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RELATIVE EFFECT OF WATER QUALITY ON ^{137}Cs ACTIVITY IN LARANGAN WATER, TEGAL

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ABSTRACT

Tegal is a city in Central Java experiencing rapid industrial development. The Larangan waters of Tegal receive household, agricultural, and industrial waste that flows directly into the Bongkok River. The purpose of this study was to determine the relative effects of water quality on ^{137}Cs activity. We found that this wastewater has caused a decline in dissolved oxygen (DO), pH and salinity and an increase in water temperature. Current velocity and water depth increased with distance from the mouth of the river and affected the correlation regression between the water quality parameters. The correlation regression becomes stronger in waters that were affected by the land waste and was very weak at stations 4 and 5, which were not strongly influenced by the waste from the land. The correlation regression between ^{137}Cs activity in the coastal waters under the influence of land showed a strong correlation with several water quality parameters such as DO, pH, temperature, as well as current velocity and water depth.

Keywords: ^{137}Cs, Larangan waters, relative effect, waste, water quality,

INTRODUCTION

An accident such as the one from Nuclear Power Plant (NPP) Fukushima on March 11, 2011, has pushed nuclear experts in the world to improve nuclear research activities in the sea. Some of the radioactive elements of concern are ^{137}Cs, ^{90}Sr, and ^{241}Am. ^{137}Cs is the most abundant anthropogenic radionuclide in the marine environment and has a half-life of 30.17 years. Its conservative nature means that the element is soluble, and is therefore easily distributed, but also can be deposited with the influence of other factors including particle size and mineral content of chemicals such as organic material (Muslim et al., 2015).

Coastal and estuarine waters are the most productive aquatic systems (Pereira - Filho et al., 2001) suitable for spawning, growth, breeding and protective areas for a wide variety of fish, mollusks, crustaceans, birds and mammals (Ohrel and Register, 2006). Estuaries also support fisheries, transportation, and tourism, and are natural buffers between land and sea. In Indonesia, today, increasing pollution from industrial and agricultural waste are discharged into estuarine waters (Sri et al., 2014). The marine environment is also a major recipient of anthropogenic global radionuclide fallout from testing of nuclear weapons by the US and the USSR in the 1960s, nuclear accidents and waste disposal from Nuclear Power Plants (Povinec et al., 2003a; UNSCEAR, 2000).

Beach areas are also important to human activity and this region becomes very sensitive to anthropogenic discharges (Jones et al., 2002; Muslim and Jones, 2003; Prasanna and Ranjan, 2010).

It is already evident that urbanization and industrialization has a great and direct influence on the rate of change of sedimentation in coastal areas (Lu and Matsumoto, 2005).

Chemical constituents in coastal or estuarine waters can be controlled by physical influences within or outside the water (Jones et al., 2002). Organic materials in the ocean that can affect the level of adsorption of chemicals (eg. ^{137}Cs), are influenced by several factors such as currents, depth of water and particle size (Arzayus et al., 2002; Kristensen and Blackburn, 1987; Muslim et al., 2015; Sun et al., 2002). Dissolution easily occurs in shallow estuarine and coastal areas due to physical and biological processes (Dellapenna et al., 1998; Hopkinson, 1985). Differences in environmental conditions cause regional differences in the biogeochemistry of the decomposition of organic matter.

Estuarine environments are generally located at a river mouth that empties into the sea (Elliott and McLusky, 2002). Therefore, estuarine mixing incorporates both saline and fresh water, assisted by wind movement. The flow of sea water is influenced by tidal currents, while the fresh water comes from the river (Priya et al., 2012). Changes in water level and mixing in estuarine environments during low tide as well as in the monsoon season affects water quality measurements including total suspended solids (Chen et al., 2006; Spellman, 2011), dissolved oxygen (Perkins, 1974), water temperature (Olausson and Cato, 1980), salinity (Ohrel and Register, 2006; Prasanna and Ranjan, 2010), pH (Spellman, 2011), conductivity (Smith, 1992), light intensity (Dennison et al., 1993), brightness (Borja and Collins, 2004; Wangersky, 2006), surface and bottom currents (Kramer et al., 1994), nitrogen (Kennish, 2002; Neil, 2005), phosphate (Van der Zee et al., 2007) and the concentration of chlorophyll a (Conley et al., 2000; Muslim and Jones, 2003; Zheng et al., 2004) that will affect the biomass and species composition of phytoplankton (Aquino et al., 2015; Canini et al., 2013; Domingues et al., 2010;.Lauria et al., 1999)

Larangan waters in the Tegal regency are located along the northern coast of Central Java Province. These waters receive various kinds of waste from the land that flowed through the Bongkok River. Coastal land is used as farms that receive fresh water from the Bongkok River. Rice fields, villages and fish market line both sides of the Bongkok River. The Tegal city is located in a very geographically strategic position, connecting the national and regional economies, namely the northern coast of Java, from west to east (Jakarta-Tegal-Semarang-Surabaya) with the central and southern regions of Java (Jakarta-Tegal-Purwokerto-Yogyakarta-Surabaya). The town has become very busy and has showed the most rapid growth in industry compared with other cities in Central Java. The average high temperature is 35 degrees Celsius and rainfall is very low. These conditions also greatly affect local water quality and impact the activity of ^{137}Cs that has been detected in some of the Indonesian waters (Suseno and Prihatiningsih, 2014). Many previous studies have observed activity concentration of ^{137}Cs in some marine area of Indonesia (Akhyar et al., 2013; Prihatiningsih and Suseno, 2007; Suseno, 2014; Suseno and Prihatiningsih, 2012; Wahyono, 2013). Research on the correlation between ^{137}Cs activities with clay mineral content in the marine sediment of Muria Peninsula has also been conducted (Suseno, 2013). To enhance the study of the influence of other environmental parameters, we conducted this research with the main purpose was to determine the relative effect of water quality on the activity of ^{137}Cs in the Larangan Water, Tegal.

MATERIALS AND METHODS

Determination of Research Station

Seven stations were chosen with attention to the circumstances of each station including depth and distance from the Bongkok River estuary. The coordinates of each station were determined by GPS (Global Positioning System). Stations 1 and 2 are located in the breakwater area that gets the direct influence of the river and has a water depth < 2.5 m, stations 3 and 7 are in the region close to the Larangan beach with a depth < 3 m. Stations 4, 5 and 6 located far away from the coast or into the open sea with a depth > 3 m. Coordinates of the location of each station are shown in Table 1 and Figure 1.

Table 1: Station site information

Station	**Coordinate**		**Depth (meter)**
	Longitude	**Latitude**	
1	06º51’35.9”	109º11’38.5”	1.44
2	06º51’27.7”	109º11’39.1”	2.37
3	06º51’22.5”	109º11’9.2”	2.63
4	06º51’0.7”	109º11’22.0”	3.41
5	06º51’0.1”	109º11’44.8”	4.13
6	06º51’8.5”	109º12’6.3”	3.16
7	06º51’38.4”	109º12’7.1”	1.68

Sampling Methods

Surface sea water for ^{137}Cs analysis was collected using 30 L polyethylene buckets that were rinsed at least twice with surface seawater and then were poured into a 100 L plastic bucket until the 90 L mark. We immediately added $K_4[Fe(CN)_6]$ and $CuSO_4$ (10 grams each) to the water samples, which binds ^{137}Cs in a water sample. In order for the binding to be completed, the water sample was stirred then allowed to stand until a precipitate containing ^{137}Cs formed. Clearwater above the sediment was removed by vacuuming with a plastic hose, and the precipitates were collected in a 5 L bucket and taken to the laboratory. While sampling, we were also measured water quality i.e. salinity, temperature, dissolved oxygen (DO), pH and water depth.

Current Modeling

Modeling was done using the current pattern Mike21 program. Bathymetry maps were used as an input model of the current pattern which is based on the depth of research sites, found using ArcGIS program to obtain the value of XYZ, where X and Y as coordinates point and Z is depth. We were then input wind data using the data ECMWF (European Center for Medium-range Weather Forecasting) one day prior to the study, the study day, and one day after the study. The result of Mike21 module program processing was a current model that showed the velocity and direction of currents.

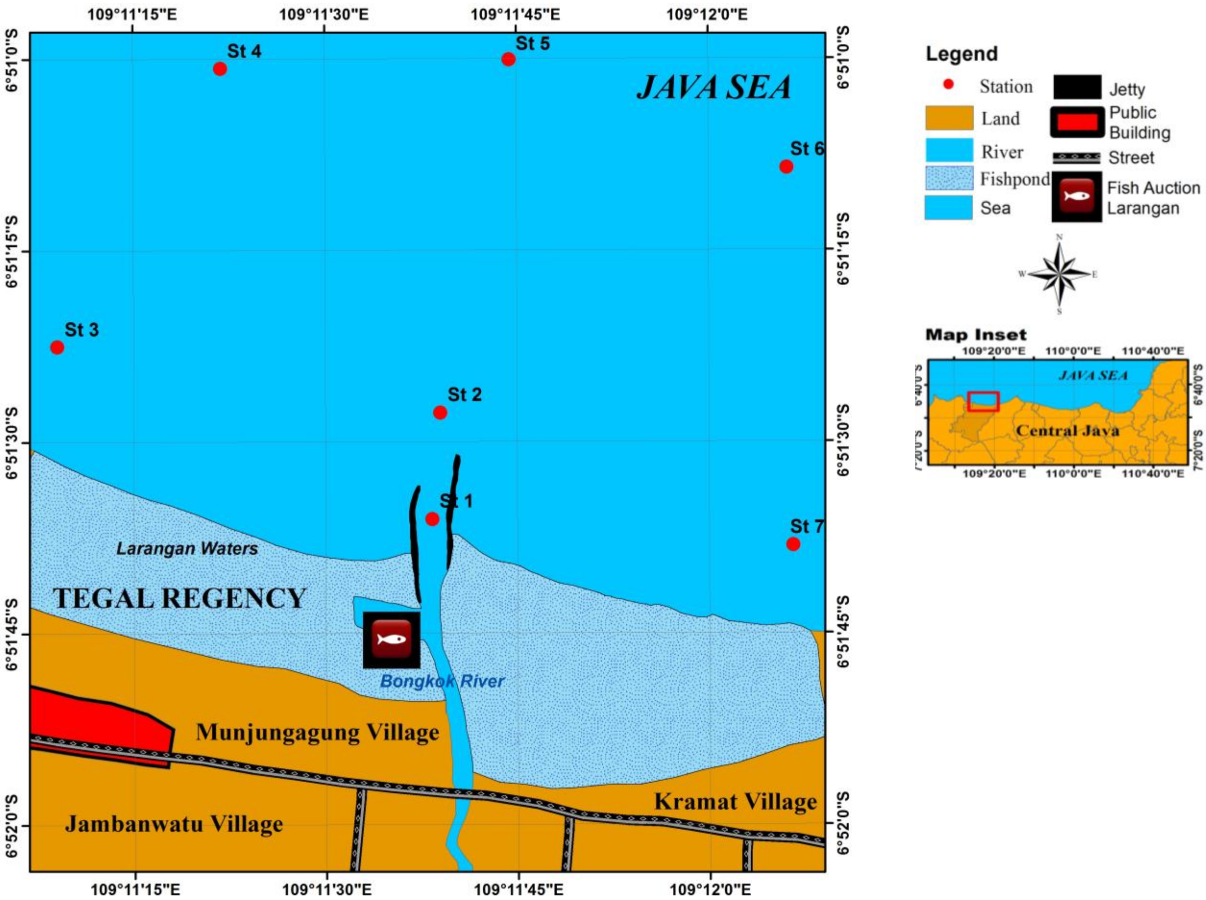

Figure 1: Location of study station

Preparation and Measurement 137Cs in the Laboratory

At the laboratory, samples in the 5 L buckets were filtered with filter paper to separate the precipitates from the water. The precipitates were then oven-dried at 70 - 80oC for about 3 days. The dried precipitates were then put in a plastic container for the measurement (counting) using Gamma Spectrometer for 3 days (IAEA, 2005).

RESULTS AND DISCUSSION

Water Quality Parameters

Water quality parameters in the Larangan waters including dissolved oxygen (DO), pH, temperature, salinity and current velocity model results are shown in Table 2. Although station 1 is still included in the basin area, the value of DO, pH, current velocity and salinity was the lowest compared with other stations. This due to 90% of organic waste from the mainland was deposited in estuaries (Tao et al., 2016) and decomposes in the water, causing the pH and DO to decrease as the organic material decomposes (Rasiq et al., 2016). The low salinity at station 1 was due to the freshwater inputs from the mainland. The low of current velocitys at station 1 due to the area is a semi-enclosed system so the current from the outside was weakened by the breakwater in front of the river mouth. The temperature at station 1 was the highest, even when sampling in the morning due to the influence of heat transfer from the mainland.

Table 2: Water quality in Larangan waters, Tegal

Station	**Environmental Parameter**				
	DO (mg/l)	**pH**	**Temperature (°C)**	**Salinity (‰)**	**Current (m/s)**
1	4.63	7.53	31.2	27	0.0487
2	5.16	7.62	30.5	29	0.0487
3	5.37	7.74	30.3	28	0.0299
4	4.78	7.82	29.6	30	0.0456
5	5.05	7.86	29.4	31	0.0609
6	5.64	7.78	30.2	30	0.0617
7	4.75	7.61	30.8	29	0.0211

Figure 1 shows that the station 4 and 5 are closest to the open sea and had the deepest water depths of 3.41 m and 4.13 m, respectively. These conditions were affected the water quality i.e. pH which was the highest at these sites (7.82 and 7.86). Stations 4 and 5 were far from the river mouth and have low organic matter content, therefore, the concentration of carbon and nutrients are reduced (Ferrari, 2000). Consequently, the pH rises, indicating the water is more alkaline as well as less influenced by the organic material from the river, which influences pH, creating an alkaline pH (Schulz et al., 2006).

The salinity at stations 4 and 5 was relatively high despite the low temperatures, because the fresh water from the river does not affect stations 4 and 5, and the level of evaporation was greater than near the coastline (Prasanna and Ranjan, 2010).

The above conditions affect the correlation regression between parameters of water quality, where the correlation between the parameters was weak for stations 4 and 5, or the correlation regression between the parameters of the water quality becomes more significant if tested on waters still heavily influenced from the mainland (terrestrial runoff) such as in stations 1, 2, 3, 6 and 7. The correlation regression between water quality parameters in all stations and excluding stations 4 and 5 can be seen in Figures 2, 3, 4, 5, 6, 7 and 8.

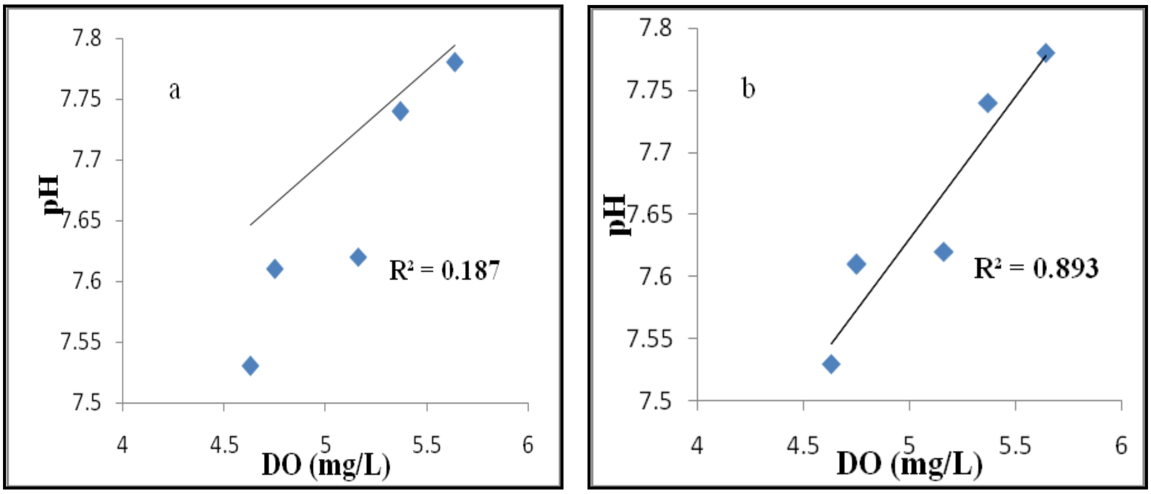

Figure 2: Correlation regression for (a) DO and pH at all stations; (b) DO and pH at station 1,2,3,6 and 7

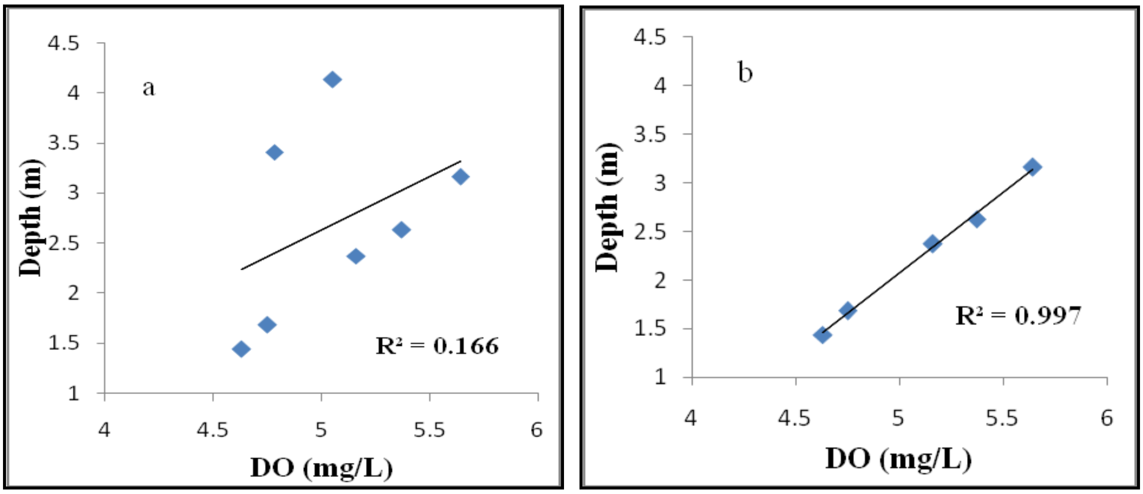

Figure 3: Correlation regression for (a) DO and depth at all stations; (b) DO and depth at station 1,2,3,6 and 7

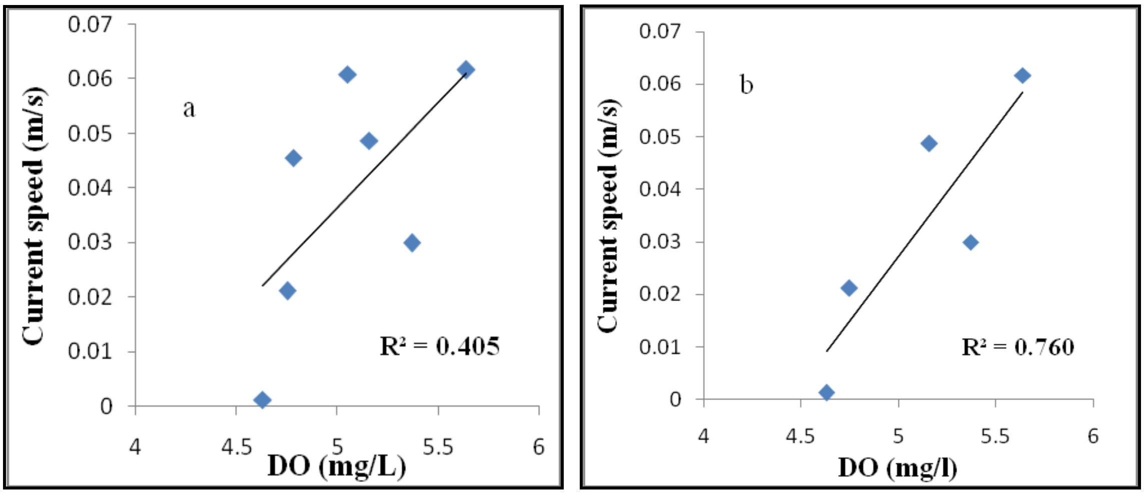

Figure 4: Correlation regression for (a) DO and current velocity at all stations; (b) DO and current velocity at station 1,2,3,6 and 7

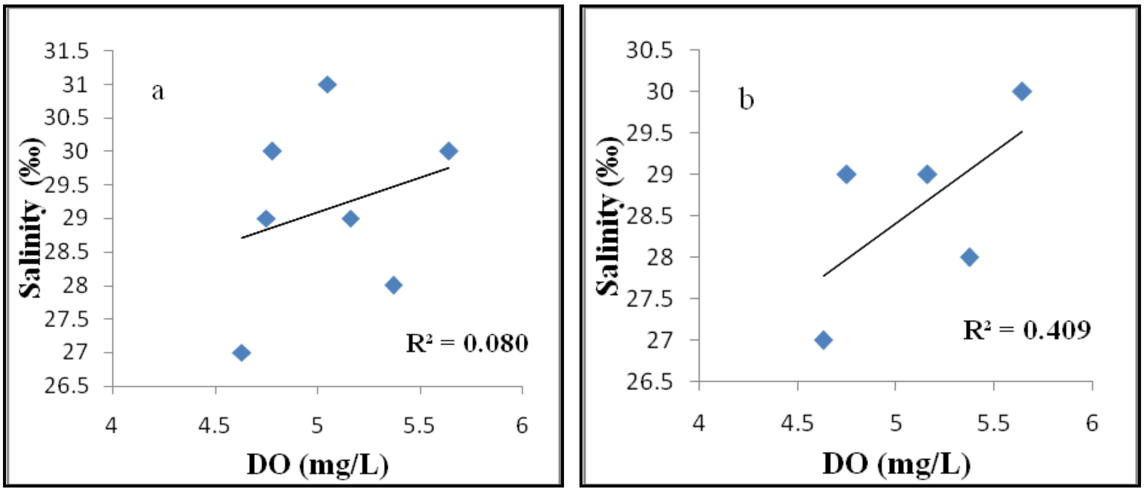

Figure 5: Correlation regression for (a) DO and salinity at all stations; (b) DO and salinity at station 1,2,3,6 and 7

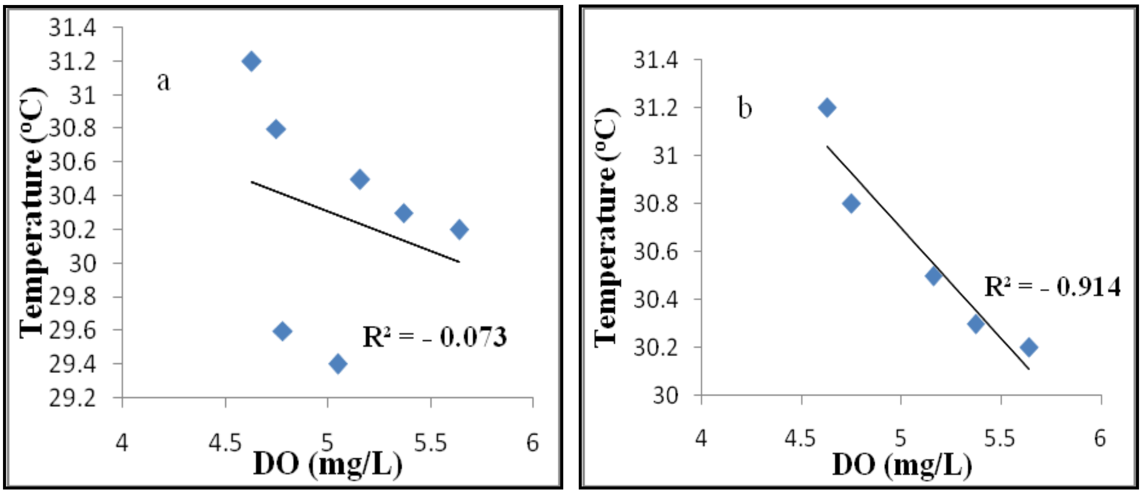

Figure 6: Correlation regression for (a) DO and temperature at all stations; (b) DO and temperature at station 1,2,3,6 and 7

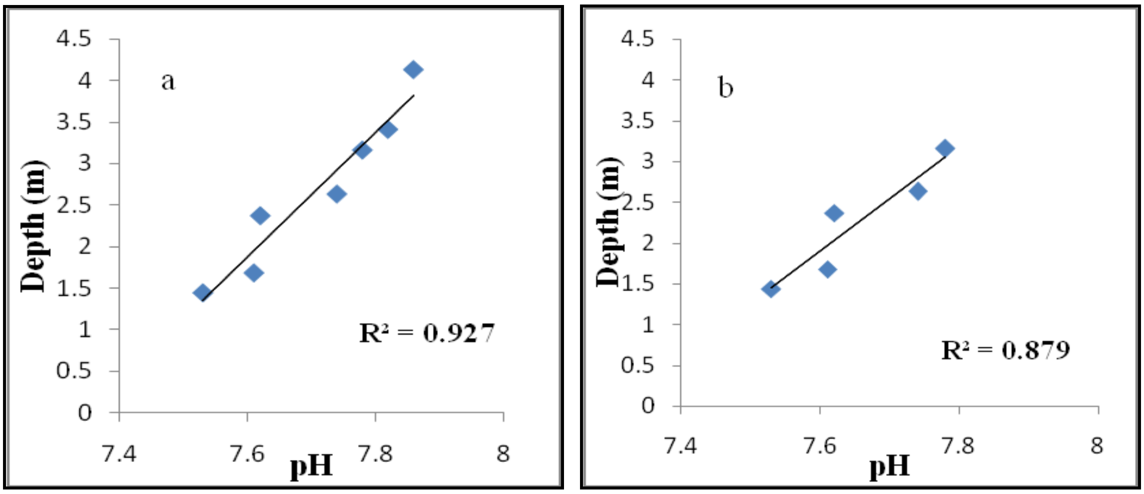

Figure 7: Correlation regression for (a) pH and depth at all stations; (b) pH and depth at station 1,2,3,6 and 7

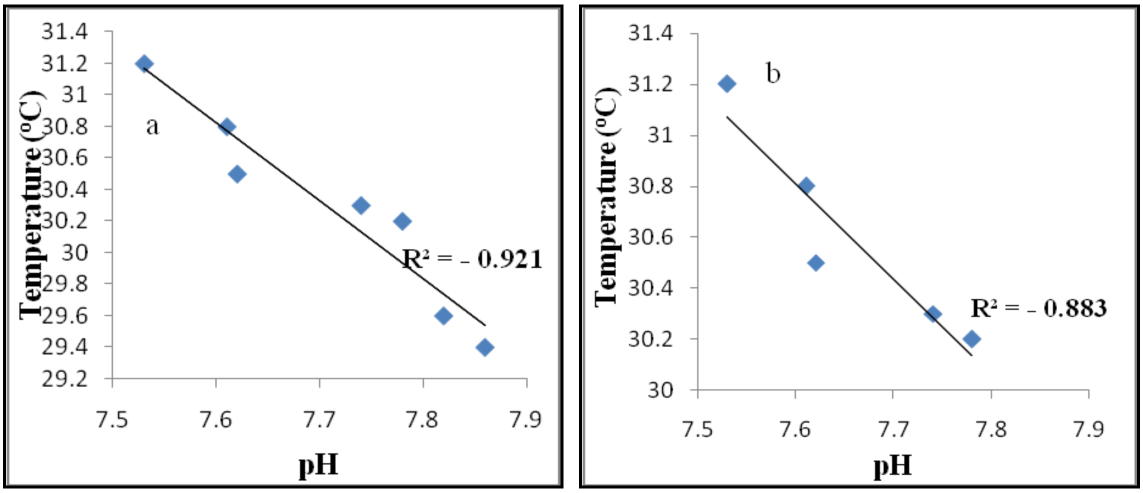

Figure 8: Correlation regression for (a) pH and temperature at all stations; (b) pH and temperature at station 1,2,3,6 and 7

Oxidation of organic materials that consume oxygen in addition to reducing DO also reduces the pH (Marion et al., 2011). In this study, the correlation regression between DO and pH occurs well in areas that are still affected by the wastewater (Figure 2b; $R^2 = 0.893$). However, the correlation regression will be weaker when we include stations 4 and 5, which are not very influenced by the wastewater runoff (Figure 2a; $R^2= 0.187$). This shows that the mainland waste (organic waste) affects DO and pH. According to Muslim (2010; 2013) the increase in organic matter in the water is proportional to the increase in the value of COD, (Chemical Oxygen Demand) therefore reducing the dissolved oxygen (DO) content and pH.

Figure 3b shows that the DO also has a strong positive correlation with depth ($R^2= 0.997$) at stations that are influenced by the mainland. According to Chen et al., (2016) the oxidation process of organic material occurs at the bottom of the water so that surface water in deeper areas (stations 4 and 5) has a higher DO content, due to the low oxidation of organic materials and photosynthesis producing an oxygen-rich layer higher in the water column. In addition to increasing DO, pH also increases with increasing depths of water. According to Ahmat et al. (2016), the correlation regression depends on environmental conditions, especially the slope of land sediment. In this study, increasing the water depth and distance from the source of waste (organic) shows that DO is also positively associated with the current velocity (Figure 4).

Figure 6 also shows that the DO has a strong negative correlation with temperature, because temperature decreases with increasing distance from the mainland, while DO increases with distance from the mainland (Tao et al., 2016). Therefore, there is a relationship between the water quality parameters above.

The concentration of salinity in waters is heavily influenced by several factors, such as the flow of fresh water from rivers, evaporation, topography and vegetation distribution (Humphries et al., 2010). The correlation regression between salinity and DO is weakly positive ($R^2 = 0.08$ and $R^2 = 0.409$) (Figure 5). Pavlov et al. (2016) found a strong correlation regression between salinity and oxygen ($R^2 = 0.96$, n = 162) in an area with freshwater inputs from the mainland. Kim et al. (2016) and Chen et al. (2016) found a negative relationship between salinity with some organic materials from the land, spreading from Changjiang River to the East China Sea. If oxidation of organic matter occurs, the DO concentration will decrease; therefore, DO shows a positive correlation with salinity.

The relative effect also occurred in regards to pH, where pH has a strong positive correlation with depth (Figure 7). The depth increases and temperature decrease with distance from the estuary. Therefore, the pH also has a relatively strong association with temperature (Figure 8).

Current Patterns

The results of modeling the direction and current velocity at high and low tide are shown in Figure 9 and 10. At high tide, the current moves from southeast to northwest, with uneven flow velocity; flow toward the offshore area is stronger than in areas near the coast (estuarine), and the most powerful location is in the eastern station, station 6 (Figure 9). At low tide, the current flows in the opposite direction, from the northwest to the southeast, with the current pace increasing in the east (Figure 10).

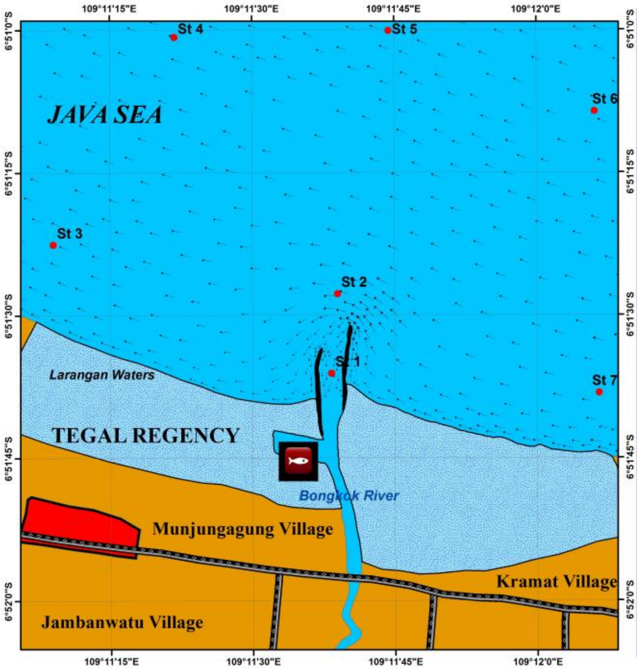

Figure 9**:** Current pattern at high tide on October 2015

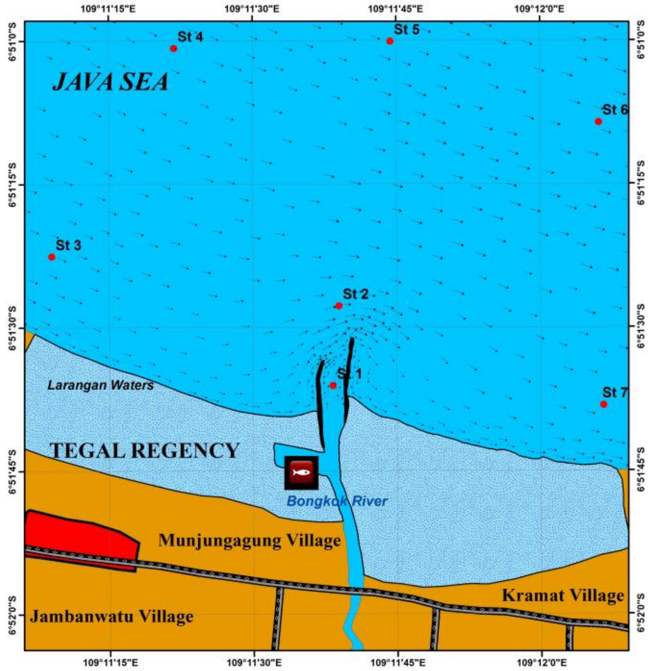

Figure 10: Current pattern at low tide on October 2015

^{137}Cs Activity in the Larangan Waters Tegal

^{137}Cs activity in the Larangan waters, Tegal was detected with values ranging between 0.03 to 1.22 mBq/L, and an average value of 0.58 mBq/L (Table 3). This value is lower than previous research in Sayung waters, Demak in October 2014, with an average value of 0.86 mBq/L (Muslim et al., In press) and higher than in Gresik waters tested in September 2013 with a value of 0.200 mBq/L (Muslim et al., 2015). The difference is due to different water conditions that ^{137}Cs in water are heavily influenced by factors such as current pattern and ion exchange with other elements such as organic matter and sulfur (Muslim et al., 2015).

Table 3: Analysis result of ^{137}Cs in sea water

Station	^{137}Cs (mBq/L)	Depth (m)	Current (m/s)
1	0.03	1.44	0.0012
2	0.95	2.37	0.0487
3	0.77	2.63	0.0299
4	0.73	3.41	0.0456
5	0.64	4.13	0.0609
6	1.22	3.16	0.0617
7	0.29	1.68	0.0211
Mean	0.58		

Table 3 shows that the highest ^{137}Cs activity (1.22 mBq/L) and current velocity (0.0617 m/sec) at station 6. While the lowest occurred in station 1 for ^{137}Cs (0.03 mBq/L) and for current velocity (0.0012 m/sec). Correlation between current velocity and ^{137}Cs activity at all stations is positively correlated R^2= 0.752 (Figure 11) and even stronger when the correlation was tested at station 1, 2, 3, 6 and 7 (R^2= 0.880). This shows that the ^{137}Cs activity levels are affected by the current velocity, because according to Povinec et al. (2003b) the radionuclides in water can be affected by advection, dispersion, and precipitation.

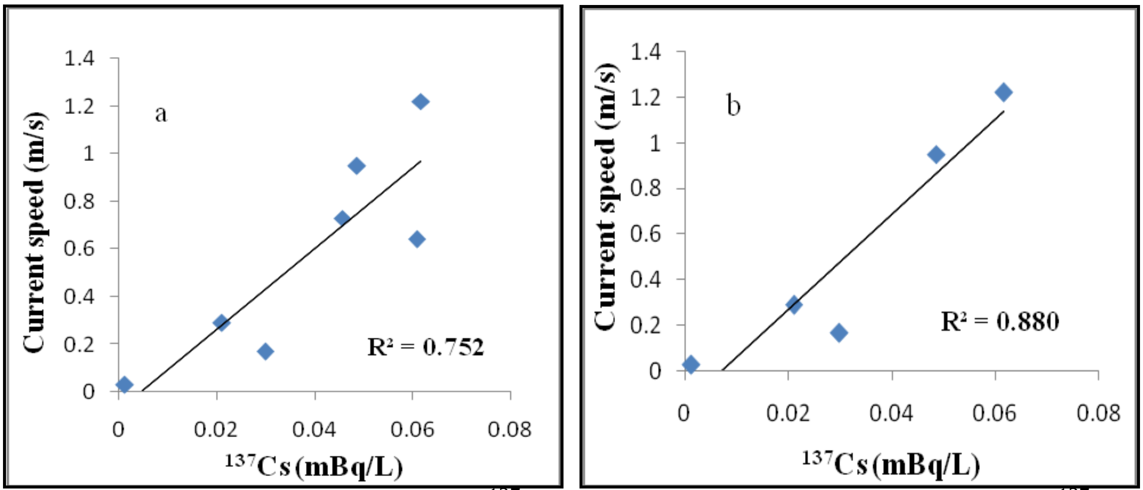

Figure 11: Correlation regression for (a) ^{137}Cs and current velocity at all stations; (b) ^{137}Cs and current velocity at station 1,2,3,6 and 7

Based on the water quality conditions above, the relative correlation occurs between the activity of ^{137}Cs with water quality parameters as shown in Figures 12, 13, 14, 15 and 16.

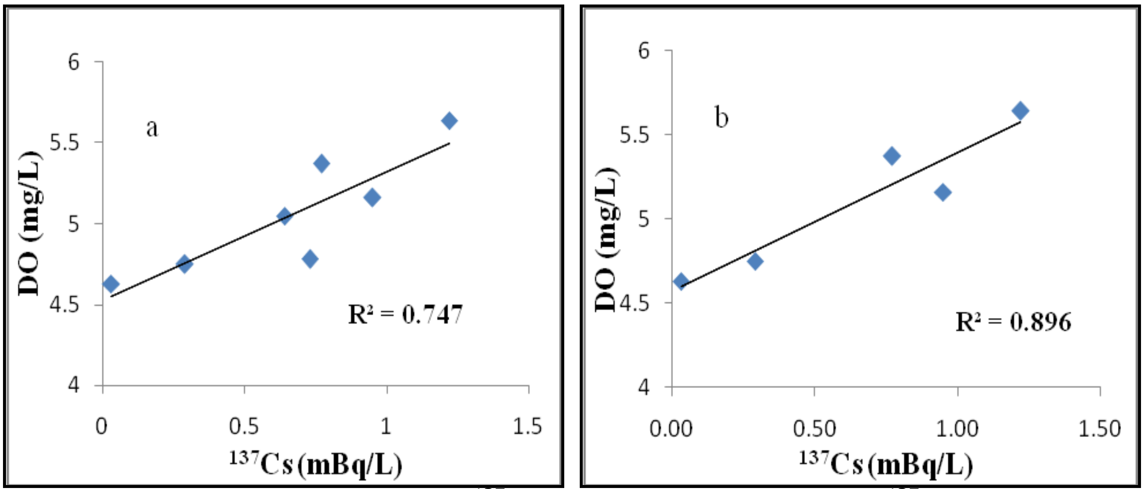

Figure12: Correlation regression for (a) ^{137}Cs and DO at all stations; (b) ^{137}Cs and DO at station 1,2,3,6 and 7

Figure 12 shows that the ^{137}Cs activity has a positive correlation with DO (R^2= 0.747 and R^2= 0.896). This correlation occurs indirectly, because ^{137}Cs have direct influence with organic material, and when organic matter increases, ^{137}Cs in the water decreases because it binds with the organic matter and settles to the bottom (Van Bergeijk et al., 1992). Therefore, more organic material reduces the degradation time for ^{137}Cs and DO in the water.

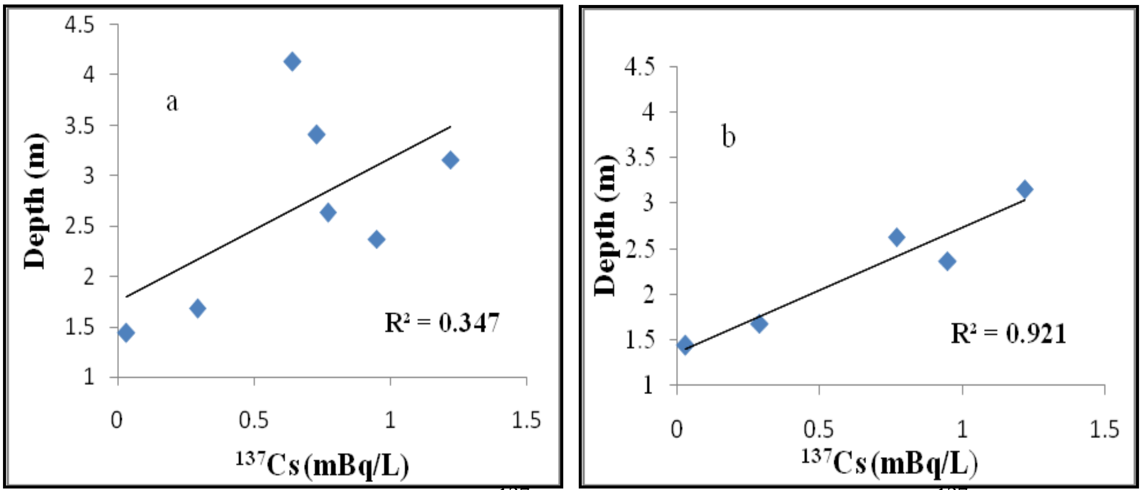

Figure13: Correlation regression for (a) ^{137}Cs and depth at all stations; (b) ^{137}Cs and depth at station 1,2,3,6 and 7

Organic materials sourced from the mainland decreased with distance from the source. This research shows that the further away from the mainland, the water also becomes deeper. Under these conditions, the DO and ^{137}Cs activity have a strong correlation with water depth (Figure 13), with R^2= 0.921 at the stations under the influence of the organic material.

pH has a strong positive correlation (R^2= 0.893) with DO (Figure 2) at stations that are affected by the wastewater (organic). DO and pH also affects the correlation between ^{137}Cs activity and pH (Figure 14). Rahman and Voigt (2004) found that increasing ^{137}Cs with an increase in pH and K content of sediment (soil potassium).

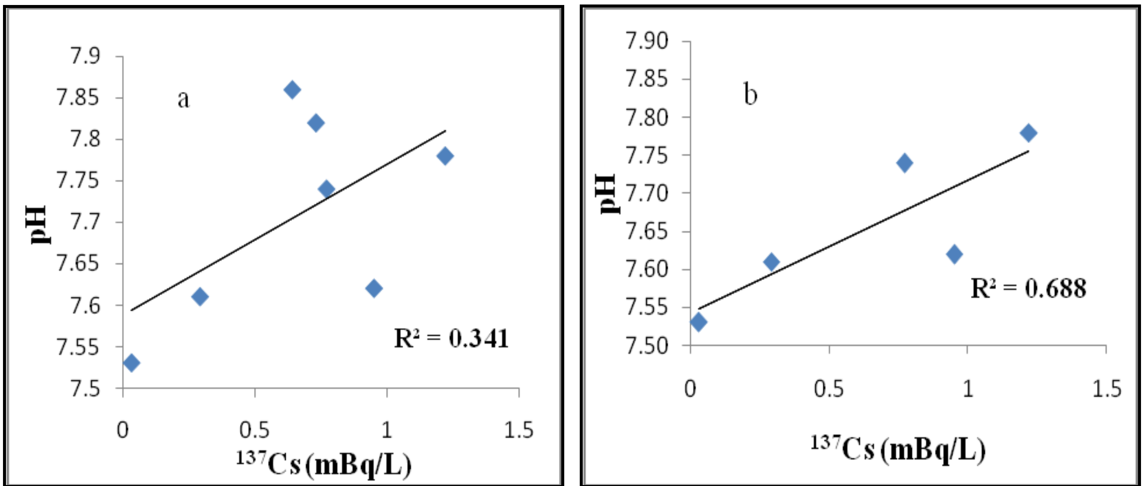

Figure 14: Correlation regression for ^{137}Cs and pH at all stations; (b) ^{137}Cs and pH at station 1,2,3,6 and 7

The variation of salinity is due to many factors, so the correlation regression between DO and salinity was not very strong with an R^2 = 0.080 and R^2 = 0.409 (Figure 5). This resulted in the correlation regression between ^{137}Cs activity with salinity becoming stronger as shown in Figure 15.

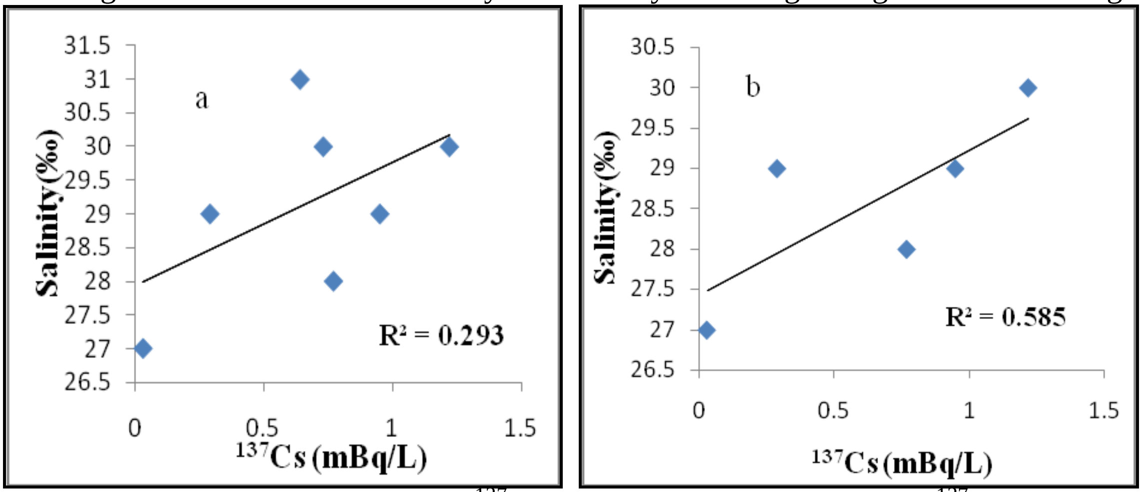

Figure 15: Correlation regression for (a) ^{137}Cs and salinity at all stations; (b) ^{137}Cs and salinity at station 1,2,3,6 and 7

The distribution of temperature in the Larangan waters Tegal decreases with distance from the mainland. The DO concentration, as opposed to temperature, increased with distance from the mainland. The conditions also affect the correlation between ^{137}Cs activity and temperature (Figure 16), which has a negative correlation (R^2 = - 0.874) to stations that are still affected by the waste organic materials from the mainland.

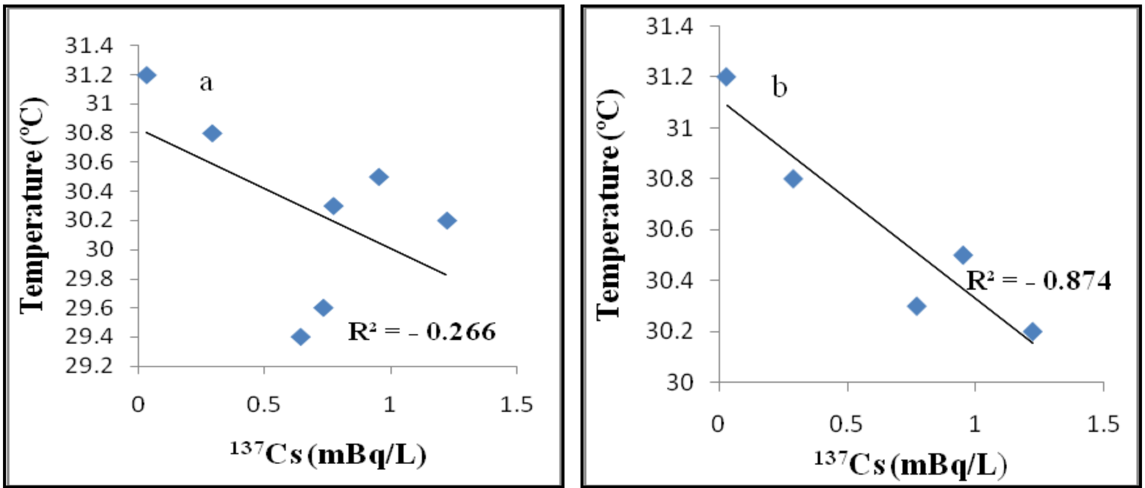

Figure 16: Correlation regression for (a) ^{137}Cs and temperature at all stations; (b) ^{37}Cs and temperature at station 1,2,3,6 and 7

CONCLUSIONS

The values of the water quality parameters in the Larangan waters, Tegal i.e. pH, DO, salinity, and temperature are influenced by the runoff from the Bongkok River which contains both organic material and warm water. Station 1 is located in the estuary of the river, the shallowest water depth, weakest current velocity and the highest water temperature. Station 1 also has the lowest pH, DO, and salinity. The low DO at station 1 is due to the decomposition of organic material, there is the highest organic material at this station compared to other stations. The result shows the strong correlated regression between the water quality parameters at other stations (1,2,3,6 and 7) without station 4 and 5that both stations are not very affected by waste from the mainland. The water quality also influenced the correlation regression with ^{137}Cs activity. The lowest ^{137}Cs activity recorded at station 1 because high content of organic material in waste at station 1. The high ^{137}Cs activity found at stations 4 and 5 due to small input of mainland waste. The correlation regression between water quality and ^{137}Cs activity was also strong correlated when this regression analysis was test at 5 stations without station 4 and 5.

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