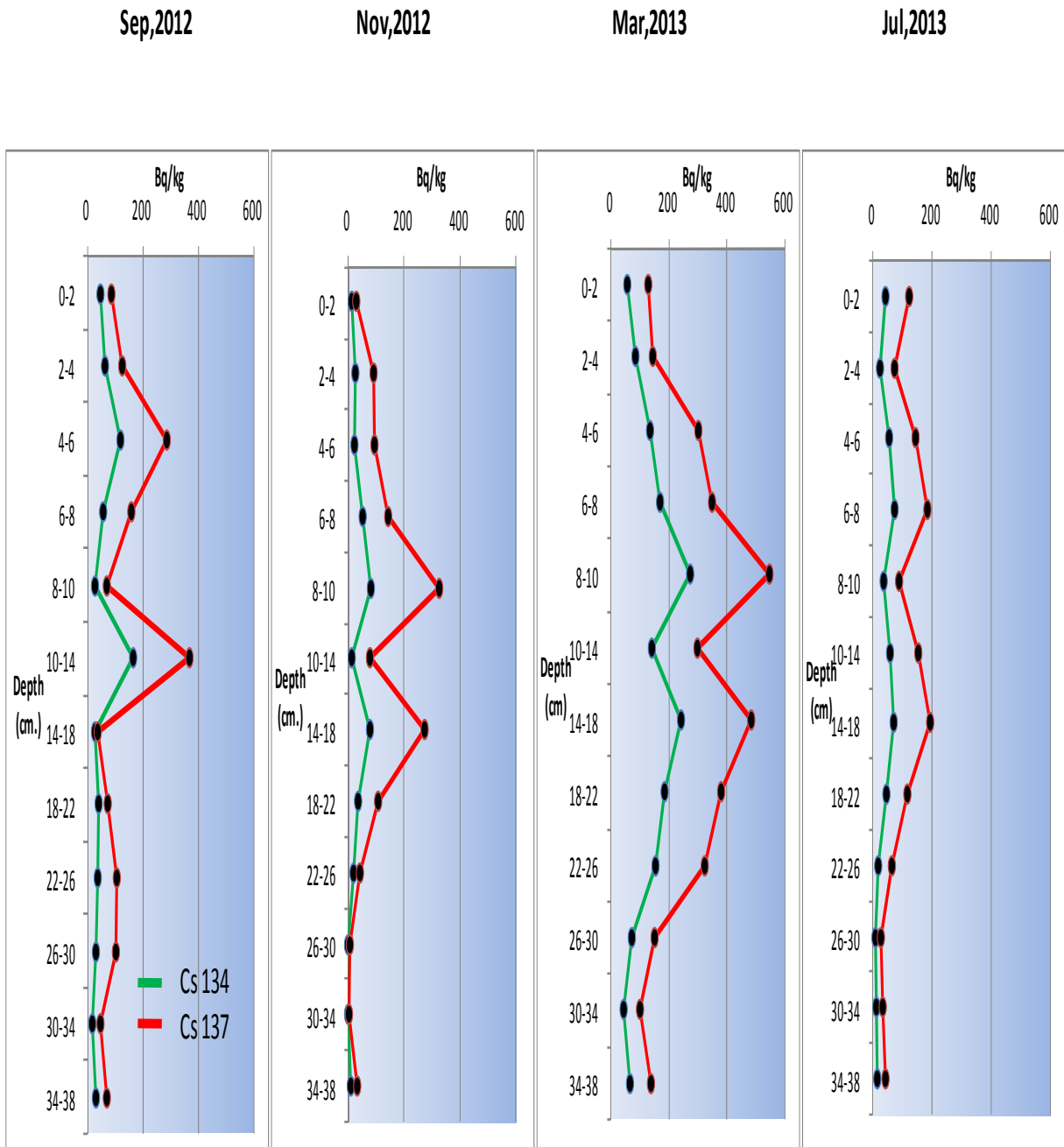


Figure 16 : Time series of Core samples result in station-9 Edogawa River.

Figure-17 shows the time series of core result of station-14 and two times, September, 2012 and July, 2013 results shown here. First time in September, 2012 there was a clear peak at the depth 10-14 cm from the sediment bed and the total Cesium concentration ( $^{134}\text{Cs}+^{137}\text{Cs}$ ) was 1358 Bq/kg, in July, 2013 the peak was in the same depth 10-14 cm and the total Cesium concentration ( $^{134}\text{Cs}+^{137}\text{Cs}$ ) was 630 Bq/kg.

## Station-14, Edogawa River

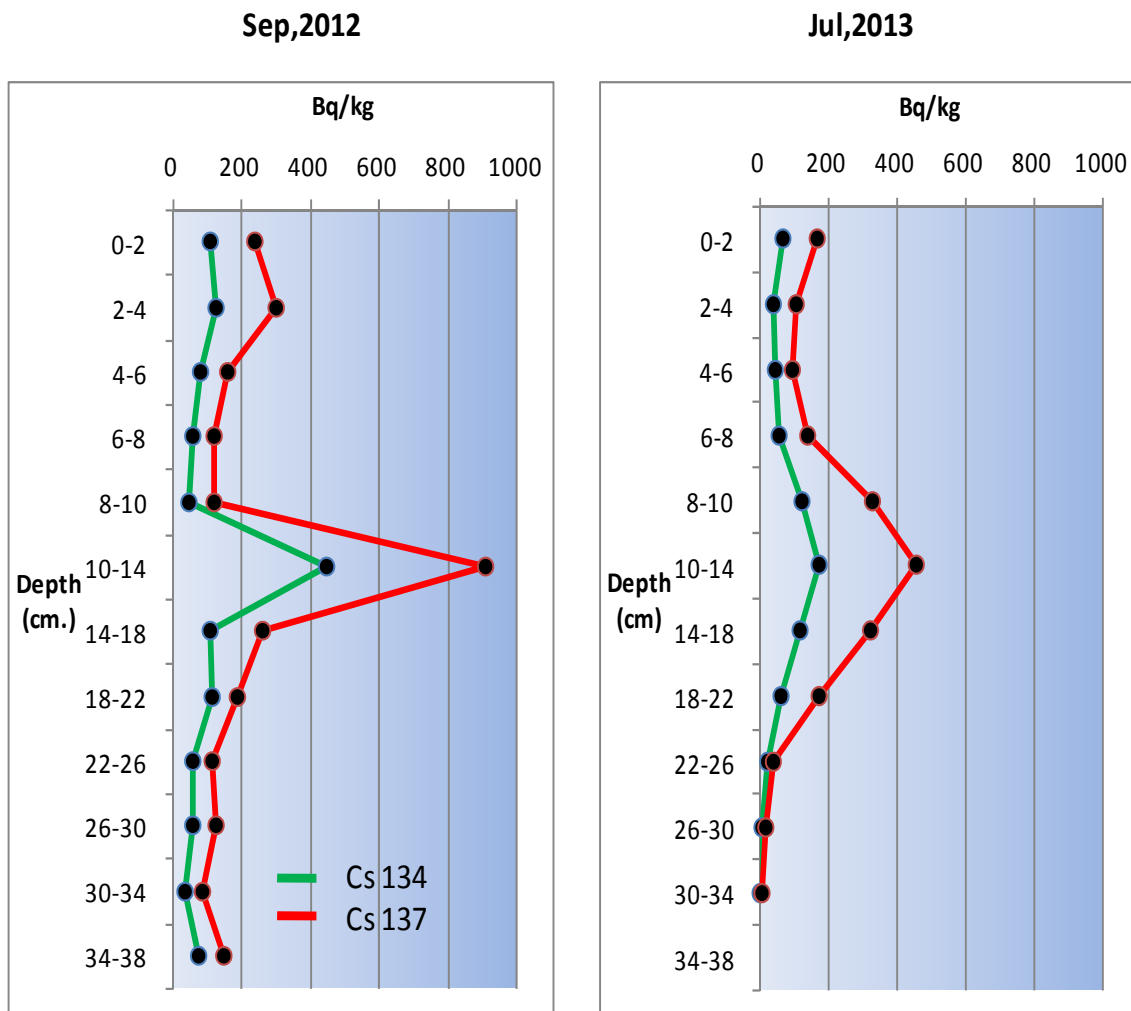


Figure 17 : Time series of Core samples result in station-14 Edogawa River.

#### 4.7 Core Samples Result of Sumidagawa River

In Sumidagawa River from two locations, station-20 which is located in the upstream region from the river mouth and another station-17 which is also located in upstream region from the river mouth (Figure-3), the core samples were taken to observe the vertical profile, movement and diffusion activity. The cores total length were 38 cm the cores were divided into the 12 layers first 5 layers width were 2 cm each and 6 to 12<sup>th</sup> layer the cores were cut into 4 cm width. Figure-18 shows the time series of core result of station-20, in December, 2012 there was a peak at the depth 6-8 cm from the sediment bed and the total Cesium concentration ( $^{134}\text{Cs}+^{137}\text{Cs}$ ) was 3725 Bq/kg, in September, 2013 the peak was in the depth 8-10 cm and the total Cesium concentration ( $^{134}\text{Cs}+^{137}\text{Cs}$ ) was 1255 Bq/kg.

#### Station-20, Sumidagawa River

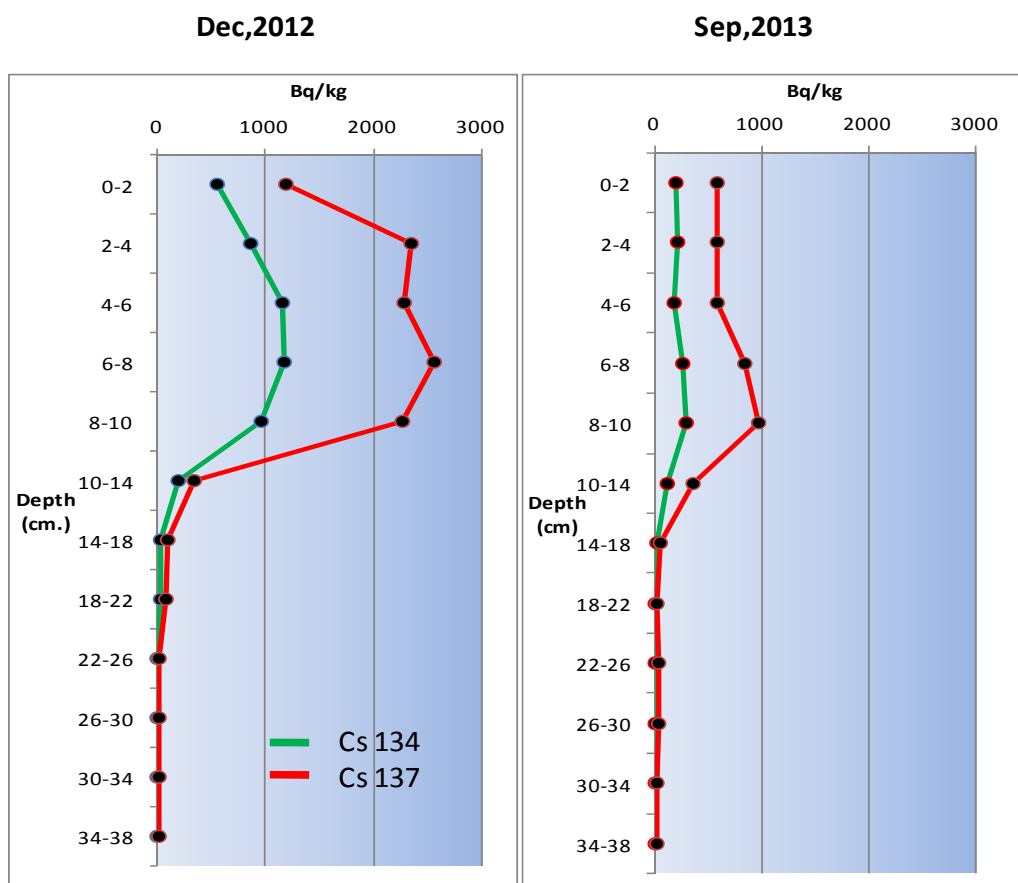


Figure 18 : Time series of Core samples result in station-20 Sumidagawa River.

Figure-19 shows the core result of station-17 of Sumidagawa River. Here only one time sample result were available and there found that there were two clear peak in the depth 4-6 cm and 8-10 cm depth and the total Cesium concentration ( $^{134}\text{Cs}+^{137}\text{Cs}$ ) was 121 Bq/kg and 111 Bq/kg respectively.

## Station-17, Sumidagawa River

Dec,2012

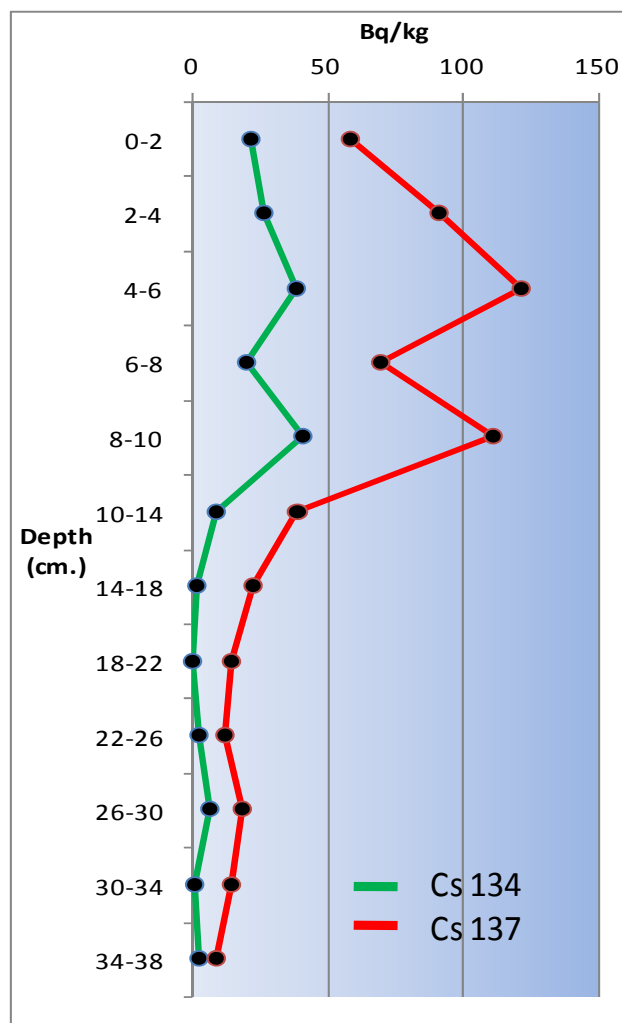


Figure 19 : Core samples result in station-17 Sumidagawa River

#### 4.8 Diffusion and Advection in Vertical Profile

The diffusion coefficient  $k_z$  was calculated according to methodology using Eq-5, thus the result of the core samples were checked by using the diffusion equation (Eq-2) and compared with the observed result and R squared value of this observed and estimated value was compared with R squared value of advection to get the information which component was dominant in those places. In Arakawa River from station-4 and station-7 core were taken, diffusion coefficient were calculated by using the equation-1. In station-4 the diffusion coefficient was  $8.91 \times 10^{-09}$  and in station-7 was  $8.98 \times 10^{-08}$  and using these diffusion coefficient and equation-1 the vertical profile for  $^{137}\text{Cs}$  was reproduced and compared with the observed data (Figure-20). Likewise in Edogawa River the core were taken from station-9 and station-14 and the diffusion coefficient was found as  $3.78 \times 10^{-08}$  and  $3.03 \times 10^{-08}$ , using these diffusion coefficient and equation-1 the vertical profile for  $^{137}\text{Cs}$  was reproduced and compared with the observed data (Figure-21). In case of Sumidagawa River the core sample were taken from station-20 the diffusion coefficient was found  $1.72 \times 10^{-07}$ , using these diffusion coefficient and equation-1 the vertical profile for  $^{137}\text{Cs}$  was reproduced and compared with the observed data (Figure-22).

Station-4, Cs 137 Sep,2012 and Mar,2013

Station-7, Cs 137 Sep,2012 and Mar,2013

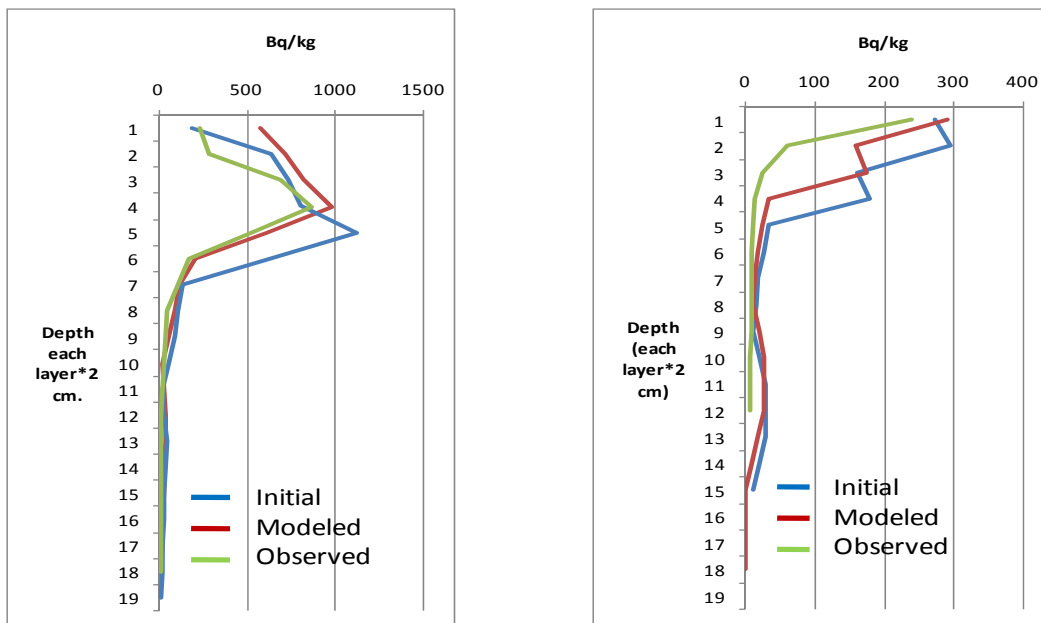


Figure 20 : The vertical profile for  $^{137}\text{Cs}$  using the diffusion coefficient and equation-1 in Station-4 and station-7, Arakawa River

Station-9, Cs 137 Sep,2012  
and Nov,2012

Station-9, Cs 137 Sep,2012 and  
Jul,2013

Station-14, Cs 137 Sep,2012 and  
Jul,2013

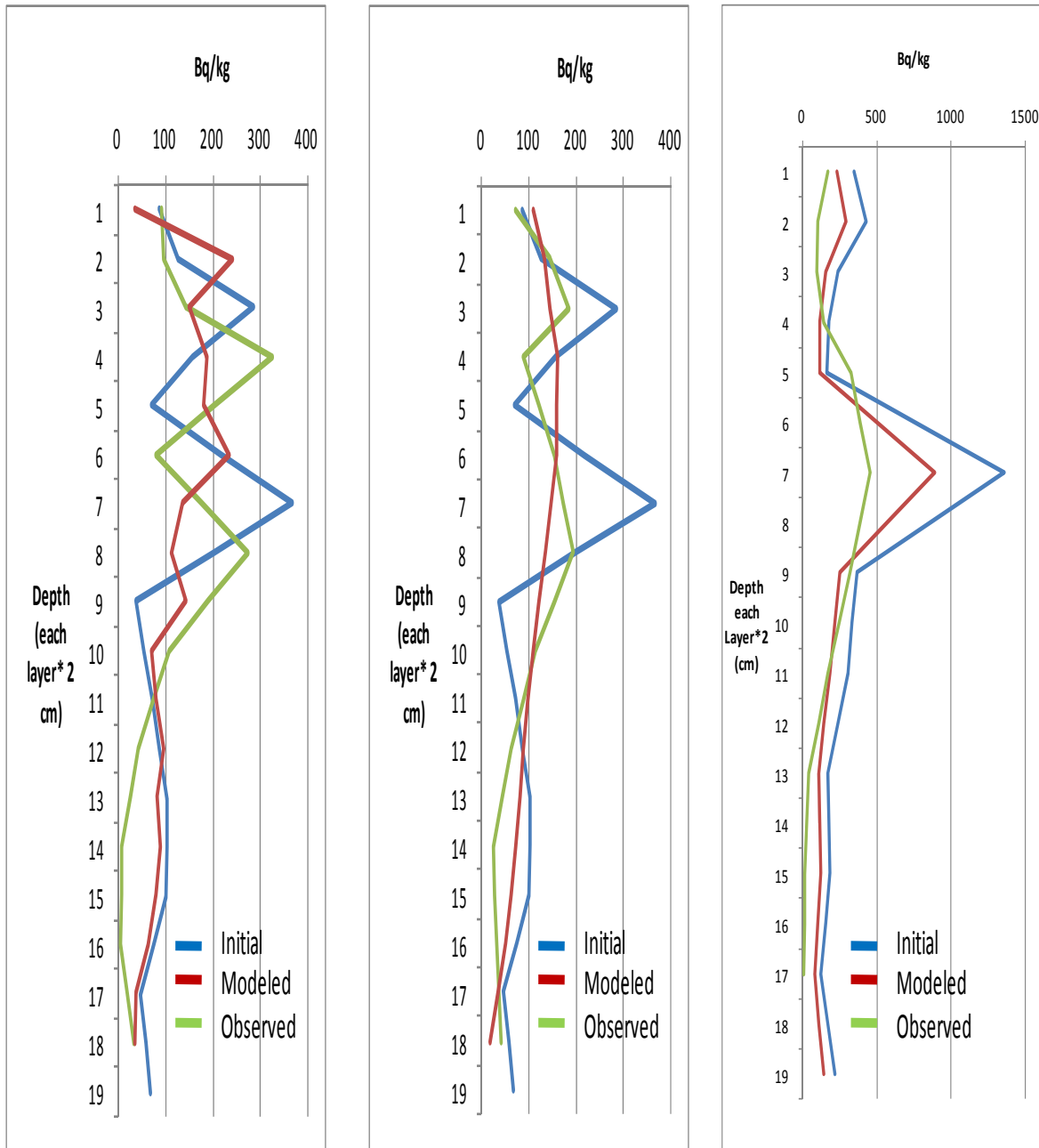


Figure 21 : The vertical profile for <sup>137</sup>Cs using the diffusion coefficient and equation-1 in Station-9 and station-14, Edogawa River

# Sumidagawa Station-20, Cs 137 Dec,2012 and Sep,2013

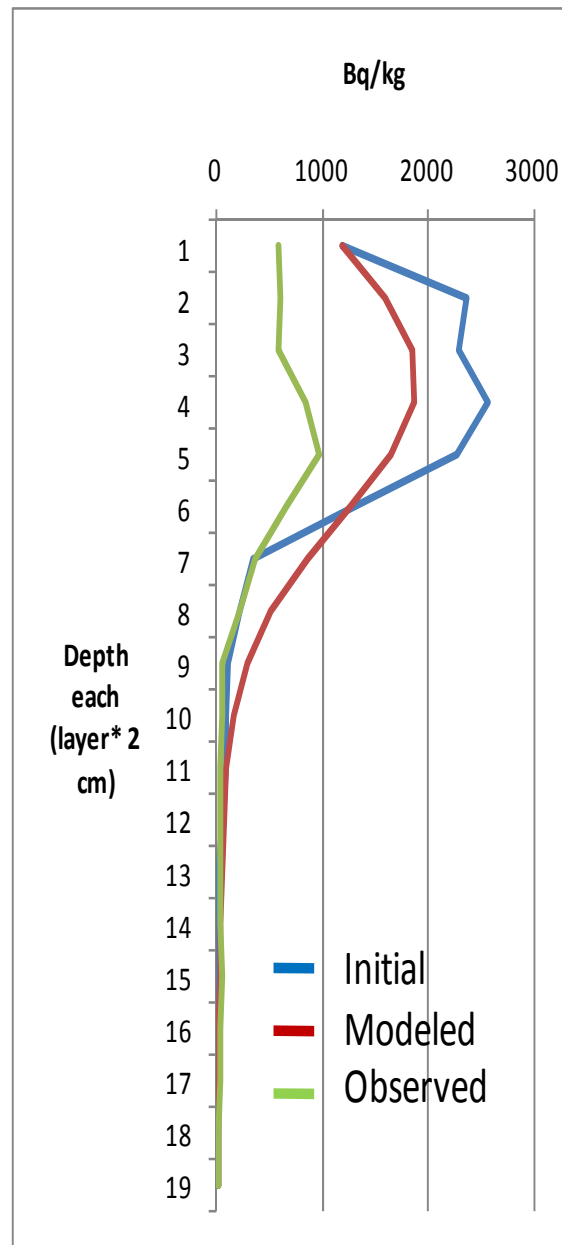


Figure 22 : The vertical profile for 137 Cs using the diffusion coefficient and equation-1 in Station-20, Sumidagawa River



According to methodology and using Eq-8, the vertical profile was reproduced and compared with the observed result and R squared value of this observed and estimated value was compared with R squared value of Diffusion to get the information which component was dominant in those places. Figure-23 shows the result of station-4 upstream area of Arakawa River, figure-24 shows the result of station-7 river mouth area of Arakawa River, figure-25 shows the result of station-9 River mouth area of Edogawa River, and figure-26 shows the result of station-14 upstream area of Edogawa River and figure-27 shows the result of station-20 upstream area of Sumidagawa River. Table-10 shows the result of R squared value of all stations. The coefficient of sedimentation varied from -0.03 to 1.

Station-4, Cs 137 Sep,2012  
and Nov,2012

Station-4, Cs 137 Sep,2012  
and Mar,2013

Station-4, Cs 137 Sep,2012  
and Jul,2013

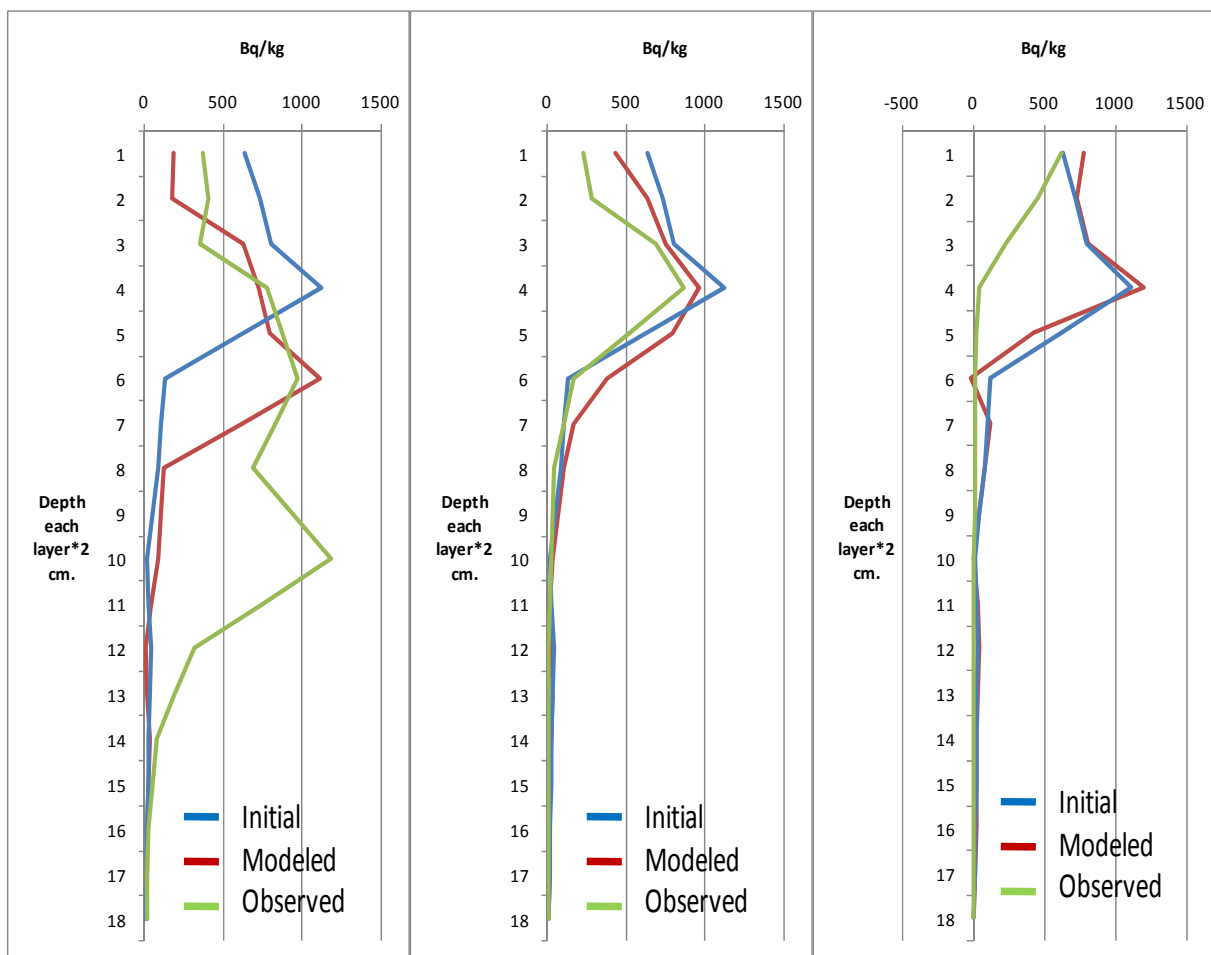


Figure 23 : The vertical profile for 137 Cs using the Advection equation in Station-4 Arakawa River.

Station-7, Cs 137 Sep,2012  
and Nov,2012

Station-7, Cs 137 Sep,2012  
and Mar,2013

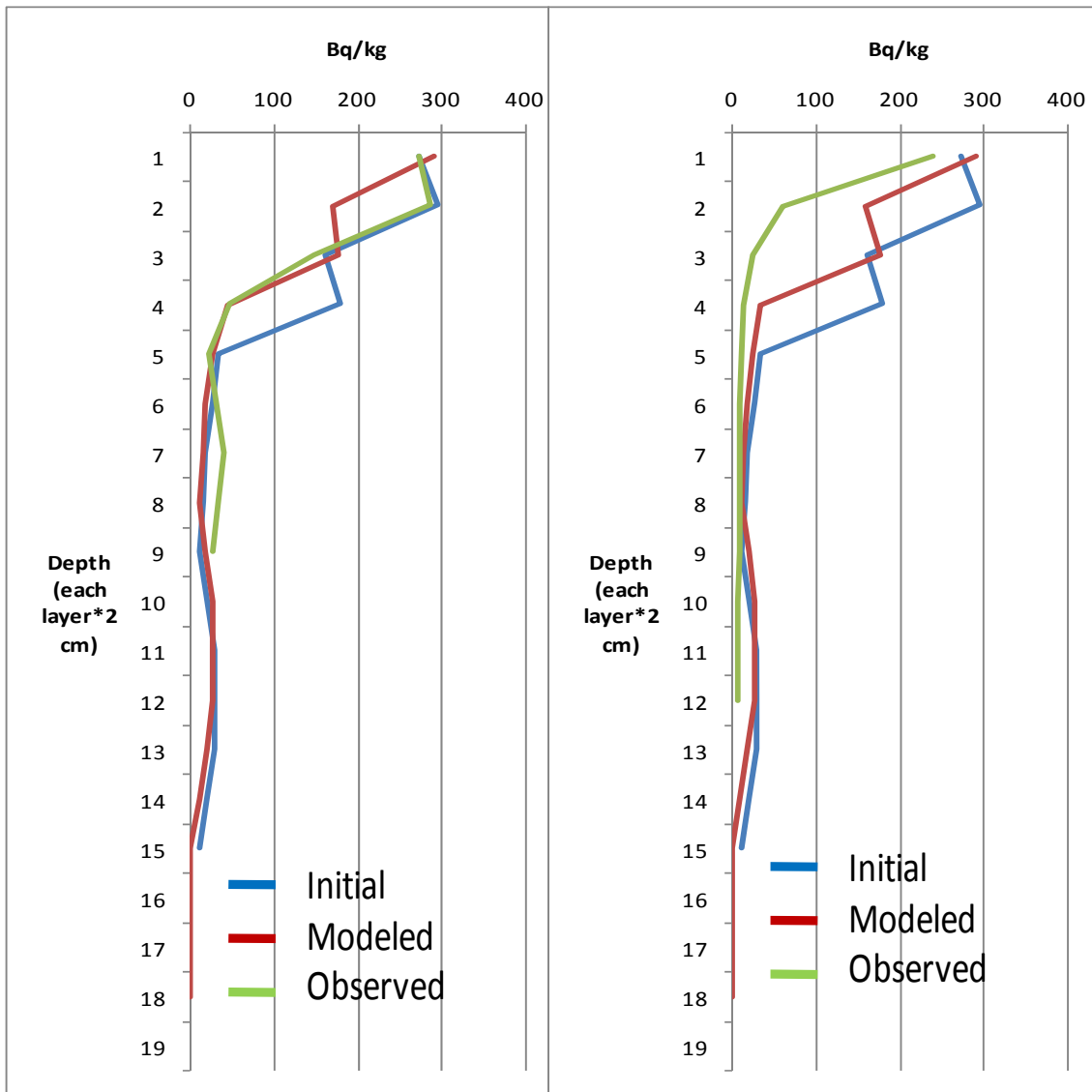


Figure 24 : The vertical profile for <sup>137</sup>Cs using the Advection equation in Station-7, Arakawa River.

Station-9, Cs 137 Sep,2012  
and Nov,2012

Station-9, Cs 137 Sep,2012  
and Jul,2013

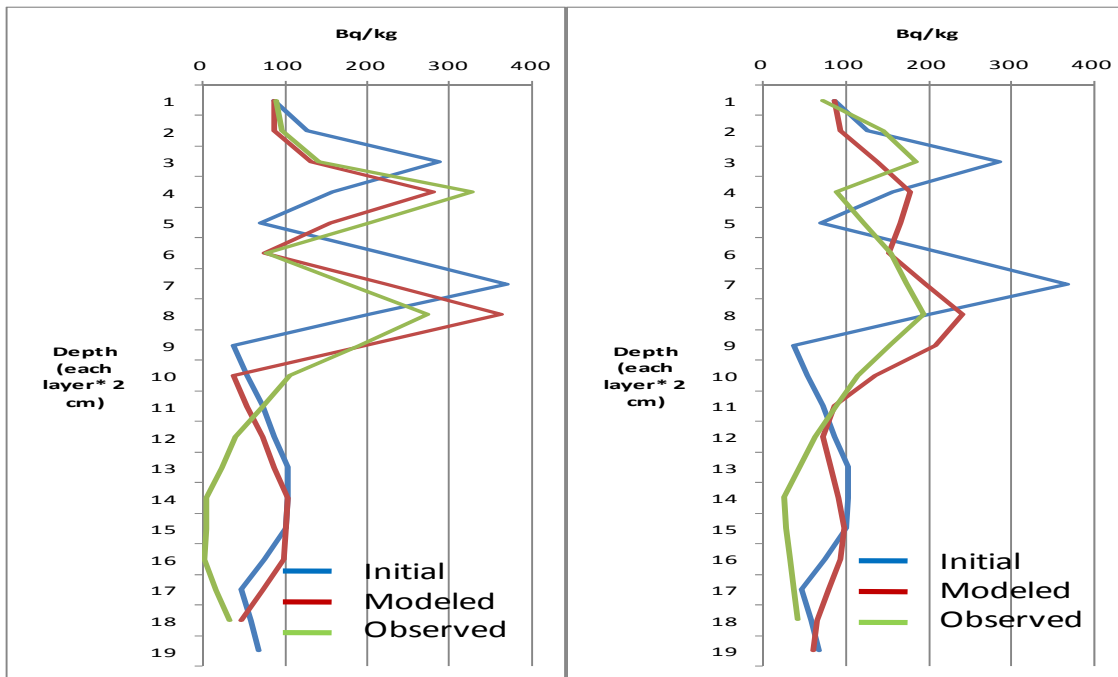


Figure 25 : The vertical profile for 137 Cs using the Advection equation in Station-9, Edogawa River.

Station-14, Cs 137 Sep,2012  
and Jul,2013

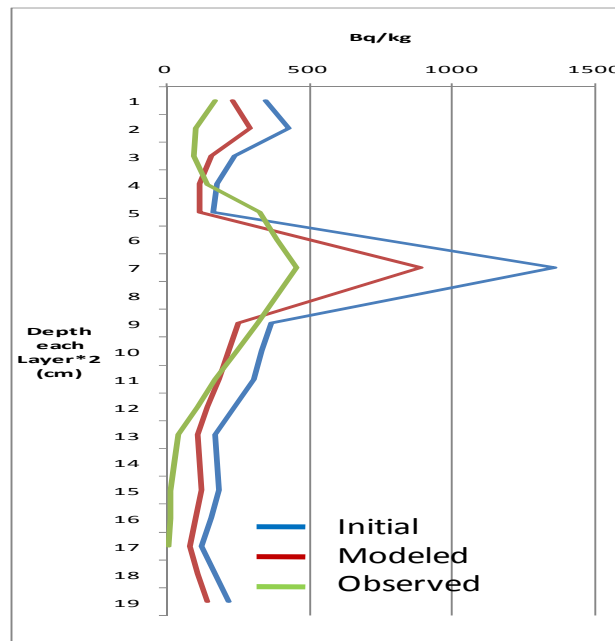


Figure 26 : The vertical profile for 137 Cs using the Advection equation in Station-14, Edogawa River.

Sumidagawa Station-20, Cs 137 Dec,2012 and Sep,2013

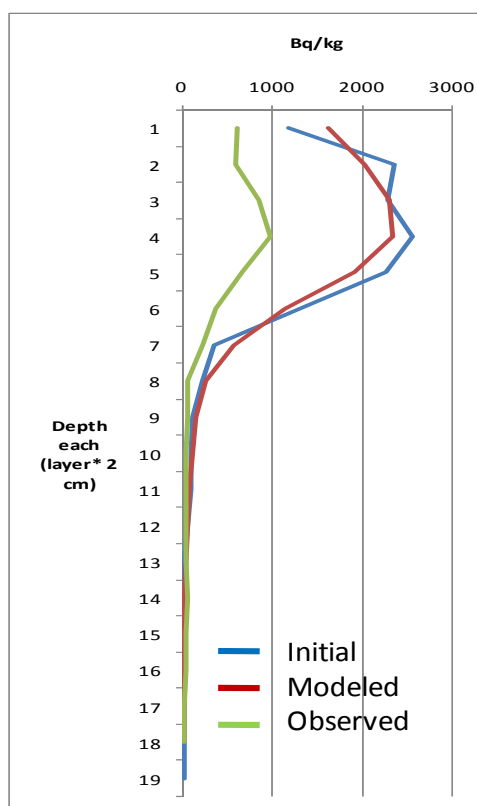


Figure 27 : The vertical profile for 137 Cs using the Advection equation in Station-20, Sumidagawa River.

Table 10 : Squared R Value for Sedimentation Rate and Diffusion Coefficient and Precipitation Data.

		In Winter (Dry season) Nov,2012	In Summer (Wet Season) Jul,2013
Location and River	Parameter	R Squared Value	R Squared Value
Station-4, Arakawa R.	Sedimentation	0.283	0.398
	Diffusion	0.095	0.341
	Prcipitation(mm)	33.5	139.5
Station-7, Arakawa R.	Sedimentation	0.776	
	Diffusion	0.795	
	Prcipitation(mm)	33.5	139.5
Station-14, Edogawa R.	Sedimentation		0.654
	Diffusion		0.964
	Prcipitation(mm)	37.5	148
Station-9, Edogawa R.	Sedimentation	0.698	0.689
	Diffusion	0.256	0.644
	Prcipitation(mm)	37.5	148

## **4.9 Sediment Particle Size and Cesium Concentration**

### **4.9.1 Sediment Particle Size and Cesium distribution in Arakawa River**

Particle size of the sediment sample of Arakawa River was measured to know the relationships between particle size and total cesium concentration, figure-28 shows the result of Particle size and cesium concentration of March, 2012 samples. The sample which contains the lowest silt fraction was in station-1 and that sediment sample contains about 18% clay, 70% silt and 12% sand and total cesium concentration ( $^{134}\text{Cs}+^{137}\text{Cs}$ ) was 270 Bq/kg. On the other hand the highest silt fraction was found in station-7 and the sediment sample contains as 18% clay, 76% silt and 6% sand and total cesium concentration ( $^{134}\text{Cs}+^{137}\text{Cs}$ ) was 449 Bq/kg. The average median size of Arakawa River sediment was 14.07  $\mu\text{m}$ .

# Particle Size and Cesium Distribution in Arakawa River

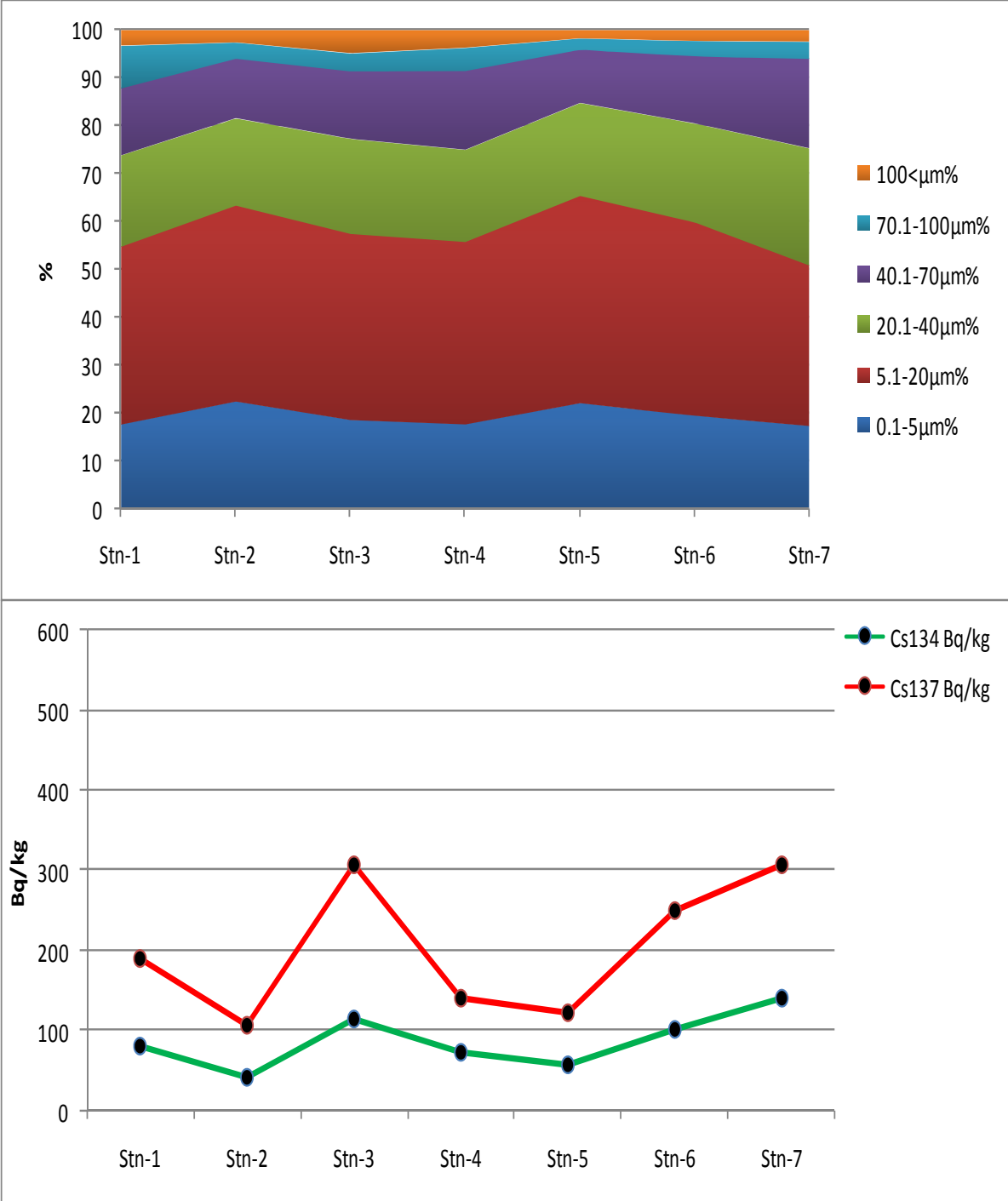


Figure 28 : Particle Size and Cesium Distribution in Arakawa River

#### **4.9.2 Sediment Particle Size and Cesium distribution in Edogawa River**

Particle size of the sediment sample of Edogawa River was measured to know the relationships between particle size and total cesium concentration, figure-29 shows the result of Particle size and cesium concentration of March, 2012 samples. In Edogawa River the lowest total cesium concentration ( $^{134}\text{Cs}+^{137}\text{Cs}$ ) 67 Bq/kg was found in station-10 and the particle size fraction found as 27% clay, 72% silt and 1% sand. On the other hand the highest total cesium concentration ( $^{134}\text{Cs}+^{137}\text{Cs}$ ) 1856 Bq/kg was found in station-15 and the particle size fraction found as 16% clay, 79% silt and 5% sand. The average median size of Edogawa River sediment was 14.47  $\mu\text{m}$ .

# Particle Size and Cesium Distribution in Edogawa River

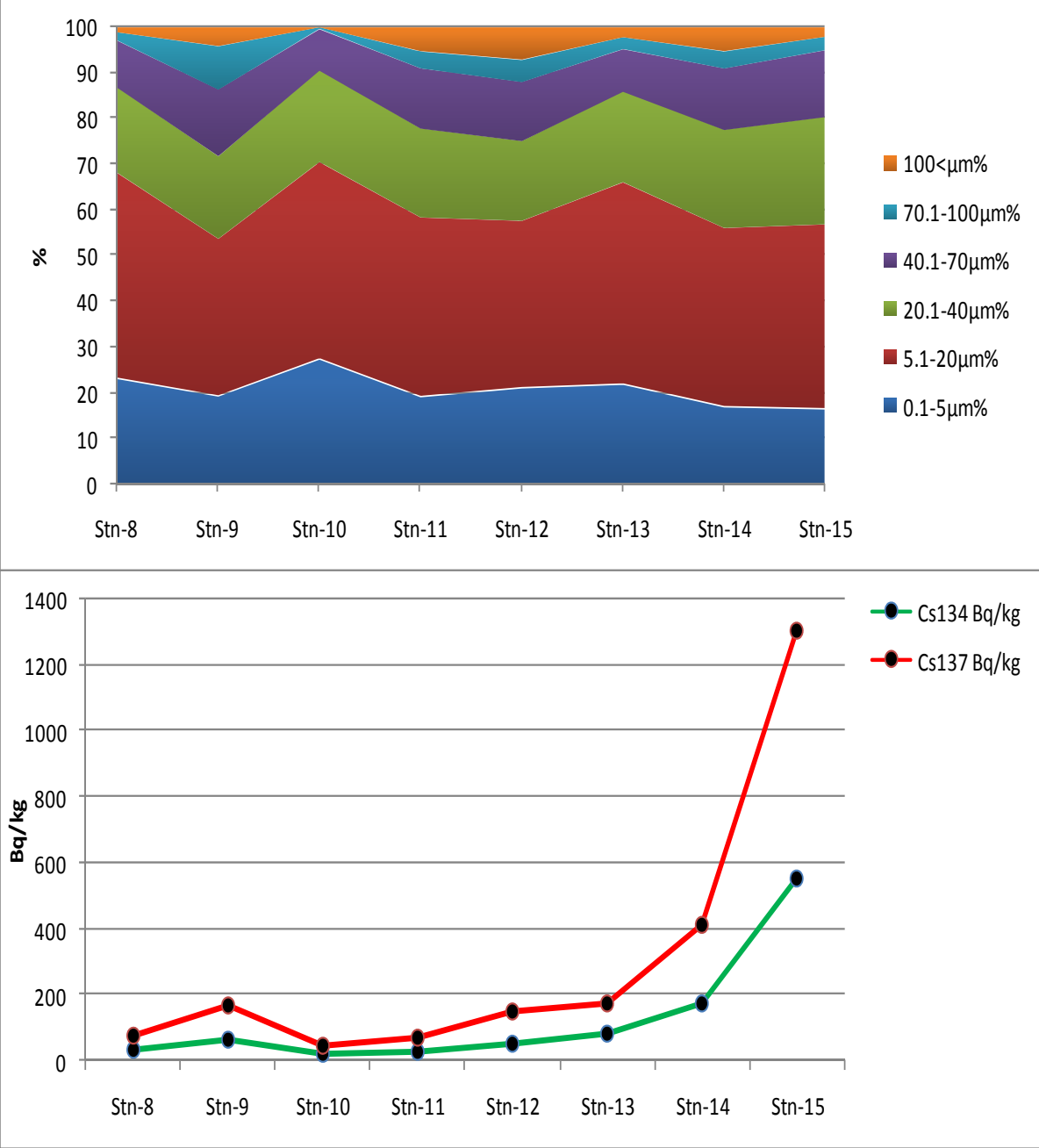


Figure 29 : Particle size and Cesium Distribution in Edogawa River.



#### 4.9.3 Sediment Particle Size and Cesium distribution in Sumidagawa River

Particle size of the sediment sample of Sumidagawa River was measured to know the relationships between particle size and total cesium concentration, figure-30 shows the result of Particle size and cesium concentration of March, 2012 samples. On an average the sediment samples contain about 25% clay, 73-74% silt and 1-2% sand in most of the stations of Sumidagawa River estuary. Highly cesium contained sediment found in the upper stream area in station 19, 20, 21, 22 and 23 and their result are as 25.5% clay, 74% silt and 0.5% sand in station-19 and total cesium concentration ( $^{134}\text{Cs}+^{137}\text{Cs}$ ) was 260 Bq/kg, 34.5% clay, 63.45% silt and 0.05% sand in station-20 and total cesium concentration ( $^{134}\text{Cs}+^{137}\text{Cs}$ ) was 265 Bq/kg, 21.5% clay, 72.5% silt and 6% sand in station-21 and total cesium concentration ( $^{134}\text{Cs}+^{137}\text{Cs}$ ) was 343 Bq/kg, 16.5% clay, 72.5% silt and 11% sand in station-22 and total cesium concentration ( $^{134}\text{Cs}+^{137}\text{Cs}$ ) was 299 Bq/kg, 21% clay, 75.5% silt and 3.5% sand in station-23 and total cesium concentration ( $^{134}\text{Cs}+^{137}\text{Cs}$ ) was 469 Bq/kg, 24% clay, 75% silt and 1% sand in station-0 which located in the Tokyo Bay and total cesium concentration ( $^{134}\text{Cs}+^{137}\text{Cs}$ ) was 170 Bq/kg . The average median size of Sumidagawa River sediment was 10.34  $\mu\text{m}$ .

# Particle Size and Cesium Distribution in Sumidagawa River

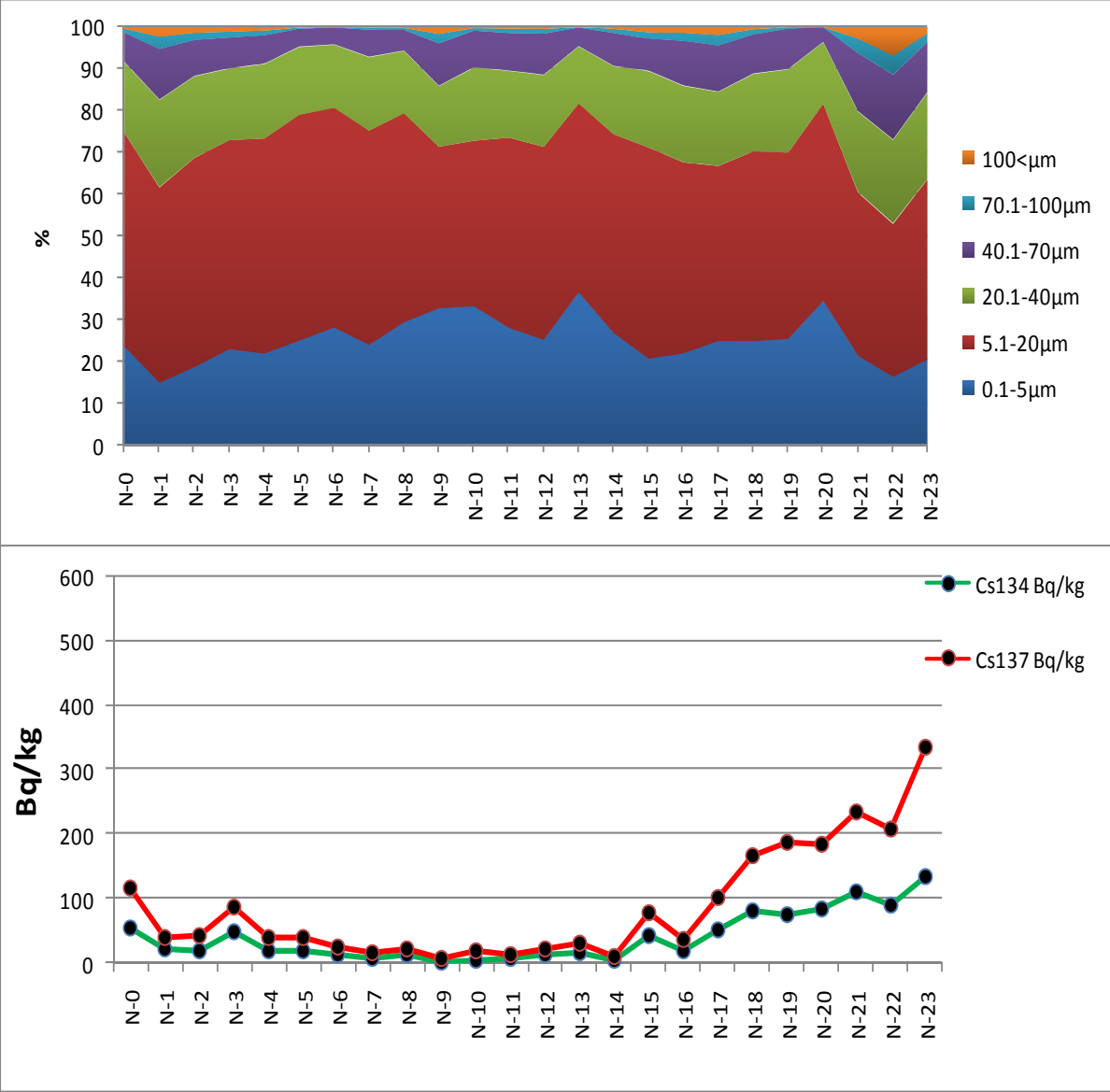


Figure 30 : Particle size and Cesium Distribution in Sumidagawa River.

## **4.10 Cesium Distribution Relationship with Water Quality Parameters**

### **4.10.1 Cesium Distribution Relationship with Water Quality Parameters in Arakawa River**

It was assumed that there was a relationship of turbidity and salinity with cesium distribution. To prove that assumption the water quality data was compared with the cesium distribution. In Arakawa River there were seven sampling points (from station 1 to station 7). Sediment samples were taken to observe the cesium distribution along with the water quality parameters of salinity and turbidity were also observed. The result of the water quality parameters and cesium distribution in Arakawa River had shown in the figure-31 of September, 2012 and figure-32 of September, 2013. Station 6 was assumed as the river mouth and station 7 is in 3.59 km. downstream area from the river mouth that means within the Tokyo Bay.

In September, 2012, from station 7 to station 3 which is about 5.12 km. upstream area from the river mouth the salinity was around 18 ppt in the surface area and it increases with the depth up to 27 ppt at 4 meter depth from the surface level of the water. But it was 11 ppt in surface and 22 ppt at 4 meter depth in station 2 and 6.6 ppt in surface and 15 ppt at 5 meter depth in station 1 which is about 9 km upstream from the river mouth. The turbidity was very low in all stations except station 3 and it was measured 106 FTU at the bottom of water column where as in other stations it was ranged between 2 to 9 FTU. The total cesium concentration was ( $^{134}\text{Cs} + ^{137}\text{Cs}$ ) 695 Bq/kg in station 7 (-3.59 km.), 607 Bq/kg in station 6 (0 km. River mouth), 395 Bq/kg in station 5 (1.5 km.), 329 Bq/kg in station 4(3.25 km.), 262 Bq/kg in station 3 (5.12 km.), 412 Bq/kg in station 2 (7.27 km.), 240 Bq/kg in station 1(8.97 km.), (Figure-31).

In September, 2013, from station 7 to station 3 which is about 5.12 km. upstream area from the river mouth the salinity was ranged around 11 to 8 ppt in the surface area and it increases with the depth up to 30 ppt at 5 meter depth from the surface level of the water. But it was ranged 3 to 7 ppt up to 2 meter depth and then it reached to 27 ppt at 4 meter depth in station 2 and 3 ppt in surface and it ranged 9 ppt up to 3 meter depth and it increased at depth 4 and 5 meter depth to 27.4 ppt. in station 1 which is about 9 km upstream from the river mouth. The turbidity was very low in all stations from surface to above the sediment bed and it ranged from 4 to 12 FTU but at the bottom of the water column turbidity found as 153 FTU in Station-7, 389 FTU in Station-6, 27 FTU in Station-5, 351 FTU in Station-4, 12 FTU in Station-3, 119 FTU in Station-2, 32 FTU

in Station-1. The total cesium concentration was ( $^{134}\text{Cs}+^{137}\text{Cs}$ ) 110 Bq/kg in station-7 (-3.59 km.), 240 Bq/kg in station-6 (0 km. River mouth), 172 Bq/kg in station-5 (1.5 km.), 523 Bq/kg in station-4(3.25 km.), 129 Bq/kg in station-3 (5.12 km.), 302 Bq/kg in station-2 (7.27 km.), 219 Bq/kg in station-1(8.97 km.), (Figure-32).

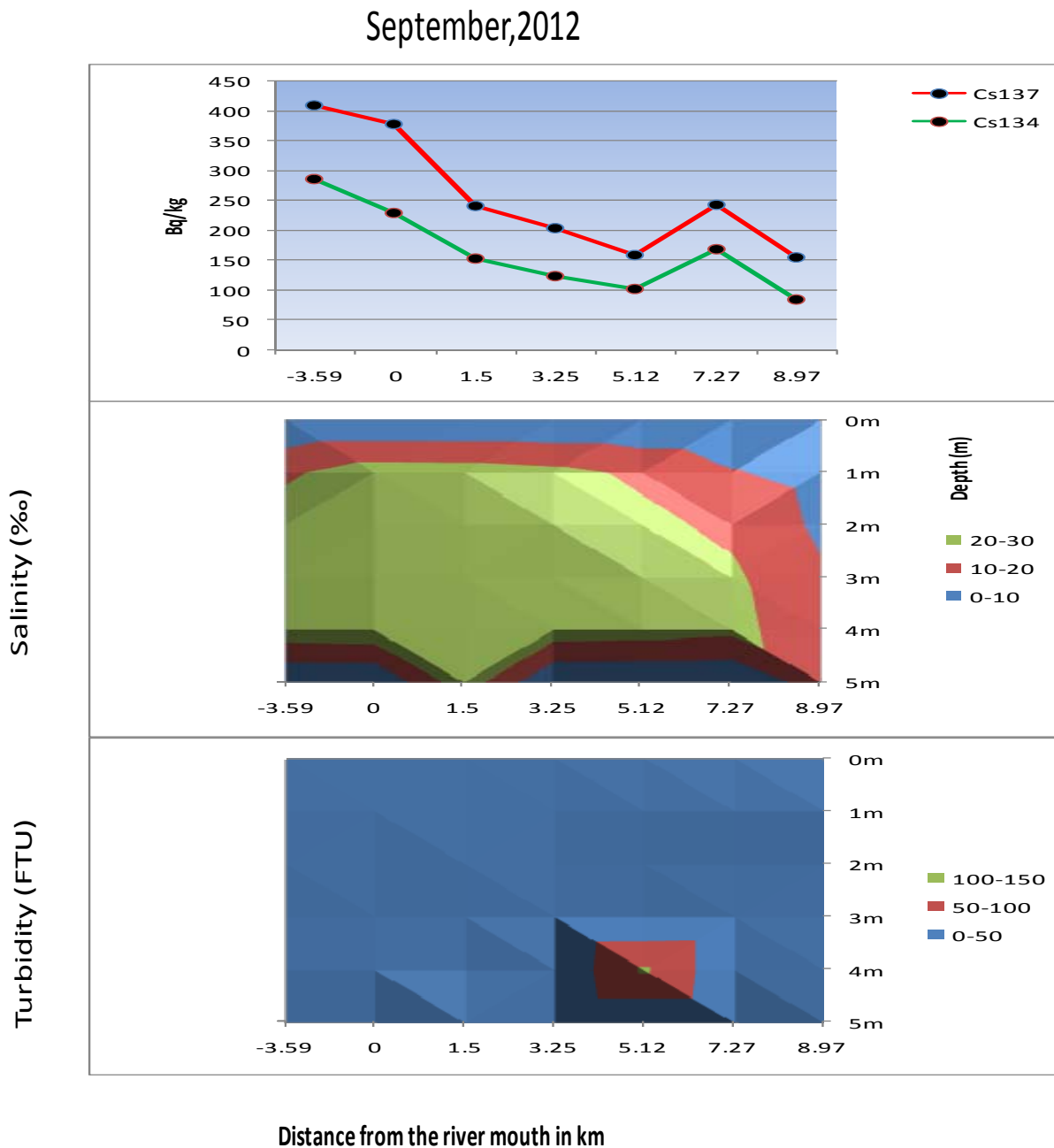


Figure 31 : Cesium Distribution and Water quality parameters in Arakawa River in September, 2012.

## September, 2013

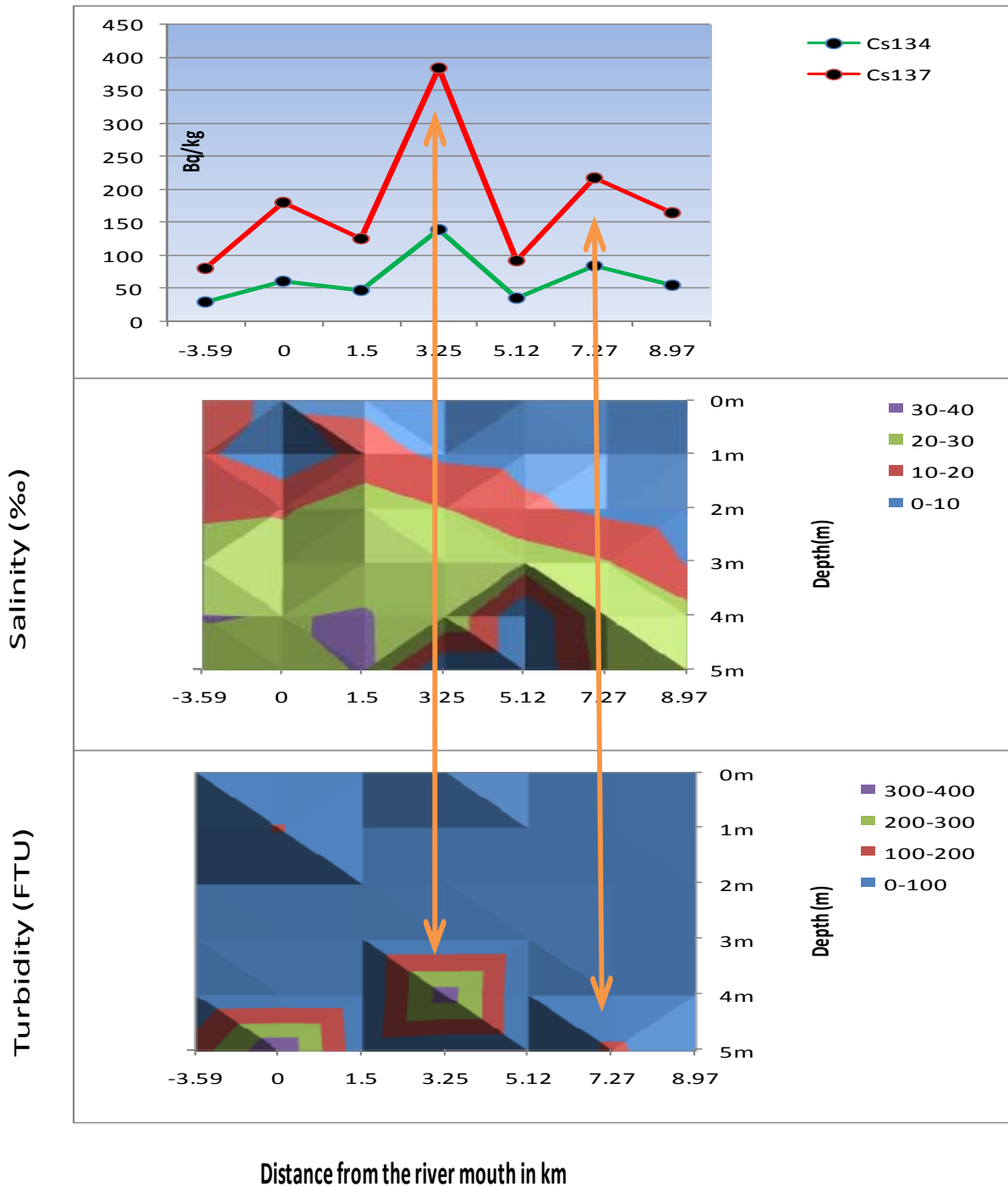


Figure 32 : Cesium Distribution and Water quality parameters in Arakawa River in September, 2013.

#### 4.10.2 Cesium Distribution Relationship with Water Quality Parameters in Edogawa River

It was assumed that there was a relationship of turbidity and salinity with cesium distribution. To prove that assumption the water quality data was compared with the cesium distribution. In Edogawa River there were seven sampling points (from station 9 to station 15). Sediment samples were taken to observe the cesium distribution along with the water quality parameters of salinity and turbidity were also observed. The result of the water quality parameters and cesium distribution in Edogawa River had shown in the figure-33 of September, 2012 and figure-34 of September, 2013. Station-9 was assumed as the river mouth (0 km.) and station-15 is in 11.33 km. upstream area from the river mouth.

In September, 2012, from station-9 to station-10 which is near to river mouth area where the river mix with Tokyo Bay, the salinity was around 19 ppt in the surface area and it increases with the depth up to 25 ppt at 4 meter depth from the surface level of the water column. Salinity was 2 ppt in surface and 23 ppt at 6 meter depth in station 11 and 0.28 ppt in surface and 18 ppt at 4 meter depth in station 12 which is about 5 km upstream from the river mouth. From station 13 to station 15 the salinity was found 0.12 to 0.13 ppt at all depths. The turbidity was very low in all stations except station 13 and it was measured 36 FTU at the bottom of water column where as in other stations it was ranged between 3 to 19 FTU. The total cesium concentration was ( $^{134}\text{Cs}+^{137}\text{Cs}$ ) 110 Bq/kg in station-9 (0 km. River mouth), 92 Bq/kg in station-10 (1.3 km.), 56 Bq/kg in station-11 (3.15 km.), 79 Bq/kg in station-12 (5.01 km.), 181 Bq/kg in station-13 (7.47 km.), 237 Bq/kg in station-14 (9.81 km.), 137 Bq/kg in station-15(11.33 km.), (Figure-33).

In September, 2013, from station 9 to station 11 which is near to river mouth area where the river mix with Tokyo Bay, the salinity was around 0.4 to 6 ppt in the surface area and it increases with the depth up to 31 ppt from the surface level of the water column. Salinity was 0.10 ppt in surface and 1.75 ppt at 6 meter depth in all stations from station-12 to station-15. The turbidity was very low in all stations above the bottom of sediment bed and ranged from 5 to 11 FTU. But at the bottom of the water column turbidity found as 19 FTU in Station 9, 98 FTU in Station-10, 60 FTU in Station-11, 42 FTU in Station-12, 72 FTU in Station-13, 353 FTU in Station-14, 144 FTU in Station-15. The total cesium concentration was ( $^{134}\text{Cs}+^{137}\text{Cs}$ ) 170 Bq/kg in station-9 (0 km. River mouth), 18 Bq/kg in station-10 (1.3 km.), 24 Bq/kg in station-11 (3.15 km.), 42 Bq/kg

in station-12 (5.01 km.), 33 Bq/kg in station-13 (7.47 km.), 188 Bq/kg in station-14 (9.81 km.), 688 Bq/kg in station-15(11.33 km.), (Figure-34).

September,2012

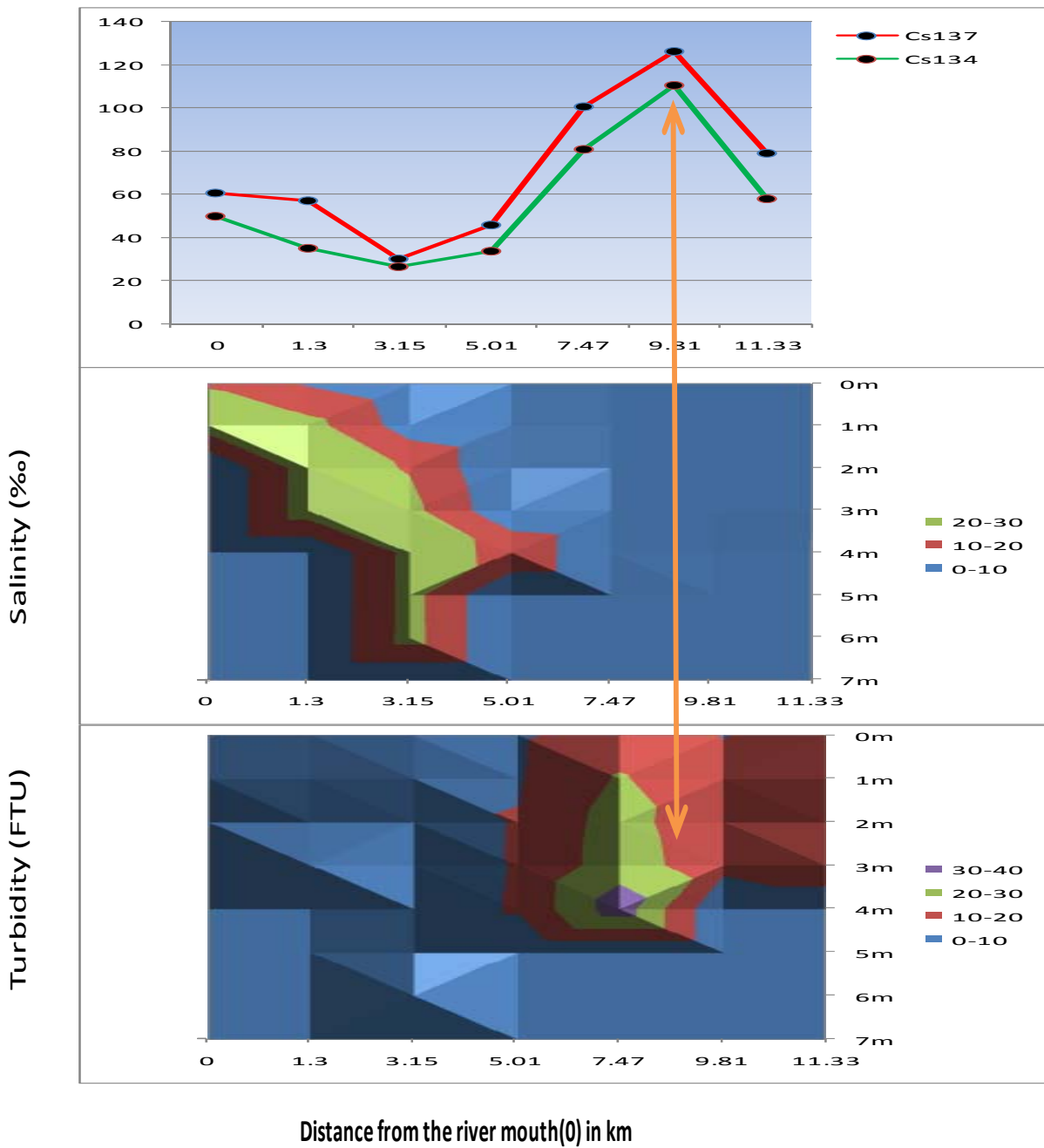


Figure 33 : Cesium Distribution and Water quality parameters in Edogawa River in September, 2012.

September, 2013

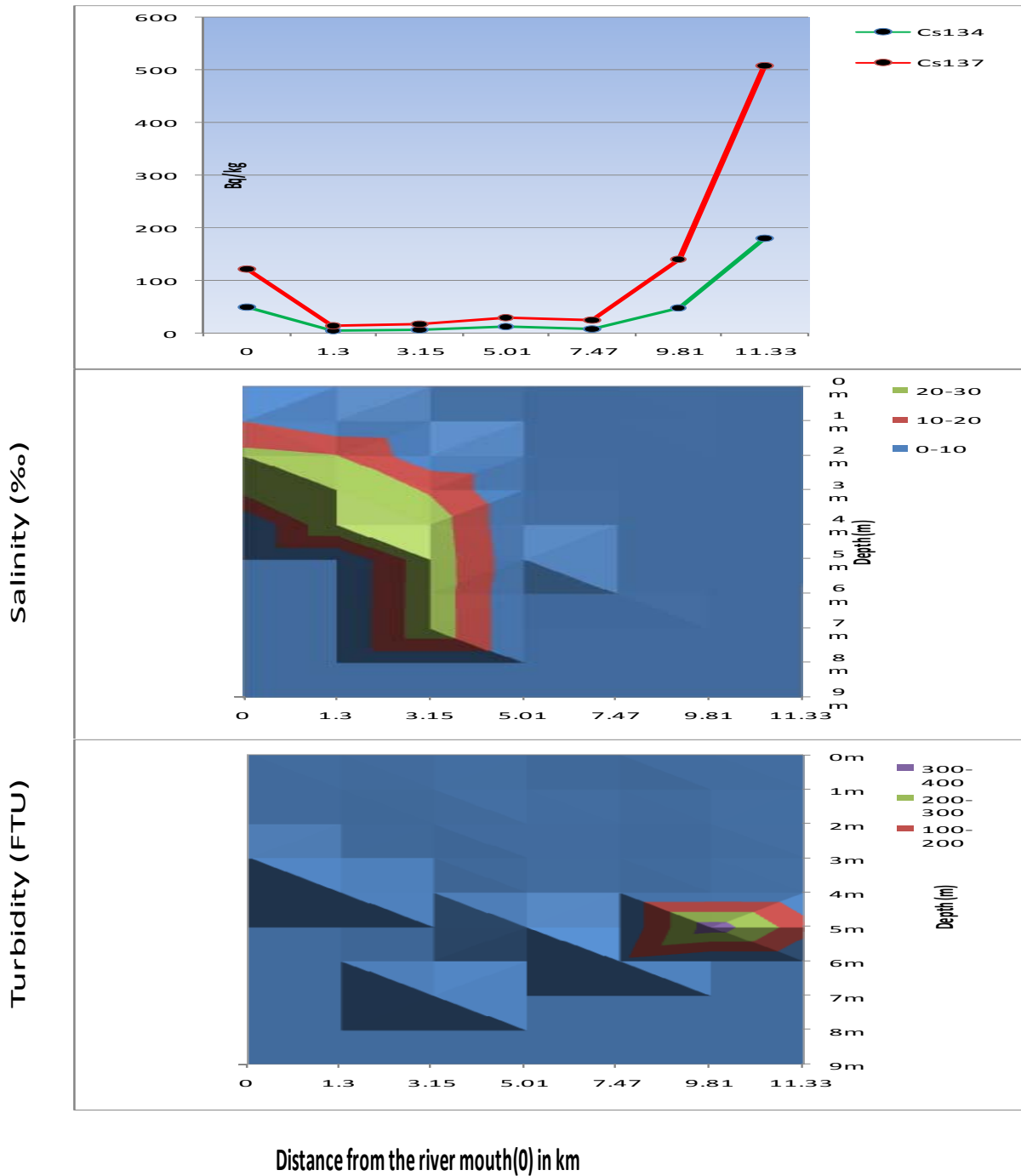


Figure 34 : Cesium Distribution and Water quality parameters in Edogawa River in September, 2013.



#### 4.11 Suspended Solid Concentration (SSC) Relationship with Water level and Cesium Concentration

To obtain the relationship of Cesium concentration with water level and also with the SSC (suspended solid concentration), the water sample were taken 12 times after rainfall event from November, 2012 to September, 2013 from Edogawa River at Noda Bridge Point. The SSC experiment results had shown in figure-35. The water level varied from 0.03 meter to 5.16 meter. The lowest water was observed on December, 17, 2012 that was 0.03 m. and at that time the SSC was found 7.22 mg/l and the total cesium concentration ( $^{134}\text{Cs}+^{137}\text{Cs}$ ) was 3951 Bq/kg. The highest water level was observed on September, 17, 2013 that was 5.16 m. and the total cesium concentration ( $^{134}\text{Cs}+^{137}\text{Cs}$ ) was 323 Bq/kg. After plotting for the regression analysis between the Cesium concentration with water level, R squared value is 0.696 and following with SSC the R squared value is 0.878. In case of the relationship between water level with SSC and water level with cesium concentration when the water level was same. Figure-36 shown the time series of calculated the SSC and cesium concentration according to relationship found through figure-19 using the same water level for both of them.

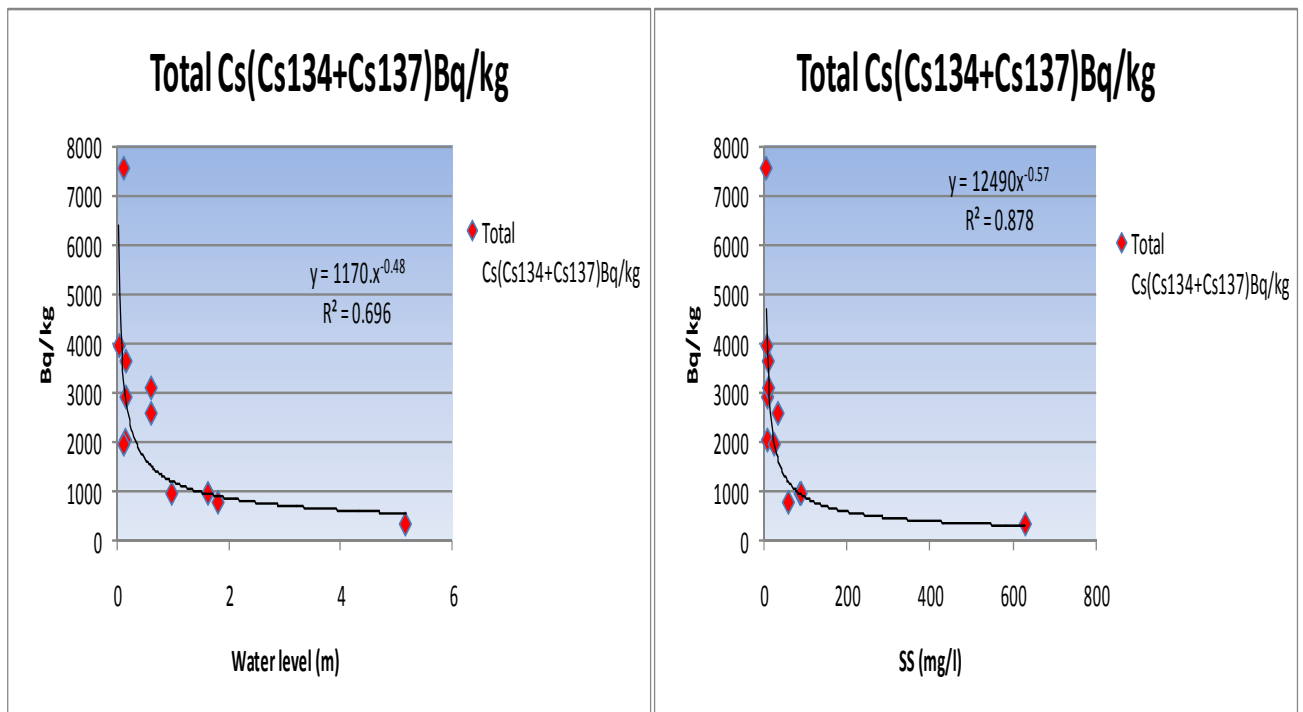


Figure 35 : The relationship of Cesium concentration with water level and with the SSC (suspended solid concentration) in Edogawa River.

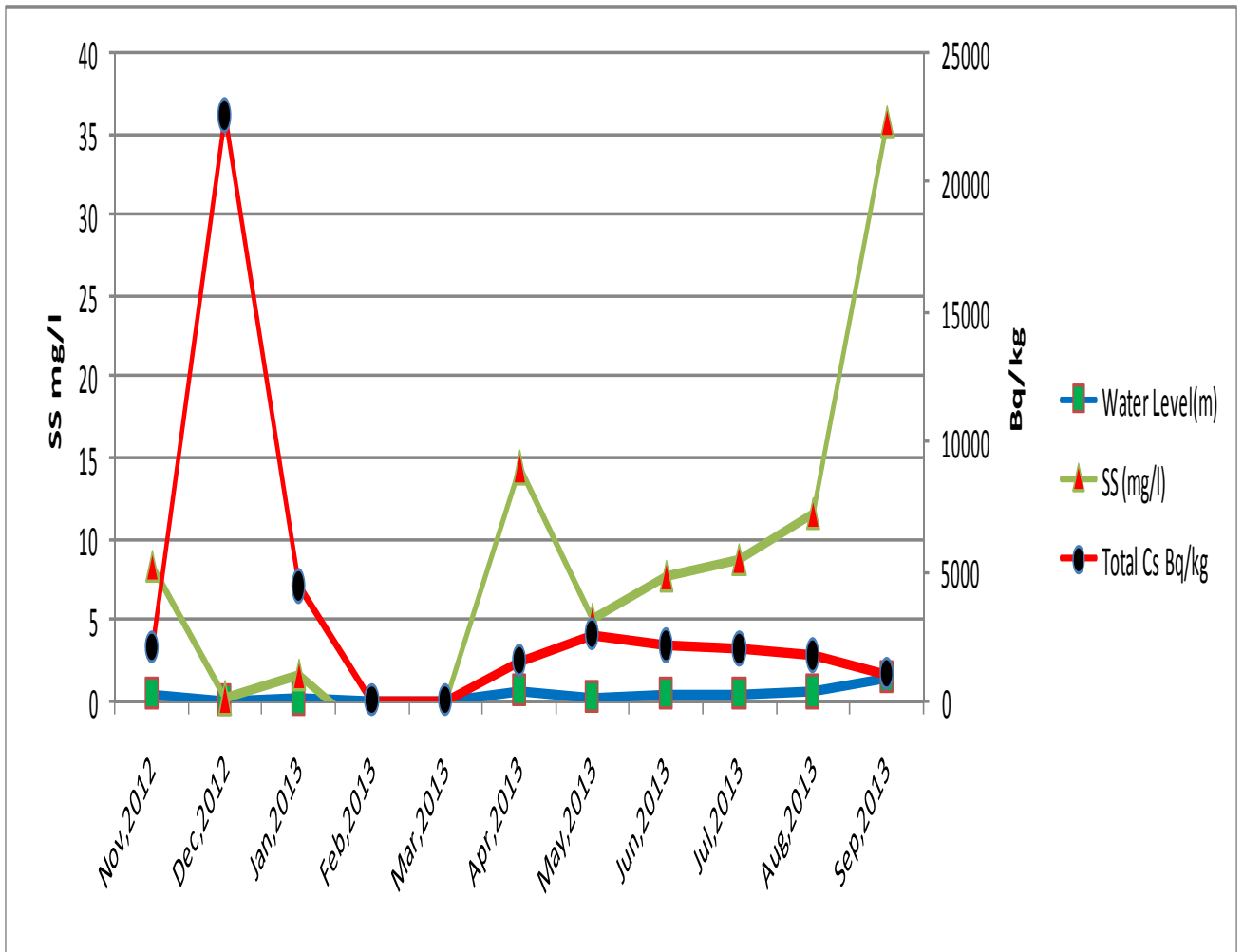


Figure 36: Time series of the relationship change of Cesium concentration with water level and SSC (Suspended Solid Concentration) in Edogawa River.

#### 4.12 Suspended Solid (SS) Flux and Total Mass of Radio-Cesium

For calculating of Suspended Solid Discharge or Load to get daily suspended-solid discharges the Eq-9 bellow was used.

$$Q_s = Q_w C_s \dots\dots\dots 9$$

Where,

$Q_s$  = suspended-sediment discharge, in tons per day or metric tons per day

$Q_w$  = water discharge, cubic meters per second

$C_s$  = mean concentration of suspended sediment in the cross-section in milligrams/liter

The water level data of Arakawa River and Edogawa River was available from the website of River Department of Japan (<http://www1.river.go.jp>). In case of Arakawa River the data was taken from the Iwabuchi Sluice Observatory and in case of Edogawa River the data was taken from Noda Bridge Observatory.

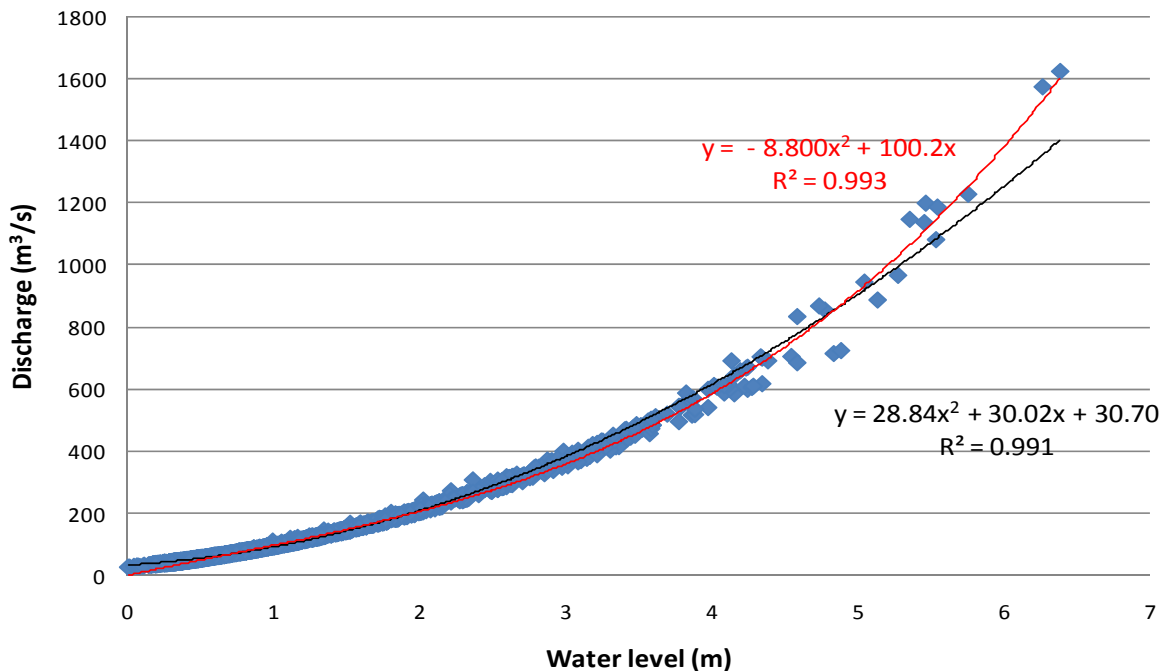
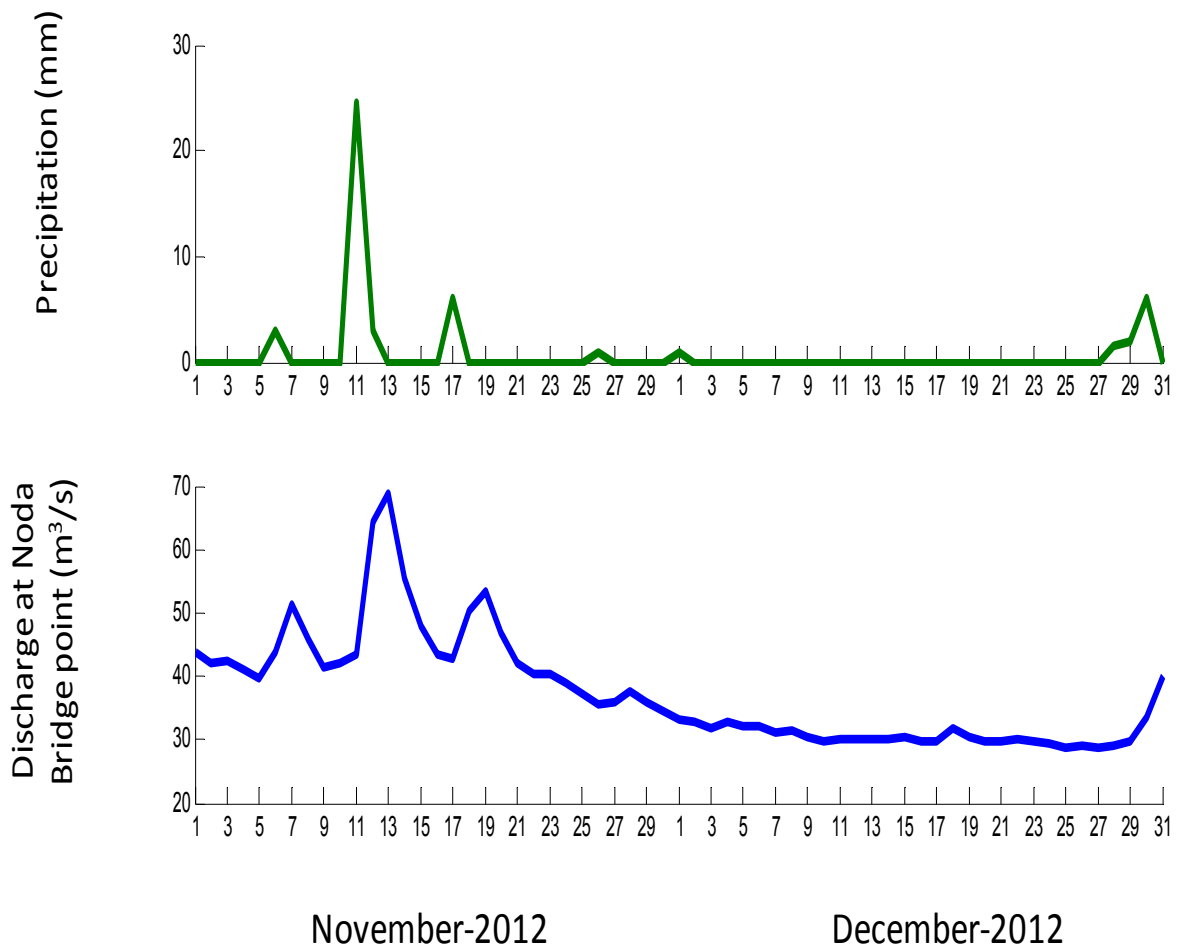


Figure 37 : H-Q Curve For Edogawa River

To calculate the water discharge rate and understand the discharge pattern, using the ten years water level and discharge data from Noda observation station and H-Q curve was made for Edogawa River and the r-squared value was 0.991, the regression equation from the H-Q curve (Figure-37) was used for Edogawa River and also another H-Q curve (Tanaka, Y. et al.,2007) for Arakawa River was used and the r-squared value was 0.904, to get the water discharge data in these two rivers and thus calculated the suspended solid flux. Figure-38 showed that the calculated water discharge data of Edogawa River of November, 2012 and December, 2012 were consistent with the precipitation data which were taken from the Isesaki rain gauge station within the watershed of Edogawa River.



**Figure 38 : Time series of Precipitation and Calculated Water Discharge.**

The calculated water discharge for Arakawa River was almost same in 2011(81.33 m<sup>3</sup>/s) and in 2012 (80.40 m<sup>3</sup>/s) and the mean suspended solid concentration was 10 mg/l but there was variation of water discharge in 2011 (110.72 m<sup>3</sup>/s) and in 2012 (78.90 m<sup>3</sup>/s) in Edogawa River and the mean suspended solid concentration (SSC) was 23 mg/l. in Arakawa River the SS flux were almost same in 2011 and 2012, it was about 0.3 million ton/year while in Edogawa River it was about 0.9 million ton in 2011 and 0.65 million ton in 2012 (Figure-39).

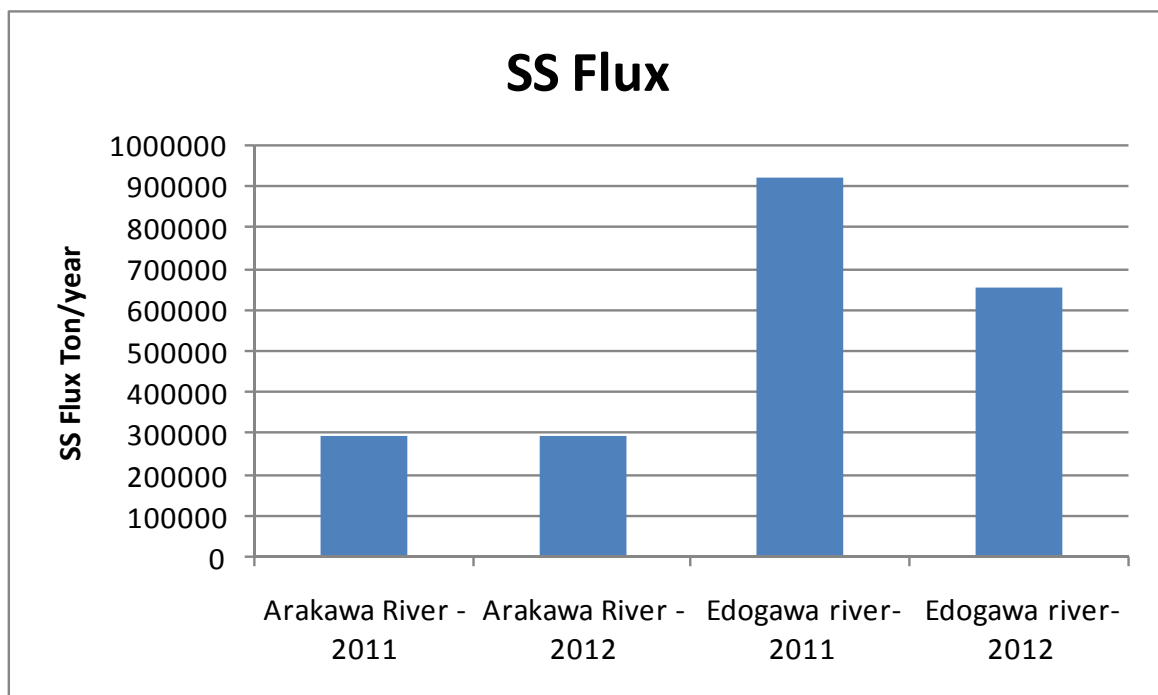


Figure 39 : SS Flux in Arakawa and Edogawa River, 2011 and 2012.

It was assumed that the contaminated sediment is moving towards downstream or in Tokyo Bay. To understand the transportation of cesium contaminated sediment in Arakawa and Edogawa River the total mass of cesium was estimated by using the data of vertical profile of cesium contained sediment. There were two core sampling points, one in upstream and another in downstream area in both Arakawa and Edogawa River. There were area data between each and every sampling points and also had the vertical profile results then the total mass of cesium were calculated by following way and integrated all layers.

Area between two sampling points= A

Depth of the contaminated sediment=D

Volume of the contaminated sediment,  $V = A \cdot D$  (m<sup>3</sup>)

Mass of the sediment =  $V \cdot 1.8$  Ton (A coefficient of 1.8 was used to get mass of sediment from sediment volume)

Cesium concentration was in Bq/kg then converted to cesium concentration in the unit of G Bq (Giga Becquerel).

Figure- 40 shows that left hand side, Arakawa River in September, 2012 the total mass of cesium was 1998 G Bq, it increased to 3031 G Bq in November, 2012 then it started decreasing in July, 2013 it was 837 G Bq and right side of figure-37 was the result of Edogawa River, in September, 2012 the total mass of cesium was 1800 G Bq it was increased and reached to 3272 G Bq in March, 2013 but again decreased to 1290 G Bq in July, 2013.

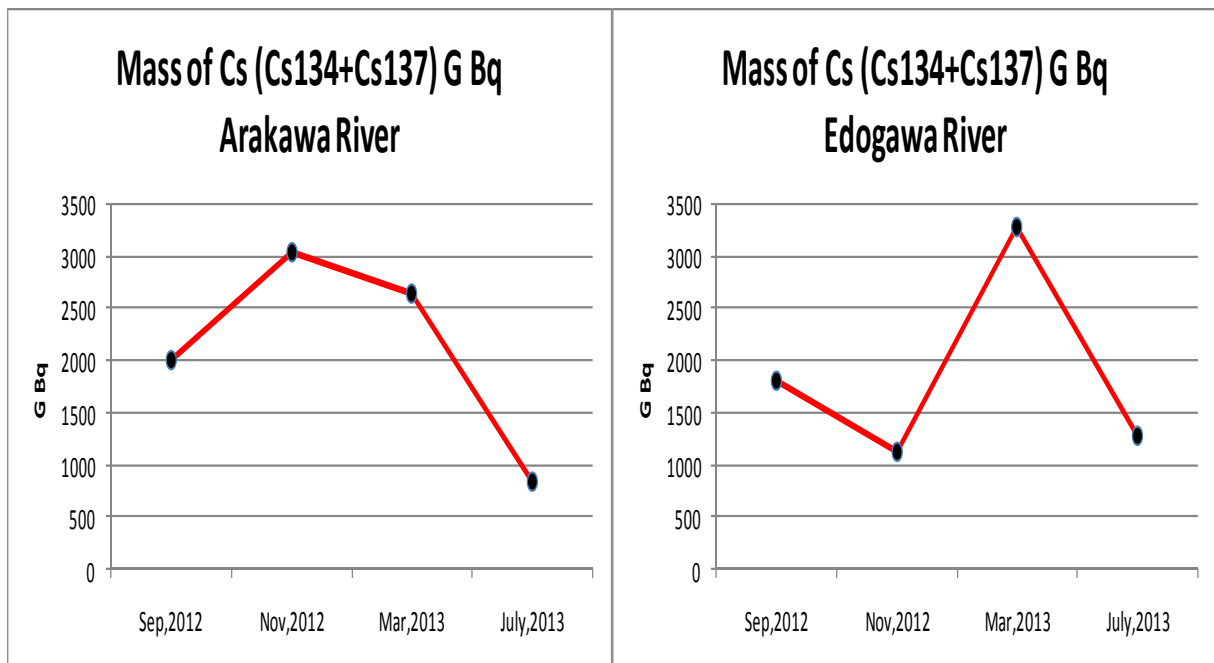


Figure 40 : Mass of Cesium in Arakawa and Edogawa River.

## Chapter 5: Discussion

### 5.1 Radio-Cesium ( $^{134}\text{Cs}$ and $^{137}\text{Cs}$ ) distribution in Tokyo Bay

There was same trend of low concentration except in the month of October 2012 in station 98 which is Northern part of Tokyo Bay and Arakawa, Edogawa and Sumidagawa river fall in this area and in the month of December 2012 in station 99 which is North –Eastern part of Tokyo Bay. This could be happen after typhoon season when large amount of sediment could reach to the estuary from the upper stream region. But again the peak disappears in the next dry season due to mixing with sediment.

To verify the result of this study, the result of this study was compared with the result of Japan Ministry of Education, Culture, Sports, Science and Technology (MEXT) measurement of radio cesium in the sediment of Tokyo Bay and the result ranging from 1.6 to 130 Bq/kg and 0.68 to 62 Bq/kg in case of  $^{137}\text{Cs}$  and in case of  $^{134}\text{Cs}$  respectively (Figure-41), which is quite similar with the result of this study except station 98. The cause of difference in station-98 may be due to the differences in getting the samples from exact sampling location.

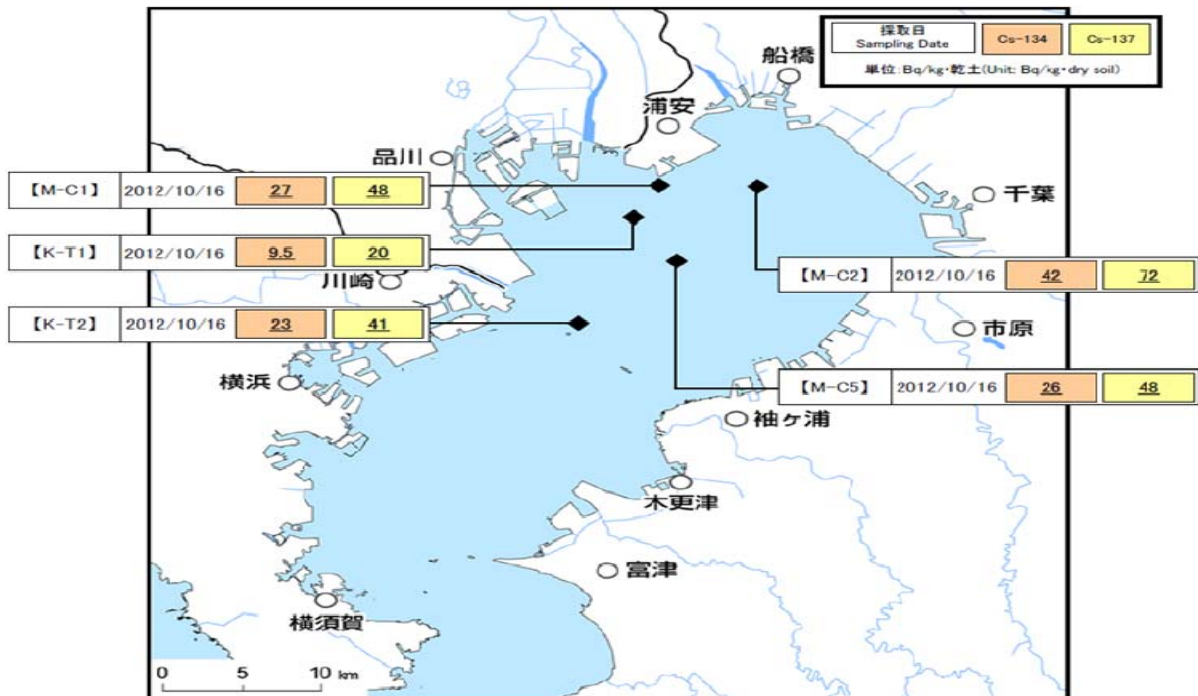


Figure 41 : MEXT Result of Tokyo Bay Sediment of Nov, 2012.

## 5.2 Radio-Cesium ( $^{134}\text{Cs}+^{137}\text{Cs}$ ) distribution in Arakawa, Edogawa and Sumidagawa River

To understand the fate of the contaminated sediment in the river and Tokyo Bay basin it was important to get the information about the cesium distribution. In Arakawa River, cesium concentration peak in the surface sediment was moving towards the downstream region from upstream region. The low concentration in the upstream area was the indication of decreasing the supply of contaminated sediment from the upper most stream region. Just after the typhoon on September 15, 2013 the new peak appeared in the station-2 and station-4 which were 7.27 km. and 3.25 km. upstream area from the river mouth. So huge water flow and rainfall event brought some new contaminated sediment into the water system or re-suspension occurred and or both were happened in the same time. In case of Edogawa River the same trend was observed. In November 2012 the upstream area at 11.33 km from the river mouth the concentration was suddenly increased to 1010 Bq/kg ( $^{134}\text{Cs}+^{137}\text{Cs}$ ). There are water flow regulation structures (Edogawa water gate and wired bridge) in that area between station-14 and station-13 which may contributed for the trapping of contaminated sediment in that site. But the concentration was also decreasing as like as Arakawa River and in July 2013 it downed to 693 Bq/kg ( $^{134}\text{Cs}+^{137}\text{Cs}$ ). After the typhoon on September 15, 2013 the contaminated sediment was transported to river mouth region and slight peak was appeared there. That means sediment was transported to the river mouth and Bay area.

**Table 11: Water Flow Rate, Total Water Discharge and Precipitation Data of Arakawa and Edogawa River.**

River Name	year	water flow rate m <sup>3</sup> /s (calculated)	yearly total discharge million m <sup>3</sup> (calculated)	Yearly total Precipitation (mm)
Arakawa	2011	81.33	2529.83	1469.5 (Chichibu)
Arakawa	2012	80.4	2500.91	1207.7 (Chichibu)
Edogawa	2011	110.72	3444.03	1459 (Iseaki)
Edogawa	2012	78.9	2454.17	1060.5 (Iseaki)

There are effect of water flow rate and discharge in the sediment transport process in the Arakawa River and Edogawa River. Table-11 shows that in 2012 the precipitation is about 27% less than year 2011 in the Edogawa river catchment area and in Arakawa river catchment area that is 17% less than 2011. In case of flow rate there was an about 32 m<sup>3</sup>/s difference in Edogawa River in 2011 and 2012 but in Arakawa River the flow rate was almost same..



Analyzing the cesium distribution data, it seemed that the sediment transport in Arakawa River is relatively large due to river topography and water discharge pattern but in Edogawa River it was relatively small and it usually happened only after large rainfall and typhoon effect.

The samples were taken four times in Sumidagawa River and estuary. First in August-2012 followed to December, 2012, March, 2013 and September-2013. First time in August, 2012, from 17 points samples were collected though the total sampling points were 24 in this river and estuary. But next three time samples were collected from all 24 sampling points. Actually this river is a branch of Arakawa River and the result showed that still the contaminated sediment was remained in the upstream region than river mouth region though the radio-cesium concentration activity is decreasing by time like other river sediment. The most noticeable thing was that the radio-cesium concentration activity in middle area of this river and estuary was always very low and it was less than 50 Bq/kg.

### **5.3 Vertical Distribution of Radio-Cesium ( $^{134}\text{Cs}$ and $^{137}\text{Cs}$ ) in Arakawa, Edogawa and Sumidagawa River**

Vertical profile provided the information of deposition layer, diffusion and advection in vertical direction. This information also used to calculate contaminated sediment mass and cesium mass in the study area. In Arakawa River from two locations, station-4 which is 3.25 km. upstream from the river mouth and another station-7 which is 3.59 km. downstream from the river mouth means within the Tokyo Bay, the core samples were taken to observe the vertical profile, sedimentation and diffusion activity. From station-4 the cores were taken four times September, 2012, November, 2012, March, 2013 and July, 2013. In September, 2012 there was a clear peak in the depth of 8-10 cm and the total Cesium concentration ( $^{134}\text{Cs}+^{137}\text{Cs}$ ) was 1683 Bq/kg. in November the peak moved to more deeper in 18-22 cm and the total Cesium concentration ( $^{134}\text{Cs}+^{137}\text{Cs}$ ) was 1179 Bq/kg, in March, 2013 the peak again moved to 8-10 cm depth and the total Cesium concentration ( $^{134}\text{Cs}+^{137}\text{Cs}$ ) was 1207 Bq/kg and in July, 2013 the peak moved to more upward 2-4 cm depth and the total Cesium concentration ( $^{134}\text{Cs}+^{137}\text{Cs}$ ) was 824 Bq/kg. In November, 2012 there some sedimentation occurred but that the sediment movement from the sediment bed happened and the peak again back to the 8-10 cm depth in March, 2013 and it

continued and in July, 2013 the peak was moved to 2-4 cm depth (Figure- 13). In station-7 which was in the 3.59 km. downstream from the river mouth and here the peak is always in 2-4 cm depth (Figure- 14) from the sediment bed and not moved. Sedimentation is very small in this area or the sediment was passing deeper part of the Tokyo Bay.

In Edogawa River from two locations, station-9 which is assumed as the river mouth where it falls in the Tokyo Bay and another station-14 which is located 9.81 km. upstream from the river mouth. The core samples were taken to observe the vertical profile, movement and diffusion activity. The cores were taken four times September, 2012, November, 2012, March, 2013 and July, 2013. First time in September, 2012 in station-9 there were two clear peaks in the vertical profile first one was in the depth 10-14 cm and second one was in 4-6 cm depth. In November also there were two peaks but the peaks moved to deeper area and first one was in 14-18 cm depth and second one 8-10 cm depth. In March, 2013 the radio-cesium concentration activity was increased. In July the second peak again moved slightly upwards to 6-8 cm depth but the first peak was in same depth 14-18 cm (Figure-15). Only in this point two clear peaks were observed every time, this information can be interpreted as the first peak that was in deeper layer may denote the accumulation of radio-cesium through atmospheric fallout just after the FDNPP (Fukushima Daichi Nuclear Power Plant) accident in March, 2011 and the second peak may denote sediment accumulation in 2012 through sediment transport and sediment wash off. Here also seemed that some movement of surface sediment happened like Arakawa River. Other place named station-14 in Edogawa River where from core samples were also observed. There was also clear sharp peak observed in the depth of 10-14 cm in September, 2012 and also in the same depth in July, 2013 though the sharpness of the peak was disappear means some diffusion activity occurred here.

In Sumidagawa River from two locations core samples were taken, one was station-20 which was located in the upstream region from the river mouth and another station-17 which was also located in upstream region from the river mouth (Figure-5). The core samples were taken to observe the vertical profile, movement and diffusion activity. There was a peak up to 8-10 cm depth in December, 2012 and the peak was in same depth in September, 2013 but the radio-cesium activity concentration decreased about half of December, 2012 (Figure-17). In station-17

there also found two clear peaks in the depth first one 8-10 cm depth and second one 4-6 cm depth (Figure-18) and this phenomena also can be interpreted as station-9 of Edogawa River.

#### **5.4 Diffusion Coefficient and Advection**

Vertical profile was used to get the Diffusion coefficient and advection rate in all six cores sampling points of three rivers. In general the most of the contaminated sediment accumulated in the depth between 8-14 cm except the station-7 of Arakawa River which is actually situated within the Tokyo Bay 3.59 km. downstream from the river mouth and here the peak was always in 2-4 cm depth. In the upstream part of Arakawa River Station-4 was less diffusion coefficient (0.03) than Edogawa River station-14 (0.17). Also in the downstream part of Arakawa River Station-7 was less diffusion coefficient (0.0001-0.07) than Edogawa River station-9 (0.48-0.7). In Sumidagawa River the Diffusion coefficient found 0.2 and here also most of the contaminated sediment accumulated in 8-10 cm depth.

For advection there was large variation of coefficient in upstream area station-4 of Arakawa River and it varied from -0.03 to 1 but in Edogawa River station-14 upstream area it was 0.02. In downstream area station-7 and station-9 of Arakawa and Edogawa River the coefficient were 0.001-0.04 and 0.15-0.98 respectively. The R squared value of observed and estimated value (Table-10) indicated that in station-4 of Arakawa River sedimentation was dominant in vertical direction in Nov, 2012 after that sedimentation was upward direction but sedimentation was dominant over diffusion (Figure-14). In station-7 downstream of Arakawa River the sedimentation and diffusion was not so active (Figure-15). In station-9 downstream of Edogawa River the R squared value was for 0.256 diffusion and R squared value was 0.698 for sedimentation, here thus sedimentation was dominant over diffusion (Figure-16). In station-14 upstream of Edogawa River the R squared value was 0.964 for diffusion and R squared value was 0.654 for sedimentation thus here the diffusion was dominant over sedimentation and the peak of cesium concentration was not moved (Figure-17). Thus according to table-10 it was observed that sedimentation was dominant in winter or dry season when the precipitation was less 33.5 mm in Arakawa water shade and 37.5 mm in Edogawa water shade but diffusion became dominant or increase than in dry season in summer of wet season when precipitation was

large 139.5 mm in Arakawa water shade and 148 mm in Edogawa water shade. Diffusion increased with the precipitation on the other hand sedimentation increased in dry season.

### **5.5 Particle size and Radio-cesium Distribution**

The particle size of the sediment of Arakawa River almost uniform in all stations and consist of about 18-22% clay, 68-76% silt and 5-12% fine sand. The average median size of the particles was 14.07  $\mu\text{m}$  and radio-cesium concentration ranges from 148 - 422 Bq/kg. In case of Edogawa River the sediment particle size consists of about 16-27% clay, 59.5-79% silt and 1-13.5%. The average median size of the particles was 14.47  $\mu\text{m}$  and radio-cesium concentration ranges from 67 Bq/kg to 1856 Bq/kg. In case of Sumidagawa River the sediment particle size consists of about 16.5-34.5% clay, 55.5-75.5% silt and 0.5-11% fine sand. The average median size sediment was 10.34  $\mu\text{m}$  and radio-cesium concentration ranges from 7 Bq/kg to 469 Bq/kg. It was difficult to conclude that which particle size may contain the more radio-cesium because the same particle size ratio sediment containing less Cs concentration in one station but more in another station. The spatial difference may have an impact on this fact and the capacity of particles to absorb radio-cesium maybe another factor that could be better to interpret this issue. But it was observed that, total cesium concentration ( $^{134}\text{Cs}+^{137}\text{Cs}$ ) were high where the silt contents were high, for example in Arakawa River the silt content was highest 76% in station-7, the total cesium concentration ( $^{134}\text{Cs}+^{137}\text{Cs}$ ) was highest 449 Bq/kg similarly in Edogawa River the silt content was highest 79% in station-15, the total cesium concentration ( $^{134}\text{Cs}+^{137}\text{Cs}$ ) was highest 1856 Bq/kg, also in Sumidagawa River the silt content was highest 75.5% in station-23, the total cesium concentration ( $^{134}\text{Cs}+^{137}\text{Cs}$ ) was highest 469 Bq/kg. In another case, the radio-cesium concentration ( $^{134}\text{Cs}+^{137}\text{Cs}$ ) in suspended solid was 22,000 Bq/kg, in November, 2012 and the suspended solid consist of about 99.18% clay and only 0.82% silt with no sand particles in Edogawa River. From this point of view it was clear that radio-cesium activities were greater in fine sediments.

As stated by Walling and He (1996) that, radionuclide activities in suspended sediment will be greater than those in equivalent source materials, due to the selective mobilization of fines.

## **5.6 Water Quality and Radio-cesium Distribution**

The relationship with the water quality parameters as salinity and turbidity with the cesium distribution was clear in the result that radio-cesium concentration increased with the turbidity but the relation with salinity is not clear and so salinity had no effect on the concentration activity of the radio-cesium in the sediment (Figure-31 to 34).

## **5.7 Suspended Solid Concentration (SSC) and Radio-cesium Distribution**

The relationship between suspended solid and cesium concentration could be an indicator of the supply of contaminated sediment from adjacent and upstream region. With the increase of water level suspended solid concentration increased exponentially and the radio-cesium concentration decreased same way. The sediment those are coming this time to the water system were less contaminated with radio-cesium or contain less amount of radio-cesium. According the result of time series of the relationship changes of Cesium concentration with water level and SSC (Suspended Solid Concentration) in Edogawa River (Figure-32), the radio-cesium concentration in suspended solid was around 22,000 Bq/kg in November, 2012 and in September, 2013 it became about 2,000 Bq/kg. The radio-cesium concentration is decreasing gradually with time. The supplies of contaminated sediment were decreasing with time from the wash off, run off from adjacent area and also from the upstream region.

## **5.8 Suspended Solid Flux and Cesium Mass**

Suspended Solid Flux is related with the water discharge and suspended sediment concentration. The total mass of cesium could be an indicator of the movement of sediment from upstream region to downstream region. In Arakawa River the water discharge were almost same ( $80 \text{ m}^3/\text{s}$ ) in 2011 and 2012 and thus the suspended solid flux also same. In Edogawa River there were variation in water discharge, it was  $110.72 \text{ m}^3/\text{s}$  in 2011 and  $78.90 \text{ m}^3/\text{s}$  in 2012 as a result the suspended sediment flux also decreased from 0.9 million ton to 0.65 million ton in 2012. The total mass of cesium in those two rivers indicated that the contaminated sediment was moving towards downstream region largely in Arakawa River from November, 2012 (figure-40). In Edogawa River total mass of cesium was sharply decreased after March, 2013 but still higher than Arakawa River (figure-40). May be it happened due to regulation of water flow through

water gate and structure in Edogawa River. These phenomena also indicated the less movement of sediment towards river mouth in Edogawa River than Arakawa River.

### **5.9 Fate of the Contaminated Sediment**

From the information about spatial and temporal distribution of radio-caesium, vertical profile and caesium concentration in suspended solid in Arakawa, Edogawa, Sumidagawa River and in Tokyo Bay, it can be said that most of the contaminated sediment passed towards the river mouth from Arakawa River as it is an independent river, for its topography and water discharge pattern. In case of Edogawa and Sumidagawa River still the contaminated sediment remained in the upper stream region because Edogawa River is not Independent River and connected to Tone River and water discharge was regulated through water gate. Sumidagawa River also a part of Arakawa River and water discharge was also regulated. In case of vertical profile in all three rivers most of the contaminated sediment was deposited or accumulated in between 4-18 cm depth from the sediment bed. Thus the contaminated sediment was buried enough depth to prevent further exposure of the contaminated sediment to Tokyo Bay or bio-available. More over the supply of contaminated sediment from upper most regions was already limited in all three rivers and the radio-caesium activity was also decreasing with time.

## Chapter 6: Conclusion

The cesium concentration in Tokyo bay was not changing rapidly and almost in low concentration. However in case of river beds, it showed variations from 2012 to 2013. The results showed that the sediment transport in Arakawa River was relatively large due to river topography, water discharge pattern. On the other hand in Edogawa River it was relatively smaller and it usually happened only after large rainfall event and typhoon effect. The half of the areas are still covered by forest (48.5%) in the Arakawa River catchment area so a large portion of radio-cesium are trapped in those undisturbed forest areas and also in vegetation covered land areas. After typhoon of September 15, 2013 when huge wash-off added to the river system though the cesium concentration is decreasing in the suspended solid (Figure-36) indicates that most of the contaminated sediment passed to the water system and thus buried in the river bed or transported to the river mouth region in Tokyo Bay. The radio-cesium concentration was decreasing gradually with time and the supply of radio-cesium attached sediment from the wash off, run off from adjacent areas and from the upper most regions also decreasing with time.

According to vertical profiles of three rivers, most of the contaminated sediment accumulated in the depth between 4-18 cm except the station-7 of Arakawa River which is actually situated within the Tokyo Bay 3.59 km. downstream from the river mouth and here the peak was always in 2-4 cm depth, diffusion and sedimentation were less active here. In Arakawa River, sedimentation was dominant in all seasons. In Edogawa River, diffusion was dominant in summer of wet season. In Sumidagawa River, sedimentation and diffusion were almost same level.

It was difficult to conclude which particle size may contain the more radio-cesium. However it was observed that, total cesium concentrations ( $^{134}\text{Cs}+^{137}\text{Cs}$ ) were high where the silt contents were high. The radio-cesium activities were greater in fine sediments than coarser particles by comparing the total cesium concentrations ( $^{134}\text{Cs}+^{137}\text{Cs}$ ) in sediment samples and in suspended solid.

The relationship of water quality parameters as salinity and turbidity with the cesium distribution was clear. The radio-cesium concentration increased with the turbidity but salinity had shown no effect on the concentration activity of the radio-cesium in the sediment bed.

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