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DISTRIBUTION OF NATURAL AND ANTHROPOGENIC RADIONUCLIDES IN SURFICIAL SEDIMENTS OF JAKARTA BAY

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ABSTRACT

DISTRIBUTION OF NATURAL AND ANTHROPOGENIC RADIONUCLIDES IN SURFICIAL SEDIMENTS OF JAKARTA BAY. The concentration and distribution of natural and anthropogenic radionuclide in surficial sediments of Jakarta Bay were investigated with the aim of evaluating its level and environmental radioactivity. Sediments were sampled in 30 locations using Smith-McIntyre Grab sampler. Sediments were dried, homogenized and sealed for 1 month for equilibration and for the detection, analysis and data acquisition, a high purity germanium (HPGe) detector coupled with a high resolution multichannel analyzer (MCA) was used. Additionally, the grain sizes were analyzed by means of hydrometer. The result shows that the specific activity of ^{238}U, ^{226}Ra, ^{232}Th, ^{40}K and ^{137}Cs range from 6.71 ± 1.01 Bq/kg to 28.63 ± 4.29 Bq/kg, 6.46 ± 0.97 Bq/kg to 28.21 ± 4.23 Bq/kg, 16.62 ± 2.49 Bq/kg to 40.46 ± 6.07 Bq/kg, 115.80 ± 12.16 Bq/kg to 358.69 ± 30.49 Bq/kg, and 0.03 ± 0.01 Bq/kg to 1.99 ± 0.34 Bq/kg with the average value are 12.83 ± 2.11 Bq/kg, 12.03 ± 1.98 Bq/kg, 26.55 ± 4.36 Bq/kg, 235.55 ± 19.37 Bq/kg, and 0.77 ± 0.13 Bq/kg, respectively. It shows that the activity of radionuclides in the research area are in natural level and the variation may be influenced by the grain size distribution in the sample. The absorbed dose rate and radium equivalent of gamma radiation was estimated to be 32.06 ± 7.72 nanoGrey/h and 68.14 ± 11.20 Bq/kg, respectively.

INTRODUCTION

Jakarta Bay is situated in the north of Jakarta Metropolitan. It is a shallow bay (average 15m) that covers an area of about 514 km^2, with coordinate $106^0 40'$E to 107^0 E and $5^0 50'$S to $6^0 7'$S and the length about 50 km from West to East. Jakarta Bay functions as the waste-water disposal site for the Jakarta Metropolitan Area. The entire waste-water production of Jakarta transported by 13 rivers and canals which pass through the Jakarta Metropolitan area ends up in the Jakarta Bay. In addition, the rivers bring a considerable amount of solid waste into the bay. Direct discharges into the Bay through the dumping of dredging spoils and sledge also take place. The water and sediment in Jakarta Bay have high concentration of several heavy metals and organic content [1]. Moreover, Jakarta Bay may also have a certain level of natural and artificial radionuclides which may be transported by rivers or direct discharges and

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deposited in the Bay. The possible sources of radionuclides are related to the growing use of nuclear and radiation devices in industry, medicine, science and technology and mining. The latter has more potential sources for natural radionuclide which is called TENORM (Technologically Enhanced Naturally Occurring Radionuclides).

Radiation of natural origin at the earth's surface consists of two components namely cosmic rays and radiation from the radioactive nuclides in the earth's crust. The latter components, the terrestrial radiation, mainly originates from the primordial radioactive nuclides that were originated in the early stage of formation of the solar system. Uranium, thorium and potassium are the main elements contributing to natural terrestrial radioactivity [2]. Uranium has two primary isotopes ^{238}U ($T½ = 9.47x10^{10}$ y) and ^{235}U ($T½ = 7.108$ y) which, at present, occur in the proportion 99.3% ^{238}U and 0.7% ^{235}U. Both exhibit long and complex decay series. Thorium ($T½ = 1.41x10^{10}$y) has only one isotope, while potassium has three isotopes (^{39}K, ^{40}K and ^{41}K) with ^{40}K ($T½ = 1.20x10^{9}$y) being only radioactive isotope of abundance 0.012% of potassium. Almost 90 % of the radiation exposure comes from natural and 10 % from artificial or man-made sources. Most of the radioactive elements produced by the atmospheric nuclear radionuclides decay rapidly, and so, only those with longer half-life remains in the environment.

Radionuclides released into the environment, reach the human body through several transfer processes. Therefore, the measurement of radioactivity level is necessary in the enviroment surrounding human beings. Moreover, knowledge of basic radiological parameters and radionuclide contents is also important since it allows us to calculate the exposure of the population to radiation from natural resources. Information about the presence of uranium, thorium and their decays in the environmental radiation context has been increasing. The knowledge of the distribution of these radionuclides in soil, rocks and sediment plays an important role in radiation protection measurement, geoscientific research and guidelines for the use and management of these materials [3]. Most countries carry out nationwide surveys to assess the level of radioactivity so as to compile data for possible radiological hazards and to take necessary action. IAEA (International Atomic Energy Agency) has also created some projects such as GLOMARD (Global Marine Radioactivity Database) and ASPAMARD (Asia-Pasific Marine Radioactivity Database) with the purpose to map the global marine radioactive database [4,5].

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No data are as yet available on the regional distributions of natural and artificial radionuclides in sediments of the Jakarta Bay Area. In the present work, surface sediment were collected from 30 locations in the Jakarta Bay region, and measured for the concentration of natural radionuclides ^{232}Th, ^{238}U, ^{226}Ra, ^{40}K and artificial ^{137}Cs to assess their content and the distribution pattern in the ecosystem of Jakarta Bay Area. The result of work provide background data on natural and artificial radionuclide and environmental pollution by radionuclide which are useful in the assessment of human radiation exposure from natural environment. The accumulation of information on natural radiation is great value for radiation protection.

MATERIALS AND METHODS

Sample collection and preparation.

Surface sediments were collected from 30 locations on the Jakarta Bay area in May 2004 (west season) using Smith-McIntyre grab sampler (Fig. 1). Sampling was done in collaboration with Research Center for Oceanography, Indonesian Institute of Sciences (P2O-LIPI), at each location, sediment was collected three times for analyzing the heavy metals, organic contents, benthos and natural and anthropogenic radionuclides. Moreover, the physical data, namely current, water depth and temperature were also measured. There was about 2 kg wet weight sediment from each sampling location which were placed in plastic bags and transported to the laboratory for radionuclides and heavy metals analysis.

The sediment samples for radionuclide contents were transferred to beaker glass and dried in an oven at 105^0C for 24 hours to achieve the constant weight. The weight of samples were measured prior and after dried for water content analysis. The dried samples were pulverized for homogenizing. 200 g of samples were packed into cylindrical plastic containers and sealed. The containers were kept for 1 month to reach secular equilibrium between the ^{238}U and ^{232}Th series and their respective progeny before measurement. It was assumed that ^{222}Rn and ^{220}Rn could not escape from the sealed containers[7].

Particle size of sediment consists of sand, silt and clay. These fractions were measured for all samples using a hydrometer.

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Radiometric analysis and calculation of radiation hazards

Gamma measurements were performed with a gamma-spectrometer equipped with 8192-channel analyzer. A coaxial type high purity germanium (HPGe) detector model GC-1020 Canberra coupled with PC based MCA acquisition board (AccuSpec-A, Canberra), cooled by liquid nitrogen was used. Detector efficiency was 10% relative to 3″x3″ NaI(Tl) and resolution was 2.1 keV for ^{60}Co peak (1332 keV). Qualitative and quantitative analysis of gamma spectra was processed with a computer program AccuSpec. The detector was housed inside a massive lead shield. Characteristic X-rays from lead have been reduced by use of 1 mm thick layer of copper. The γ-ray energies of ^{212}Pb (238.63keV) was used to determine the concentration of ^{232}Th. The γ-ray energy of ^{234}Th (93keV) and ^{214}Pb (351.9keV) were used to measure the activity of ^{238}U and ^{226}Ra, respectively. The ^{40}K and ^{137}Cs radionuclides were measured from their respective γ-ray energies 1460 and 661.66 keV, respectively [8,9,10]. All measurement were compared to the IAEA-375 Standard for quantitative analysis and the geometry of counting samples was the same as that of the standard sample and the counting time for all the samples was 86400 seconds. The background due to naturally occurring radionuclides in the environment around the detector was measured during weekend in the period of measurement and its value was subtracted from that of each sample. The calculated standard deviation is $\pm$ 1σ.

The reliablity of the method was checked through analyzing two certified samples supplied by IAEA (IAEA-300 and IAEA-368 sediment) and the obtained results were in good agreement with the recommended or information values.

Since the concentration and distribution of radionuclides is not uniform throughout the world so uniformity in respect of exposure to radiation has been defined in terms of radium equivalent activity and absorbed dose rate. In order to compare the specific activities of materials containing different concentration of radium, thorium and potassium, the radium equivalent activity concentration index, Ra_{eq}, was calculated using the formula [7]:

$$Ra_{eq} = C_{Ra} + 1.43C_{Th} + 0.077C_K$$

where C_{Ra}, C_{Th} and C_k are the specific activities of ^{226}Ra, ^{232}Th and ^{40}K (in Bq/kg) respectively. This equation is based + on the estimate that 1 Bq/kg of ^{226}Ra, 0,7 Bq/kg of ^{232}Th or 13 Bq/kg of ^{40}K generate the same γ-ray dose rate.

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The absorbed dose rate in outdoor air one meter above the ground surface due to the radionuclides ^{238}U, ^{232}Th and ^{40}K in soil was estimated using the formula [7]:

$$D = (0.427C_u + 0.662C_{Th} + 0.0432C_K)$$

where the unit of dose rate is nanoGrey/h and C_U, C_{Th} and C_K are the average activity concentrations of ^{238}U, ^{232}Th and ^{40}K, in soil or sediment samples.

RESULTS AND DISCUSSION

Table 1 represent the concentrations with the statistical analysis of the radionuclides in sediment surface of Jakarta Bay, the values being given on dry weight basis. The specific activity of uranium series were; ^{238}U ranged from 6.71 ± 1.01 Bq/kg to 28.63 ± 4.29 Bq/kg with mean value of 12.83 ± 2.11 Bq/kg and ^{226}Ra ranged from 6.46 ± 0.97 Bq/kg to 28.21 ± 4.23 Bq/kg with mean value 12.03 ± 1.98 Bq/kg. The concentration of ^{232}Th ranged from 16.62 ± 2.49 Bq/kg to 40.46 ± 6.07 Bq/kg with the mean value 26.55 ± 4.36 Bq/kg. Potassium has concentration ranged from 115.80 ± 12.16 Bq/kg to 358.69 ± 30.49 Bq/kg with the average 235.55 ± 19.37 Bq/kg and anthropogenic ^{137}Cs was from 0.03 ± 0.01 Bq/kg to 1.99 ± 0.34 Bq/kg and the mean value was 0.77 ± 0.13 Bq/kg.

Fig 1. The map of sampling area of surface sediment in Jakarta Bay.

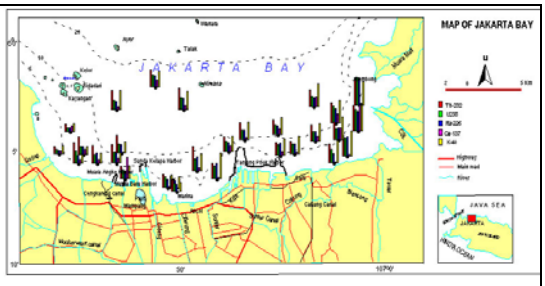

Fig 2. Surface distribution of ^{238}U, ^{232}Th, ^{40}K and ^{137}Cs in the bottom sediment.

Fig. 2 shows the natural and anthropogenic radionuclides distributed in Jakarta Bay which have higher concentration in the eastern part the bay. The radionuclides are preferentially adsorp on clays or organic sedimentary fractions [3]. The analysis of particle fractions (sand, silt and clay) on the sediment shows the clay fraction was higher on the eastern than the western part of the bay. The distribution of each

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radionuclide, radium equivalent and dose rate in surface sediment of study area which is tabulated in normal curve is shown in Fig. 3. The distribution of all radionuclides follow normal distribution based on the goodness of fit χ^2-test with the 95% confidence level. The skewness of the graph is the asymmetry of the shape, a value more than or less than zero indicates skewness in the data. The skewness of all radionuclides in sediment and dose rate are positive which is indicated of the graphs have a right tail. Moreover, ^{238}U and ^{226}Ra have long right tail due to the high value of skewness compared to ^{232}Th, ^{40}K and ^{137}Cs. The high value of skewness of ^{238}U and ^{226}Ra is caused due to the high concentration of both radionuclides in the location 27 compare to the average concentration of both radionuclides in the sampling area (Fig.2).

Table 1. Statistical analysis of natural and anthropogenic radionuclides in surface sediment of Jakarta Bay.

Nuclide	Mean	Range	Skewness
^{238}U (Bq/kg)	12.83±4.39	6.71±1.01- 28.63±4.29	2.04
^{226}Ra (Bq/kg)	12.04±4.47	6.46±0.97 - 28.21±4.23	2.02
^{232}Th (Bq/kg)	26.55±5.88	16.62±2.49 - 40.46±6.07	0.49
^{40}K (Bq/kg)	235.55±71.03	115.80±12.16 - 358.69±30.49	0.002
^{137}Cs (Bq/kg)	0.77±0.53	0.03±0.01 - 1.99±0.34	0.92
Radium eq (Bq/kg)	68.14±11.20	42.04±6.31 - 110.15±16.52	0.76
Dose rate (nGrey/h)	32.06±7.72	20.47±3.07 - 52.52±7.88	0.67

The ratio between natural radionuclides are tabulated in Table 2 and Fig. 4. In general, all radionuclides represent the positive trend between its activity which is shown by their correlation factor. The thorium to uranium concentration (^{232}Th/^{238}U) ratio has a mean value 2.02±0.07 which is more than unity. The activity of ^{238}U in the surface sediment are lower than the ^{232}Th activity due to uranium high geochemical mobility [3]. Thus, a preferential accummulation of uranium by sediments might explain the relatively lower ^{238}U values than ^{232}Th values. The world average of ^{232}Th and ^{238}U activity is 25 Bq/kg and the ^{232}Th/^{238}U quotient is 1.0 [2]. Moreover, the ^{232}Th activity is 26.55±5.88 Bq/kg which is similar to the world average value. ^{226}Ra is one of the progeny of ^{238}U series and the ratio of ^{238}U/^{226}Ra is 1.06 which means that ^{226}Ra and ^{238}U are in seqular equilibrium. The ^{238}U/^{40}K and ^{232}Th/^{40}K ratio ranges from 0.04±0.01 to 0.07±0.02 with the average 0.06±0.01, and 0.08±0.02 to 0.18±0.03 with an average of 0.12±0.02, respectively. The world average for both quotients being 0.067 [2]. This

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value is almost a half of ratio $^{232}Th/^{40}K$, this difference could be related to the low solubility of thorium.

^{137}Cs is an artificial radionuclide produced by nuclear fission. Widespread global distribution of ^{137}Cs into the environment began with high-yield atmospheric tests of atomic weapons in the 1950s and early 1960s. ^{137}Cs and other radionuclides were released into the stratosphere, distributed globally, moved back to the troposphere and from the troposphere to the earth's surface as fallout, which was strongly related to the local precipitation patterns and rates. Temporal and spatial distribution of fallout on the earth's surface has been extensively documented and ^{137}Cs fallout inputs were much greater in the northen hemisphere than in the southern hemisphere, because more atmospheric nuclear tests took place in the northern hemisphere [4]. The concentration of ^{137}Cs is very small in the Jakarta Bay in comparing with the northern hemisphere due to the difference in lattitude. Moreover, the distribution in the local area of Jakarta Bay tend to be higher in the eastern than the western due to the higher fine particle in the eastern, as the ^{137}Cs absorp strongly to the clay minerals. The mean acitivity of ^{137}Cs observed in sediment of Jakarta Bay is 0.77 ± 0.53 Bq/kg.

Table 2. Activity's ratio of natural radionuclides in sediment of Jakarta Bay.

Activity ratio	Mean	Range
$^{232}Th/^{238}U$	2.02 ± 0.07	1.41 ± 0.06 - 2.68 ± 0.11
$^{238}U/^{226}Ra$	1.06 ± 0.16	0.87 ± 0.11 - 1.35 ± 0.27
$^{238}U/^{40}K$	0.06 ± 0.01	0.04 ± 0.01 - 0.07 ± 0.02
$^{232}Th/^{40}K$	0.12 ± 0.02	0.08 ± 0.02 - 0.18 ± 0.03

The radium equivalent activity range from 42.04 ± 6.31 Bq/kg to 110.15 ± 16.52 Bq/kg with the average of 68.14 ± 11.20 Bq/kg. The radiological hazard of natural radionuclides which is indicated by absorbed dose rate, range from 20.47 ± 3.07 nGray/h to 52.52 ± 7.88 nGray/h and the average is 32.06 ± 7.72 nGray/h. This value is lower than the international permissible level of 55 nGray/h [2].

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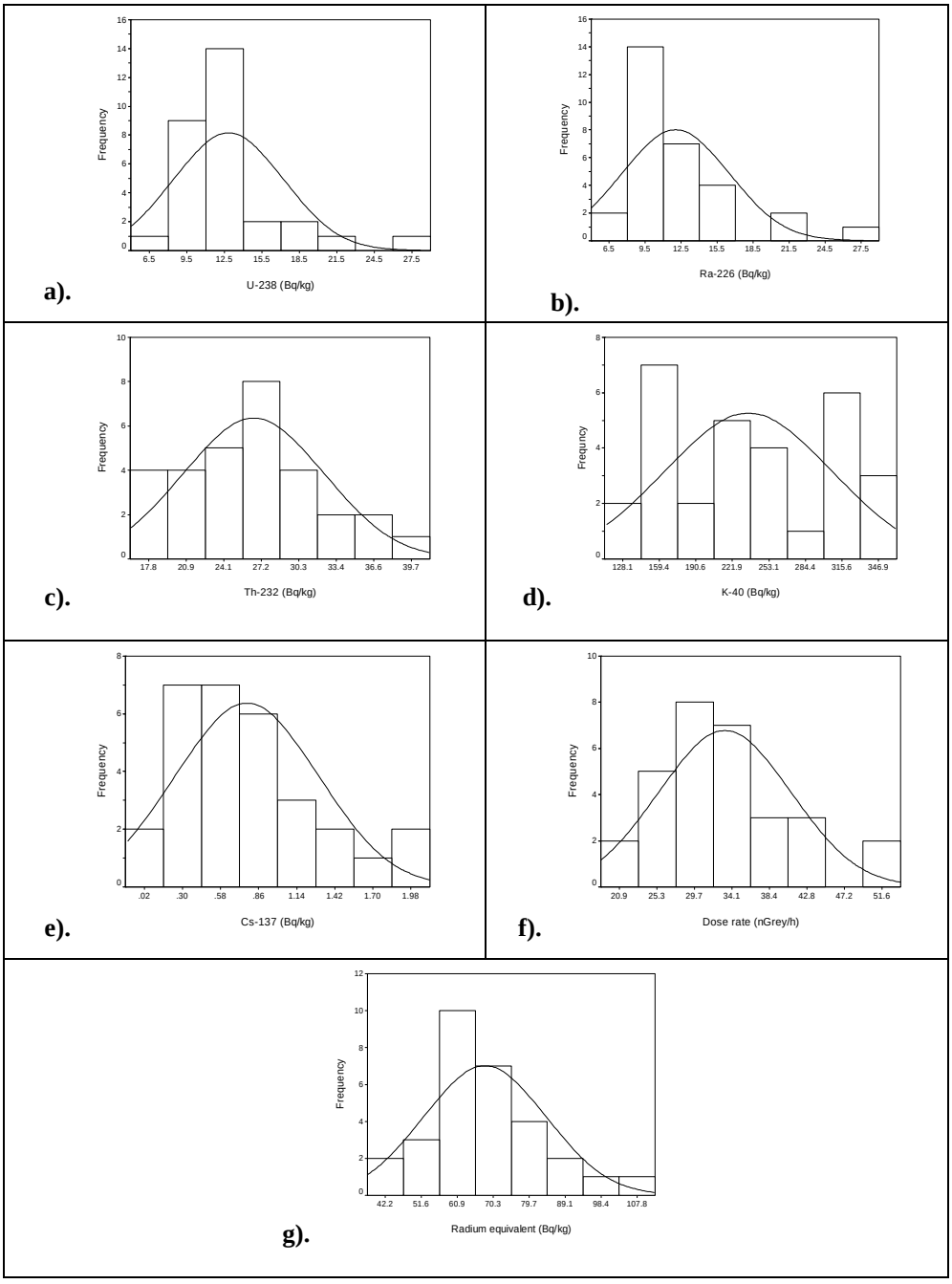

Fig. 3. Normal distributions of natural and antrophogenic radionuclides, dose rate and radium equivalent of surface sediment of Jakarta Bay, a). ^{238}U, b). ^{226}Ra, c). ^{232}Th, d). ^{40}K, e). ^{137}Cs f). Dose rate and g). Radium equivalent.

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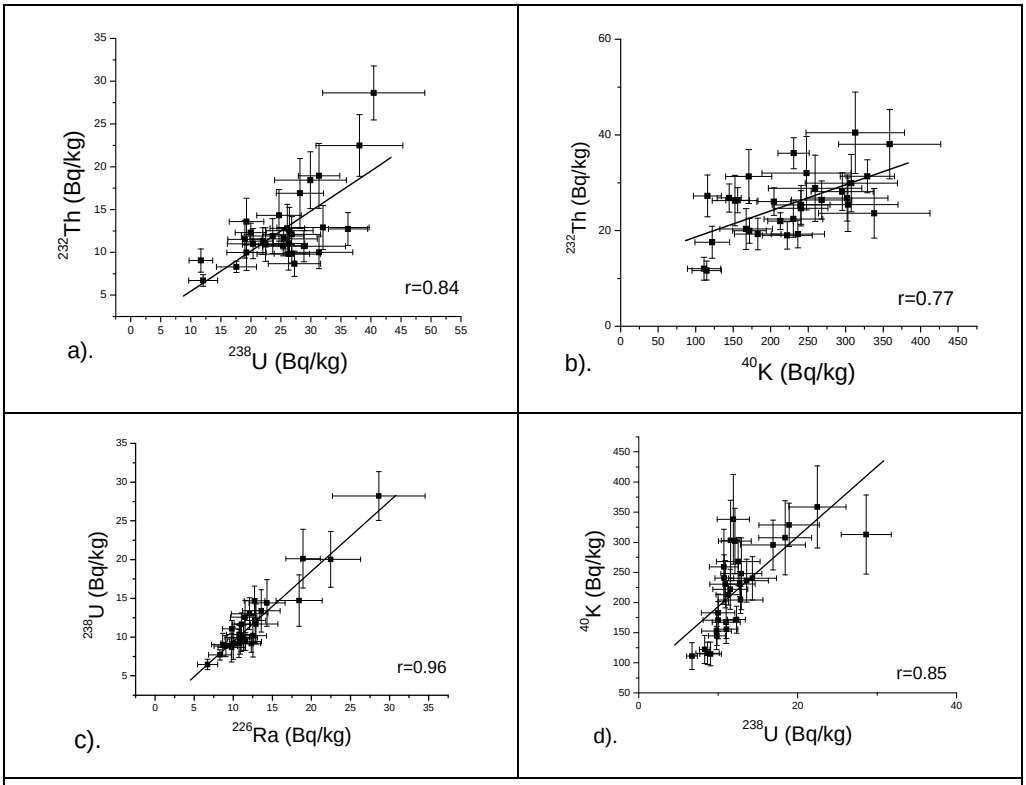

Fig. 4. Linear correlation curve of activity concentration of a). ^{238}U and ^{232}Th, b). ^{40}K and ^{232}Th, c). ^{226}Ra and ^{238}U and d). ^{40}K and ^{238}U (r = correlation factor).

CONCLUSION

Natural and antrophogenic radionuclides distributed in the surface sediment of Jakarta Bay and followed normal distribution based on the statistical analysis (goodness of fit χ^2-test). The concentration of all radionuclides is higher in the eastern than the western part of the bay due to the more clay particle in the eastern. The absorbed dose rate was less than their respective limiting values showing that the surveyed area has no significant hazard from the environment and health point of view.

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