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DISTRIBUTION OF NATURAL RADIONUCLIDES ^{40}K AND ^{208}Tl IN SURFICIAL SEDIMENTS OF SEMARANG WATERS

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ABSTRACT

Distribution of natural radionuclides in surficial sediments of Semarang waters has been carried out by applying the gamma spectrometry analysis. Using the gamma spectrometry technique was employed to analyze the radiation exposure. Six natural radionuclides (^{40}K, ^{208}Tl, ^{212}Pb, ^{214}Pb, ^{228}Ac and ^{226}Ra) can be identified in sediments, along with two natural radionuclides (^{40}K and ^{208}Tl) in water. Distribution factor can be estimated from the ratio of these radionuclides in surficial sediment and the water. Result shows that the distribution factor for radionuclide ^{40}K is 24.5097 - 33.5206 and for ^{208}Tl is 13.4383 - 42.1509.

Keywords: radionuclides distribution, surficial sediments of Semarang waters.

I. INTRODUCTION

Semarang waters is a specific ecosystem. From the radioecological point of view, Semarang estuary has been contaminated by various radioactive and nonradioactive waste from releases of nuclear installation, bedrock trap, deposition from mainland as product of geomorphological process, fall-out from atmosphere and extra-terrestrial radiation (Sasongko, 1997). Indeed, Semarang waters has various functions and allotments: harbour activities, bonded zone, industries and settlements, reclamation area for industrial estate, urban domestic sewage discharge, and fishing activities.

Surface sediments analysis can represent a useful way to evaluate the radionuclides distribution (Aarkrog, 1990), thereby the presence of radionuclides in Semarang waters could hopefully be distinguished whether the sources are natural or anthropogenic in origin. Counting of radionuclides distribution in surficial sediment could determine the radioecological quality of Semarang waters.

This study aims it identifying the nature of radionuclides in Semarang waters, in order to determine the radionuclides distribution found in the surficial sediment.

II. MATERIAL AND METHODS

Sampling location was purposively determined. Each site was chosen to represent the environment of fresh-water/river (A), brackish-water/estuary (B), sea-water (C), and heavily polluted sea-water (D). In each site, 200 litres sample was taken from 3 depths with ratios 0.2, 0.6 and 0.8; then 1 ml of HCl or HNO_3, as fixative, was added to each litre of water.

Following the routine protocol adopted from Nareh and Shaleh (1993), preparation began by filtering the sample through 1μm Millipore paper to let approximately 100 ml filtrate, which were then dripped evenly onto the planset. Having been dried under a 1000 watts light bulb for 6 hours in room temperature (31,3°C), dry sample of approximately 1 mm thick were ready to be counted.

Surface sediments, taken by means of a dredger, were burnt for 3 hours in a muffle furnace of 1500°C in the laboratory. The remaining ashes were evenly spred onto planset to make approximately 1 mm thickness sample for further analysis.

The γ-counting consist of coaxial Ge(Li) detector, supported by Phillips Stabilizer 400 VA, Ortec 4001 Power Supply, Canberrra 2021 Spectroscopy Amplifier, Canberra High Voltage Power Supply and Ortec 7010 Multi Channel Analyzer. This was also equipped by Lead House as a shielding (external cover) and an Omnigraphic Houston Instrument 2000 Plotter-Recorder.

According to Susetyo (1988), stability of the gauge-meter was examined statistically by means of *the least square method*, by repeat test result of observations in the same condition. The efficiency of γ-counting was calculated by energy calibration curve constructed from standard source multigamma ^{152}Eu. The relationship of energy and channel-number was sought by calculating the standard source for which the energy was known exactly. Relationship between the energy and the channel-number was linear.

Measurement of gamma energy was carried out by the γ-spectrometer and the efficiency of counting was also determined by *yield* (absolute intensity). The γ-activity of samples was counted by using the efficiency calibration and was measured by comparing the sample activity to the standard source activity.

According to IAEA (1985), the following equation was applied in calculating "Distribution Factor" (B_p):

$$B_p = (C_p / C_w)$$

where:

$C_{p,i}$ = radionuclides concentration-i in sediments (Bq/kg)

$C_{w,i}$ = radionuclides concentration-i in water (Bq/ltr)

III. RESULT

3.1. Preparation of Water and Sediment

Thickness factor for water in sampling site A was 0.04363 grl/l, 0.04405 grl/l in B, 0.04319 gr/l and 0.04355 gr/l in C and D respectively. Whereas the thickness factor for sedi-ments were 0.8245 in A, 0.8852 in B; 0.8649 and 0.8871 in C and D respectively.

3.2. Curve of Calibration

Curve of calibration of γ-energy produces an equation of line-calibration, i.e. **Y = 0.5658 X - 0.3157.** Meanwhile regression line of calibration efficiency was **Y = -0.99 X - 0.95**

3.3. Radionuclides Identification

The isotopes table of Erdtmann (1976) and Erdtmann & Soyka (1979) was used to identify the presence of radionuclides in this study. Accordingly, the results are given in Table 1 and 2.

Table 1. The identification of radioinuclides in the water

Location	Energy (keV)	Intensity (%)	Isotopes	Half-life	Sources/series
A	510.7	22.5	Tl-208	3.1m	Th-232
	1460.7	10.7	K-40	1.28E+9y	Natural
B	510.7	22.5	Tl-208	3.1 m	Th-232
	1460.7	10.7	K-40	1.28E+9y	Natural
C	510.7	22.5	Tl-208	3.1 m	Th-232
	1460.7	10.7	K-40	1.28E+9y	Natural
D	510.7	22.5	Tl-208	3.1 m	Th-232
	1460.7	10.7	K-40	1.28E+9y	Natural

Table 2. The identification of radioinuclides in the sediments

Location	Energy (keV)	Intensity (%)	Isotopes	Half-life	Sources/series
A	186.2	3.28	Ra-226	1600y	U-238
	238.6	43.10	Pb-212	10.64h	Th-232
	351.9	37.10	Pb-214	26.8m	U-238
	510.7	22.50	Tl-208	3.1m	Th-232
	583.1	86,00	Tl-208	3.1m	Th-232
	1460.7	10.70	K-40	1.28E+9y	Natural
B	238.6	43.10	Pb-212	10.64h	Th-232
	351.9	37.10	Pb-214	26.8m	U-238
	510.7	22.50	Tl-208	3.1m	Th-232
	583.1	86,00	Tl-208	3.1m	Th-232
	1460.7	10.70	K-40	1.28E+9y	Natural
C	238.6	43.10	Pb-212	10.64h	Th-232
	295.2	19.20	Pb-214	26.8m	U-238
	351.9	37.10	Pb-214	26.8m	U-238
	510.7	22.50	Tl-208	3.1m	Th-232
	583.1	86,00	Tl-208	3.1m	Th-232
	911,0	29,00	Ac-228	6.13h	Th-232
	1460.7	10.70	K-40	1.28E+9y	Natural
D	238.6	43.10	Pb-212	10.64h	Th-232
	351.9	37.10	Pb-214	26.8m	U-238
	510.7	22.50	Tl-208	3.1m	Th-232
	583.2	86,00	Tl-208	3.1m	Th-232
	911,0	29,00	Ac-228	6.13h	Th-232
	1460.7	10.70	K-40	1.28E+9y	Natural

3.4. Radioactivities and the Concentration of Radioisotopes in the Water and Sediment

The result of γ-activities and the concentration of radioisotopes in the water and sediment was given on Tables 3 to 6.

Table 3. The activities and the concentration of ^{40}K $^{(1460.7\ keV)}$ in the water

Location	Sample weight	Net count (cps)	Efficienc (%)	Activities (Bq)	Atomic numbers	Weight (gr)	Activity (Bq/gr)	Concent. (%)
A	20.508	0.0075	2.85E-4	245.94	1.43E+19	09.53E-4	11.99	4.65E-4
B	24.708	0.0075	2.85E-4	245.94	1.43E+19	09.53E-4	09.95	3.86E-4
C	24.296	0.0085	2.85E-4	278.73	1.62E+19	10.80E-4	11.26	4.36E-4
D	24.748	0.0075	2.85E-4	245.94	1.43E+19	09.53E-4	09.94	3.85E-4

Table 4. The activities and the concentration of ^{40}K $^{(1460.7\ keV)}$ in the sediments

Location	Sample weight	Net count (cps)	Efficiency (%)	Activities (Bq)	Atomic numbers	Weight (gr)	Activity (Bq/gr)	Concent. (%)
A	20.033	0.0095	2.85E-4	311.53	1.82E+19	12.07E-4	15.55	6.03E-4
B	20.043	0.0075	2.85E-4	245.94	1.43E+19	09.53E-4	12.27	4.75E-4
C	20.027	0.0085	2.85E-4	278.73	1.62E+10	10.79E-4	13.92	5.39E-4
D	20.038	0.0100	2.85E-4	327.92	1.91E+19	12.70E-4	16.36	6.34E-4

Table 5. The activities and the concentration of ^{208}Tl $^{(510,7\ keV)}$ in the water

Location	Sample weight	Net count (cps)	Efficiency (%)	Activities (Bq)	Atomic numbers	Weight (gr)	Activity (Bq/gr)	Concent. (%)
A	20.508	0.0220	8.06E-4	121.31	32560	1.12E-17	5.92	5.46E-17
B	24.708	0.0170	8.06E-4	93.74	25160	0.87E-17	3.79	3.52E-17
C	24.296	0.0195	8.06E-4	107.53	28860	0.99E-17	4.43	4.10E-17
D	24.748	0.0170	8.06E-4	93.74	25160	0.87E-17	3.79	3.85E-17

Table 6. The activities and the concentration of ^{208}Tl $^{(510,7\ keV)}$ in the sediments

Location	Sample weight	Net count (cps)	Efficiency (%)	Activities (Bq)	Atomic numbers	Weight (gr)	Activity (Bq/gr)	Concent. (%)
A	20.033	0.0135	8.06E-4	74.44	19980	0.69E-17	3.72	3.45E-17
B	20.043	0.0110	8.06E-4	60.66	16280	0.56E-17	3.03	2.81E-17
C	20.027	0.0230	8.06E-4	126.83	34040	1.18E-17	6.33	5.87E-17
D	20.038	0.0255	8.06E-4	140.61	37740	1.30E-17	7.02	6.51E-17

3. 5. Distribution factor of ^{40}K and ^{208}Tl in the sediments

The distribution factor of ^{40}K and ^{208}Tl in the sediment is given in Table 7.

Table 7. Distribution factor of ^{40}K and ^{208}Tl

No	Location	$C_{w,i}$ (Bq/l)$^{(1)}$	$C_{s,i}$ (Bq/gr)$^{(2)}$	$K_{d,i}$ (l/gr)$^{(3)}$
Based on gross-α				
1	A	09,7708E-3	0,1292	13,2231
2	B	10,4649E-3	0,0517	4,9403
3	C	09,4219E-3	0,0568	6,0285
4	D	08,9219E-3	0,0675	7,5657
Based on gross-β				
1	A	0,8807E-3	0,2685	304,8711
2	B	6,8487E-3	0,4697	68,5824
3	C	5,3309E-3	0,5051	94,7495
4	D	4,1238E-3	0,3927	95,2277
Radionuclide ^{40}K				
1	A	0,5231	12,8210	24,5097
2	B	0,4383	10,8614	24,7807
3	C	0,4371	12,0394	27,5438
4	D	0,4329	14,5130	33,5206
Radionuclide ^{208}Tl $^{(4)}$				
1	A	0,2583	3,4711	13,4383
2	B	0,1669	3,5497	21,2684
3	C	0,2155	5,8986	27,3717
4	D	0,1650	6,9549	42,1509

Note: (1). $C_{w,i}$ = activity in water
(2). $C_{s,i}$ = activity in sediments
(3). $K_{d,i}$ = distribution factor
(4). Sum of 510.7 keV and 583.1 keV

IV. DISCUSSION

The existence of radionuclides in Semarang waters can be identified by using gamma-spectrometry methods which was identified from eight γ-peak energy (Table 1 and Table 2). From the characteristics of gamma energy, 6 radionuclides of natural sources can be identified, i.e.: ^{40}K (1460.7 keV), from natural decaying, ^{208}Tl (510.7 keV) from natural decaying series of ^{232}Th, ^{212}Pb (238.6 keV) from natural decaying series of ^{232}Th, ^{214}Pb (195.4 keV and 351.9 keV) from natural decaying series of ^{238}U, ^{226}Ra (186.2 keV) from natural decaying series of ^{238}U, and ^{228}Ac (911.0 keV) from natural decaying series of ^{232}Th.

To estimate the distribution factor of these radionuclides in surficial sediment their presence should be identified from both water and sediment samples. Table 1 shows that only ^{40}K and ^{208}Tl found in the water, while others can be detected only in the sediments studies. This might be in line with what Odum (1993) state that the identification of radionuclides and description of its pathways and behavior is a part of marine radio-ecology (or radiation ecology) which correlate the radioactive compound, type of radiation and the ecology of marine environment. Those six natural radionuclides mentioned may come from cosmic rays and/or natural materials. The largest natural radiation contributed by radionuclide ^{40}K (1.28×10^9 y), ^{238}U (4.56×10^9 y), decaying series of ^{232}U (8.8×10^8 y) with 0.714% content, decaying series of ^{238}U (9.56×10^9 y) with 99.238% content, and ^{234}U radionuclides (2.48×10^9 y). Marine environment may contribute less than 1% from ^{210}Po in fish and shellfish (UNSCEAR, 1988).

Beiser (1987) stated that radioactive elements are unstable elements and therefore always emit radioactive rays to be stable. In doing so, they create radioactive series that show the change of elements and type of radiation. In nature, there are four radioactives series characterized by different mass-number of nuclides, they are 4n, 4n+1, 4n+2 and 4n+3, where n is integer. Each series consist of succession of daughter products, all ultimately derived from a single parent nuclide.

Anthropogenic radioactives made by nuclear reaction have now been widely used. This in turn, increases the background radiation on the environment, coming from global fall-out, the operation of both military and civilian or the operations and accident of nuclear power plant, fuel reprocessing and the use of radioisotopes in various living field (Aarkrog, 1988).

According to Smith (1984), radionuclides behaviour in aquatic environment might be performed by various complex models within mathematical representation. The model developed in accordance to the various types of water bodies are e.g.: river, estuary, and sea. Physical conditions affecting the distribution of radionuclides in marine environment are temperature, density and salinity of water, current pattern, wave pattern, as well as the depth of water.

In the water, radionuclides will be diluted and spread, and presumably transfered to biological matter, sediments and suspended particles. Among others, these factors affect the concentration of radionuclides in aquatic environment: mixing process, distribution and interaction with sediments and biological matters (IAEA, 1982). According to Ophel (1977), the concentration of radionuclides in the water is determined

by the distribution, transfer route and decay factors.

The most important parameter concerning radionuclides distribution in aquatic environment is the movement of water mass. Whilst in the estuary, it depends on size, food-stuff production and interaction of river-to-sea waters. Radionuclide elements which were absorbed in small amount by the sediment have a probability to be spread widely (Ophel, 1977).

Foodweb has an important role because each chain absorbs radionuclides, and interchain transfer process of radionuclides has a constant absorbtion factor. To monitor radiation contamination in aquatic environment, one can utilize surface sediment indicator by monitoring of samples as indicator in regular time interval (Dahlgaard, 1991).

V. CONCLUSION

The results of radionuclides identification by γ-spectrometry method shows that there are six radionuclides from eight identified γ-energy peaks. All of these identified radionuclides in Semarang waters are natural in origin. Therefore, on the basis of this data, Semarang waters have not been yet contaminated by anthropogenic radionuclides either from the activation or fision processes. However, radionuclide ^{40}K (1460 keV) and ^{208}Tl (510.7 keV) identified from both kind of samples (water and sediment) which indicate various activities and contents as well as numerous elements.

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