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Original Paper

DISPERSION MODELING OF NATURAL RADIONUCLIDES ^{238}U, ^{232}TH, ^{226}RA, ^{40}K IN MURIA COASTAL WATERS

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ABSTRACT

Dispersion modeling of natural radionuclides ^{238}U, ^{232}Th, ^{226}Ra, ^{40}K in Muria coastal waters has been carried out in sea water and sediment surounding Tanjungjati B coal-fire power plant and nuclear power plant site's candidate by applying the hydrodynamics model of unsteady 2-dimensional flexible grid. Oceanography data collecting of bathimetry, current, wave, tide and wind had been carried out on May 28, 2006 until June 2006. Updating data was conducted on April 27 up to April 29, 2011 by using Acoustic Doppler Current Meter Profiler (ADCP) to measure the wave and subsurface current with duration of 2x24 hours. Sea water and sediment samples were collected on April 22, 2011 in six locations (surounding Tanjungjati CPP) and on April 23, 2011 in 10 locations (surounding NPP site's candidate). Samples were analyzed at Research Center for Safety Technology and Radiation Metrology Laboratory, National Nuclear Energy Agency, Jakarta on May 2011 until September 2011 by using spectrometri-γ analysis. Result shows that it can be identified and measured the natural radionuclides of ^{238}U, ^{232}Th, ^{226}Ra, ^{40}K in sea water and sediment. The study can be justified that natural radionuclides of ^{238}U, ^{232}Th, ^{226}Ra, ^{40}K was leachated from fly ash and bottom ash of coal burned Tanjungjati CPP to sea water. The hiyrodynamics model of unsteady 2-dimensional fexible grid by using CD Oceanography software for current plotting, ArcView GIS 3.3 software for bathimetric contouring and SMS 8.1 software for modeling of natural radionuclides dispersion in coastal waters one can applied for radionuclides dispersion of ^{238}U, ^{232}Th, ^{226}Ra, ^{40}K in Muria coastal waters.

Keywords: Hydrodynamics model; natural radionuclides; Muria coastal waters

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INTRODUCTION

Muria coastal waters were contaminated by radioactive and nonradioactive waste from releases of coal-fired power plant, bedrocks trap, deposition mainland as product of geomorphological process, fall out from atmosphere or extraterrestrial radiation. Indeed, coastal waters has various functions and activities: harbour activities, coal-fired power plant, industries, settlements, domestic sewage discharge and fishing activities. The present of radionuclides in Muria coastal waters could be distinguished wether the sources are naturally or anthropogenic in origin. Dispersion modeling of natural radionuclides in sea water and sediment could determine the radioecological quality of coastal waters. The study aims to apply the hydrodynamics model of unsteady 2-dimensional fexible grid to analyze the dispersion of natural radionuclides of ^{238}U, ^{232}Th, ^{226}Ra,^{40}K in sea water and sediment.

MATERIALS AND METHODS

Sampling location was purposively determined. Sea water and sediment sample were collected on April 22, 2011 in six locations surrounding Tanjungjati CPP and on April 23, 2011 in 10 locations surrounding NPP site's candidate. Samples were analyzed in Research Center for Safety Technology and Radiation Metrology Laboratory, National Nuclear Energy Agency, Jakarta. Sample preparation was conducted on May to July 2011, radionuclides counting was done on August to September 2011.

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In each site, 2x20 l of water sample was taken from 3 depths i.e. 0.2d, 0.6d, 0.8d, was added 1 ml of HNO_3 to each volume of water as fixative (Nareh and Shaleh, 1993). Preparation began by filtering the sample through 1 μm millipore paper. In each site, 2 kg of sediment sample was taken from sea bottom by using a dredger. The samples were then burnt for 24 hours in a furnace at 105^0C. The remaining ashes were evenly filtered by using 100 mesh onto Marinelli 1 l for further analysis.

The γ-counting consists of HP Ge detector equipped by lead house as a shielding. According to Susetyo (1988), stability of the gauge-meter was examined statistically, efficiency of γ-counting was calculated by energy calibration curve constructed from multi-γ standard source of ^{152}Eu. The (linearly) relationship of energy and chanel-number was sought by calculating the standard source for which the energy was known exactly. Measurement of γ-energy was carried out by the γ-spectrometer and the counting efficiency was also determined by yields (absolute intensity). The γ-activity of sample was counted by using the efficiency calibration and was measured by comparing the sample activity to the standard activity.

Oceanography data collecting was conducted since 2006 for bathimetric, current, wave, tide and wind (May 28, 2006 until June 2006) after secondary data was collected. This data then be updated on April 27 until April 29, 2011 by using *Acoustic Doppler Current Meter Profiler* (ADCP) *Sontek Argonaut-XR Extended Range* to measure the waves and current with duration of 2x24 hours.

Wind data which will be converted into wave-rose was collected from 2000 to 2010 from Semarang Station of Meteorology. Tide measurement was conducted each 30 minutes of 24-hours for 30 days (in 2006) and completing with tides data from LPWP Undip Station (January 1 to April 30, 2011). Bathimetric survey was done in 2006 by using GPS and echosounder, measured for 1600 location (grid size: 100 m x 100 m).

Wave analysis based on wind data was conducted by using Sverdrup-Munk-Breschneider method. Wave modeling was conducted by using CEDAS software ST-WAVE module for 3-direction wave probability (northwest, north, northeast). Bathimetric contouring was done by using ArcView GIS 3.3 software. Current plotting was done by using CD Oceanography software. Harmonic constant of tide was calculated by using admiralty method. Mathematical modeling for natural radionuclides dispersion of ^{238}U, ^{232}Th, ^{226}Ra, ^{40}K in coastal waters was constructed by using SMS (*Surface-water Modeling System*) version 8.1. Contouring of radionuclides activity in sea water was conducted by using RMA4 Module and contouring in sediment was conduct by using Sed2D Module (US Army, 2001, 2002, 2003a, 2003b, 2003c).

Time interval for modeling was taken at Δt = 1 h, T total = 360 hours (15 days). Grid size, dx and dy, near source 10 m, outer border 100 - 1500 m. River flowrate 5 m^3/s, *outlet* flowrate 73 m^3/s. Radionuclides releases source was assumed from leaching of *fly ash* and *bottom ash* in 0,6 d *depth*. Advection-diffusion coeffisient was taken per 10 m.

RESULTS AND DISCUSSION

Results

Current velocity pattern in location of ADCP can be divided into: average velocity, surface velocity (0.2d=2.32 m in cell-4: 0.3–2.3 m), mid depth velocity (0.6d=6.96 m in cell-3: 6.3–8.3 m) and bottom-velocity (0.8d =9.28 m in cell-2: 8.3-10.3 m). Monogram the current velocity pattern presented in **Fig. 1 and Fig. 2**.

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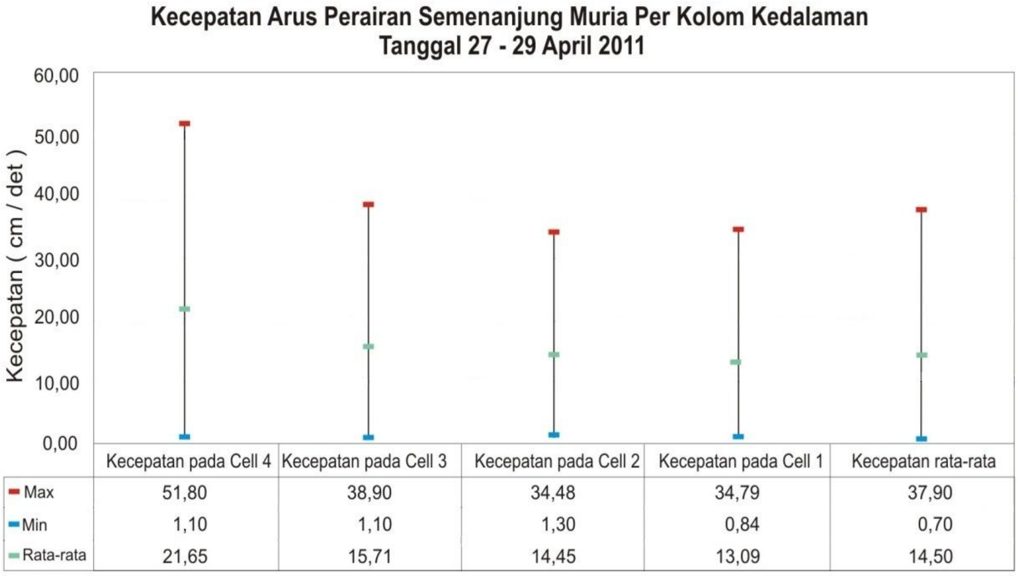

Fig. 1. Current velocity per depth column read by ADCP (April, 2011)

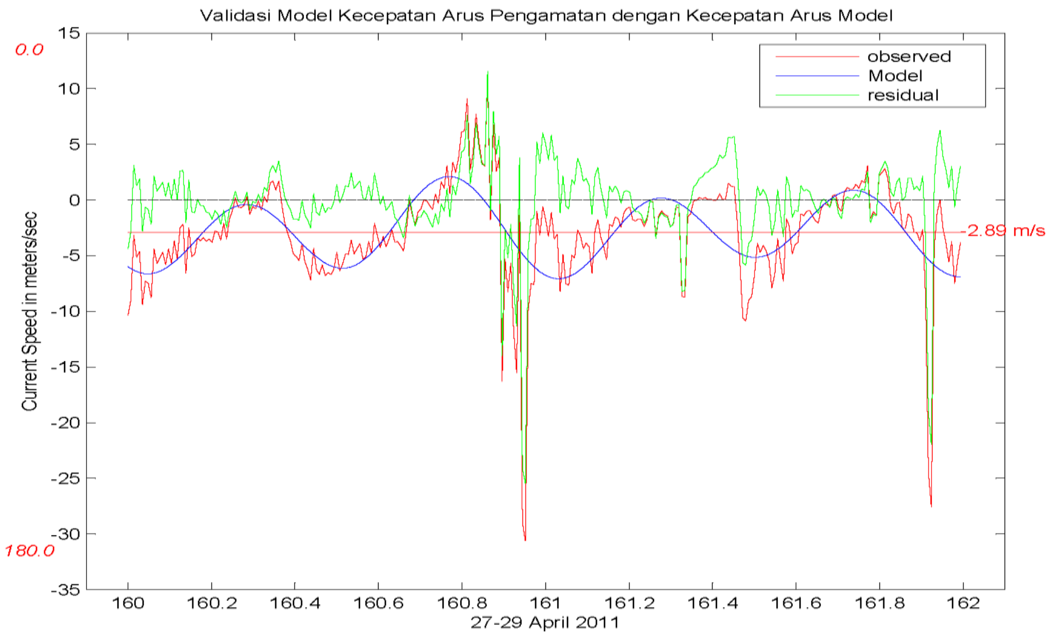

Fig. 2. Validation for current velocity

The harmonic constants were calculated by using *Admiralty* method (**Table 1**).

Table 1. The harmonic constants of tide

Constant	S_0	M_2	S_2	N_2	K_1	O_1	M_4	MS_4	K_2	P_1
A (cm)	82.28	9.94	4.79	3.77	27.34	9.40	0.87	0.77	1.29	9.02
g°	-	34.77	270.66	181.91	78.10	8.03	202.29	344.56	270.66	78.10

M_2 is semi diurnal tide, by moon; S_2 is semi diurnal tide, by solar gravity; N_2 is semi diurnal tide, by moon path changes; K_1 is diurnal tide, by moon and sun declination; O_1 is diurnal, by moon declination changes; M_4 is angular velocity twice of M_2; MS_4 is

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interaction product of S_2 and M_2; K_2 is affected by earth-sun revolution distance; P_1 is diurnal tide, by sun declination change; A is amplitude; g^o is phase.

Based on the Formzahl formula it can be determined the value of F = 2.49, HHWL = 149.48 cm, MSL = 82.28 cm, LLWL = 15.08 cm. Simulation results of natural radionuclides dispersion modeling in sea water and sediment in the study site can be presented in **Fig.3** until **Fig. 10.**

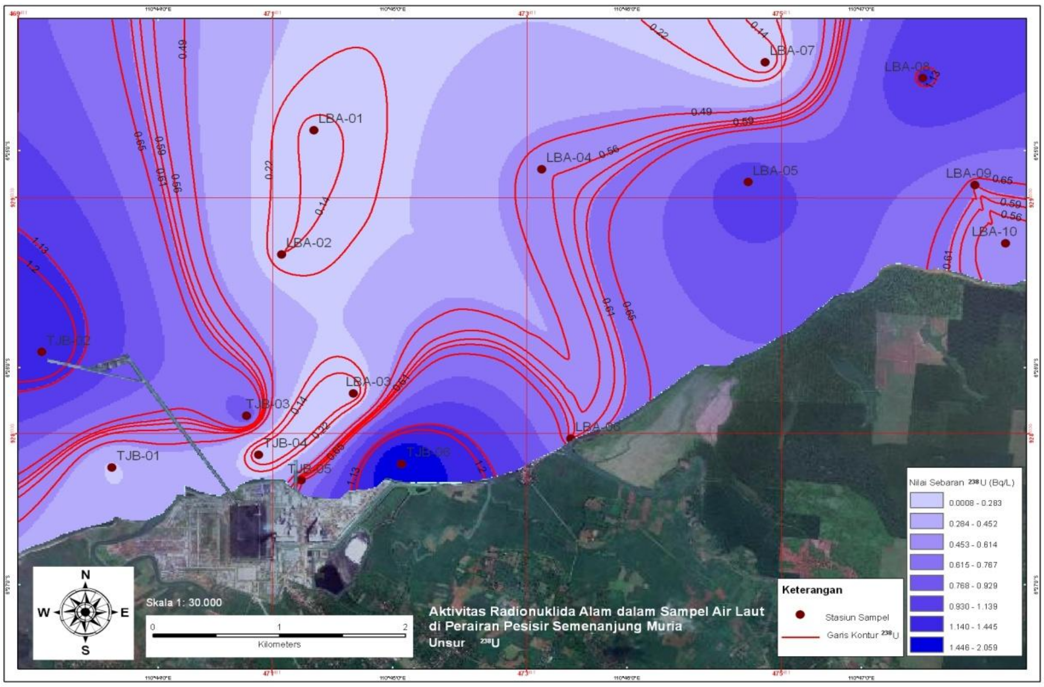

Fig. 3. Iso-Activity of ^{238}U in Sea Water Sample

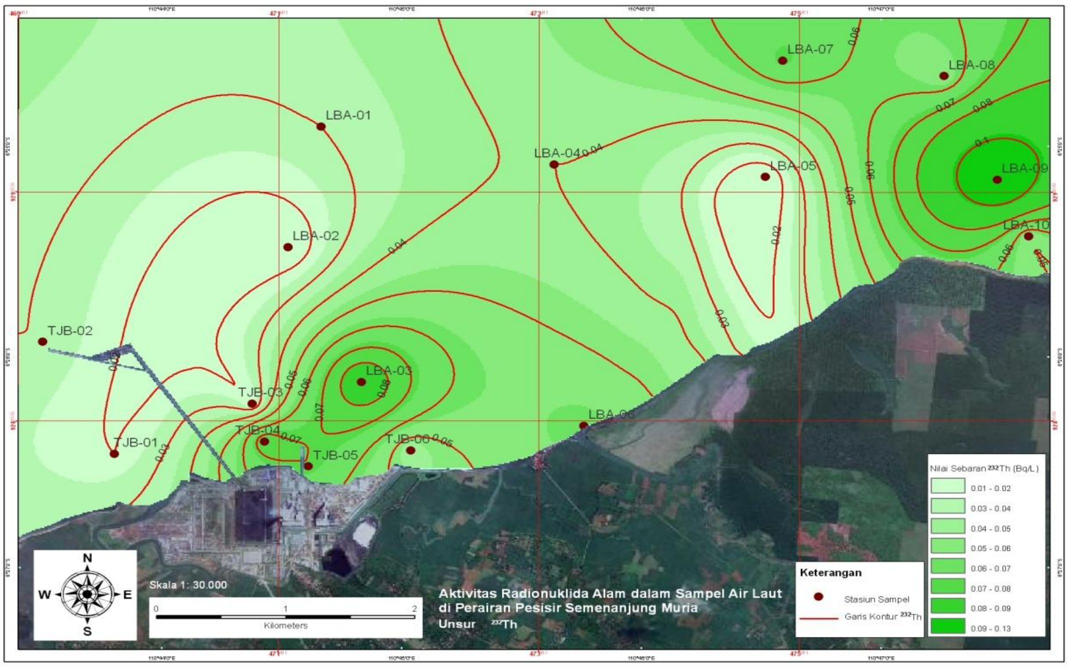

Fig. 4. Iso-Activity of ^{232}Th in Sea Water Sample

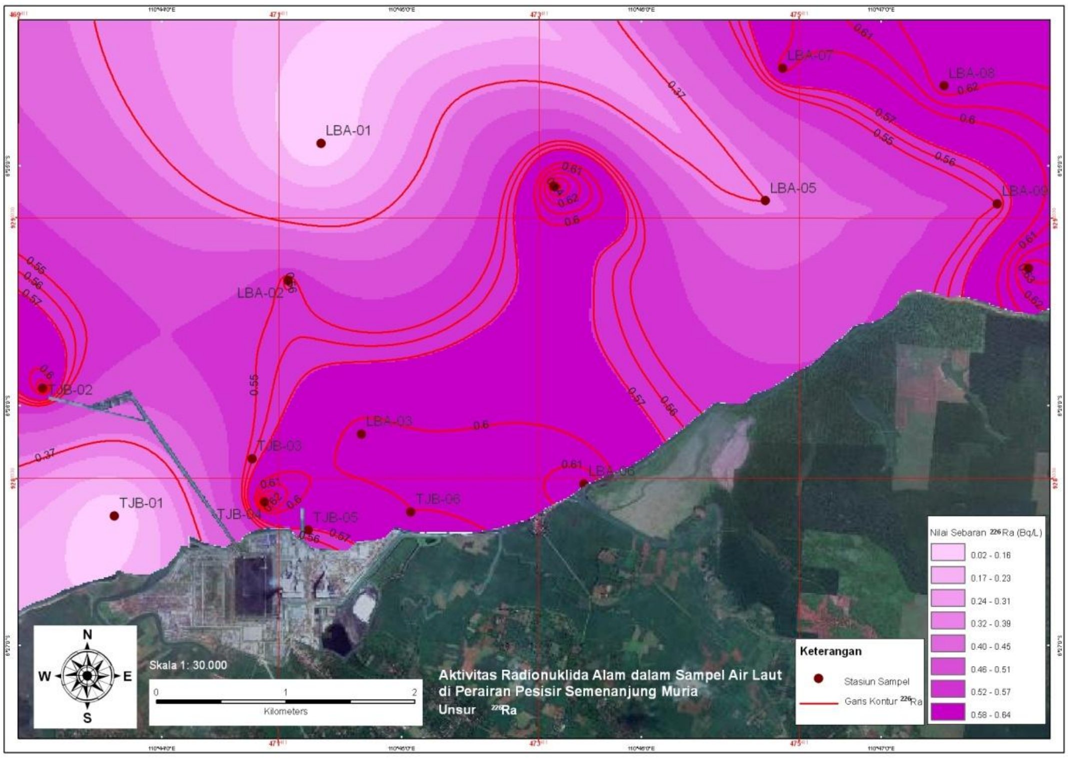

Fig. 5. Iso-Activity of ^{226}Ra in Sea Water Sample

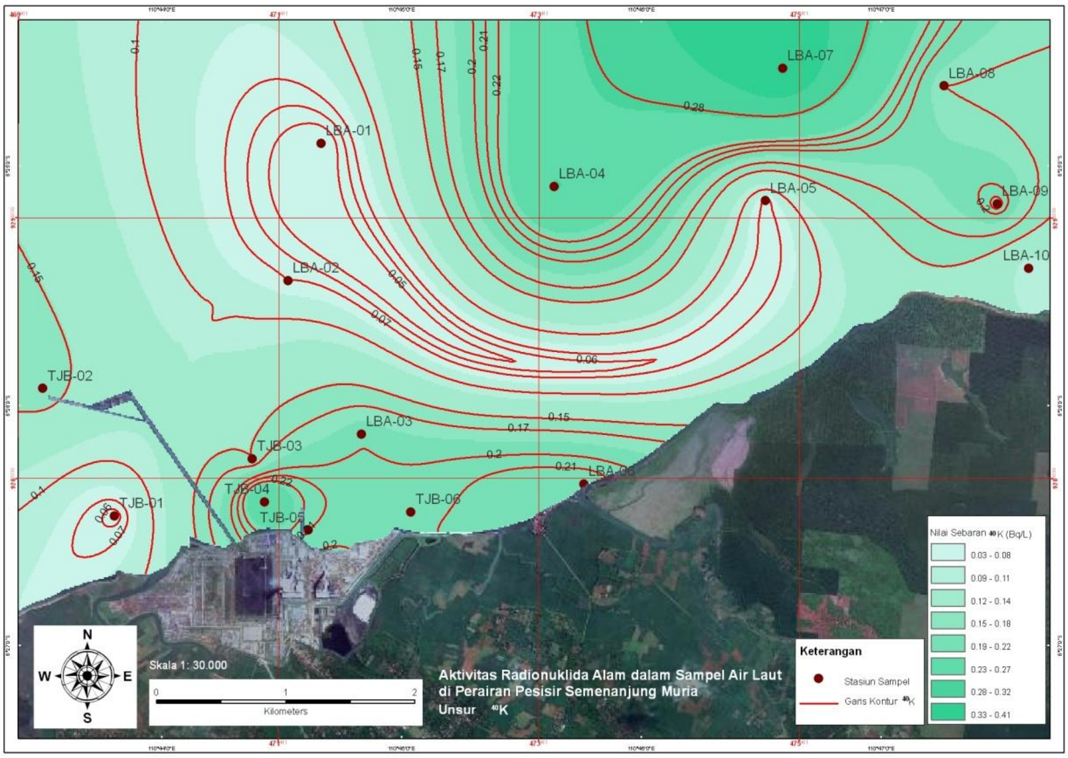

Fig. 6. Iso-Activity of ^{40}K in Sea Water Sample

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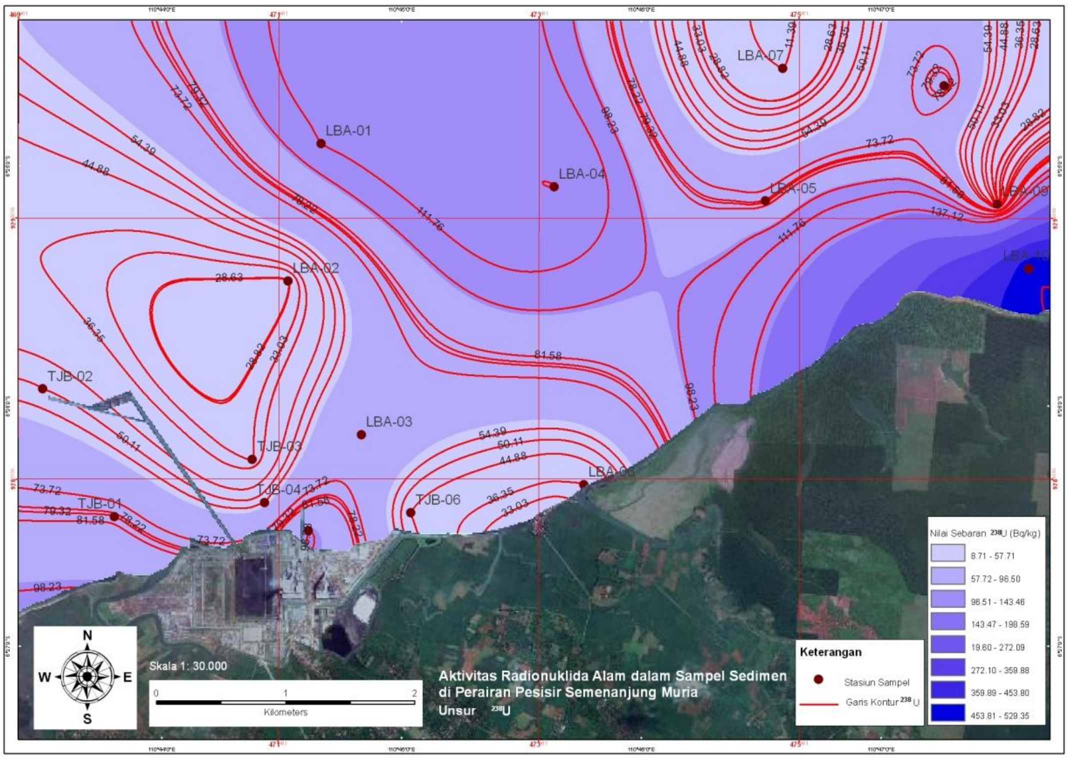

Fig. 7. Iso-Activity of ^{238}U in Sediment Sample

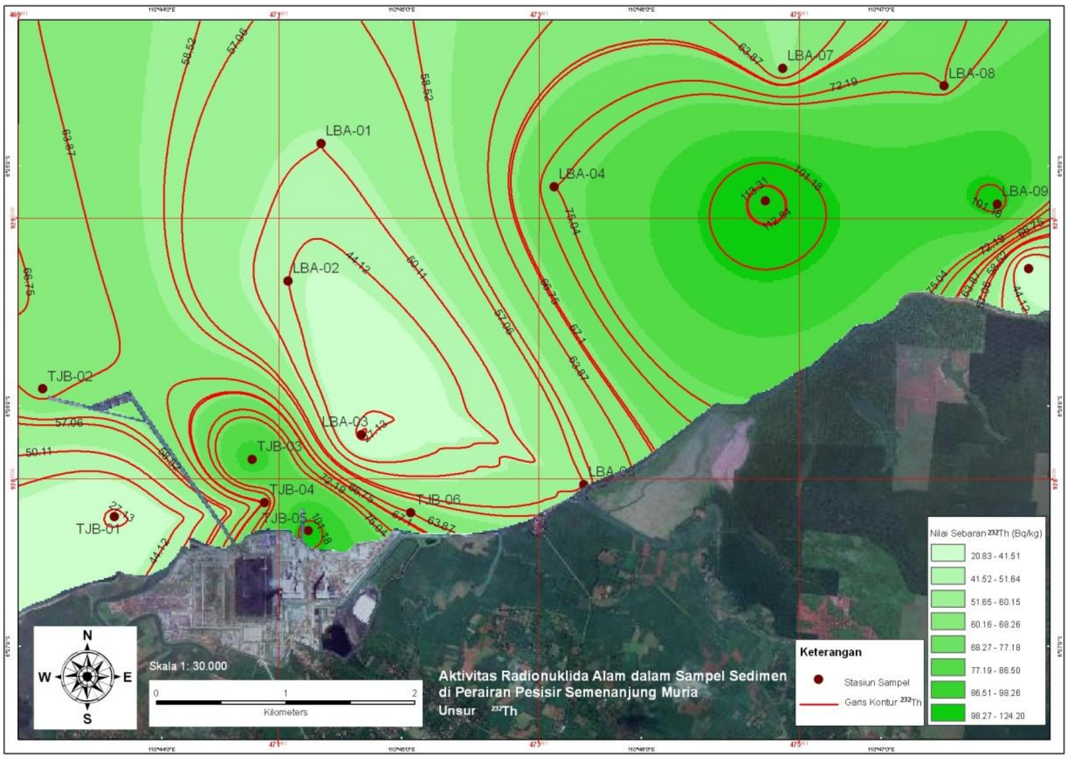

Fig.8. Iso-Activity of ^{232}Th in Sediment Sample

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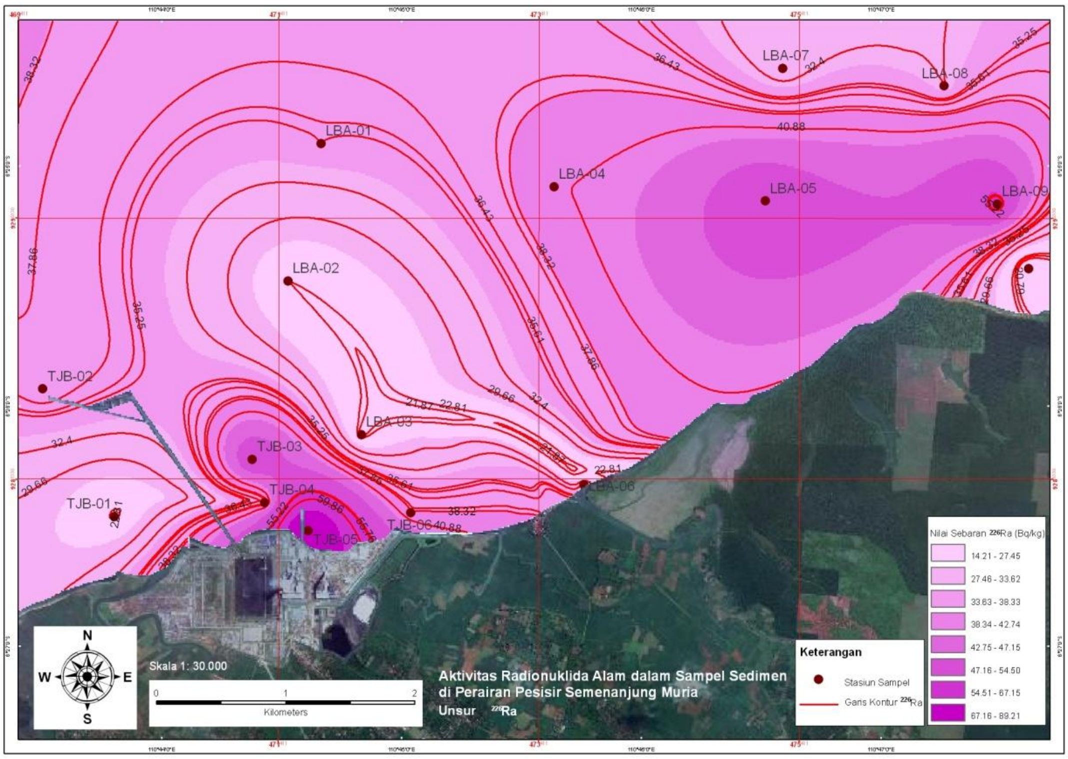

Fig. 9. Iso-Activity of ^{226}Ra in Sediment Sample

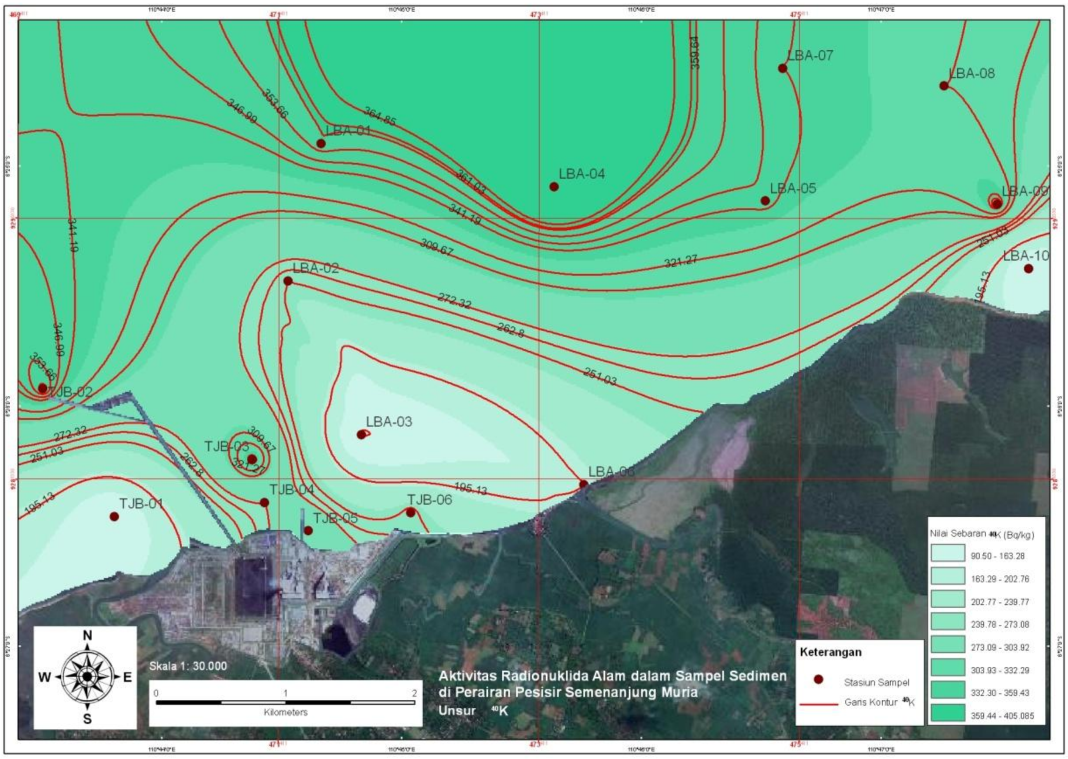

Fig. 10. Iso-Activity of ^{40}K in Sediment Sample

Discussion

Muria coastal waters is a part of *coastal sediment cell* 4 of Central Java *Coastal Cell* (DKP, 2003). According to Tanjungjati B coal-fired power plant (2007), Tanjungjati B CPP was operated with capacity of 2x660 MW with coal consumption of 305 tons/hour/unit with ash content 4.8%. Coal burning in CPP will produced fly ash and bottom ash which can be leacheated to coastal waters. Mass-water movement will disperse that leachate to sea water, suspend in suspended-load sediment, deposite to bed-load sediment and will accumulated by biota.

According to Marinkovic, *et al.*, (2010), coal as mining product from earth mantle contains natural radionuclide of ^{238}U, ^{232}Th, ^{226}Ra and ^{40}K which can increasing the environment radiation exposure and marine pollution. He shows that contain of ^{238}U (129 ± 36 Bq/kg in *fly ash* and 161 ± 42 Bq/kg in *bottom ash*), ^{232}Th (86 ± 9 Bq/kg in *fly ash* and 63 ± 6 Bq/kg in *bottom ash*), ^{226}Ra (126 ± 13 Bq/kg in *fly ash* and 86 ± 8 Bq/kg in *bottom ash*), ^{40}K (396 ± 40 Bq/kg in *fly ash* and 358 ± 36 Bq/kg in *bottom ash*) (**Table 2**).

Table 2. Spectrometry-γ analysis of coal ash in Nicola Tesla CPP

Nuclides	*Fly ash* (Bq/kg)	*Bottom ash* (Bq/kg)	Standard
^{238}U	129 ± 36	161 ± 42	-
^{235}U	10 ± 1	6.3 ± 0.8	-
^{40}K	396 ± 40	358 ± 36	5000$^{(*)}$
^{226}Ra	126 ± 13	86 ± 8	400$^{(*)}$
^{232}Th	86 ± 9	63 ± 6	300$^{(*)}$

Source: Marinkovic *et al.*, 2010.
(*) According to Polish BN-87/6713-02

Study of Uslu and Gokmese (2010) over 11 CPPs in Turkey which can detected natural radionuclides in coal, *fly ash* and *bottom ash*, coming from 3 decaying series: Uranium-Radium (^{238}U series), Uranium-Actinium (^{235}U series), Thorium (^{232}Th series) and ^{40}K. After burned, coal volume will decreased, so the natural radionuclides contains in *fly ash* and *bottom ash* increasing.

Table 3. Activities in fly ash vs coal (Bq/kg)

Radionuclides	^{238}U	^{238}Th	^{40}K
Coal	20	20	50
Fly ash	200	70	265
Concentration factor	10	35	5,3

Source: Uslu dan Gokmese, 2010.

The study of Marinkovic, *et al.*, (2010), Uslu and Gokmese (2010) give the justification of natural radionuclides in coal, *fly ash* and *bottom ash* from coal burning. Orescanin, *et al.*, (2005) and Orescanin, *et al.*, (2006) showed the influence of *fly ash* and *bottom ash* deposition on sediment quality of Kastela Bay and also detected the natural radionuclides ^{238}U, ^{232}Th, ^{226}Ra, ^{40}K in sediment. This study give a justification of leaching the natural radionuclides ^{238}U, ^{232}Th, ^{226}Ra, ^{40}K to sea water, suspended-load and bed-load sediment.

Mljak dan Krizman (1996) found the natural radionuclides ^{226}Ra, ^{238}U, ^{232}Th, ^{40}K and ^{210}Pb in Paka river leaching from Sostanj CPP, Slovenia. This study give a justification that natural radionuclides leaching from *fly* ash and bottom ash can exist in fluvic and limnic environment.

The leaching of natural radionuclides in coastal waters can be justified from this study (**Table 4** and **Table 5**).

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Table 4. Activities of natural radionuclides in sea water samples of Muria coastal waters (Bq/l)

Location	^{238}U	^{232}Th	^{226}Ra	^{40}K
TJB-01	0,22	0,02	0,02	0,05
TJB-02	1,48	0,03	0,63	0,17
TJB-03	0,99	0,02	0,56	0,15
TJB-04	0,00	0,08	0,65	0,28
TJB-05	0,65	0,07	0,57	0,21
TJB-06	2,08	0,04	0,60	0,21
LBA-01	0,09	0,03	0,03	0,03
LBA-02	0,14	0,01	0,57	0,07
LBA-03	0,00	0,10	0,61	0,20
LBA-04	0,56	0,04	0,65	0,28
LBA-05	1,13	0,01	0,37	0,06
LBA-06	0,59	0,06	0,62	0,22
LBA-07	0,00	0,07	0,61	0,42
LBA-08	1,20	0,05	0,63	0,17
LBA-09	0,61	0,14	0,55	0,22
LBA-10	0,49	0,05	0,64	0,10

Table 5. Activities of natural radionuclides in sediment samples of Muria coastal waters (Bq/kg)

Location	^{238}U	^{232}Th	^{226}Ra	^{40}K
TJB-01	79.32	22.24	21.87	96.68
TJB-02	50.11	66.75	37.86	364.85
TJB-03	33.03	101.18	55.76	353.66
TJB-04	54.39	67.10	36.43	272.32
TJB-05	98.23	113.31	89.77	309.67
TJB-06	44.88	63.87	38.32	262.80
LBA-01	111.76	50.11	35.25	359.64
LBA-02	28.63	44.12	22.81	251.03
LBA-03	73.72	20.29	20.79	87.95
LBA-04	137.12	75.04	40.88	405.13
LBA-05	78.22	124.31	55.22	346.99
LBA-06	36.35	57.06	35.61	195.13
LBA-07	11.39	58.52	29.66	341.19
LBA-08	81.58	72.19	32.40	321.27
LBA-09	28.82	112.84	59.86	361.03
LBA-10	520.11	27.13	14.06	93.15

According to **Table 4** and **Table 5,** coal as mining product from earth mantle contains natural radionuclides ^{238}U, ^{232}Th, ^{226}Ra and ^{40}K. It can be concluded that the natural radionuclides which dispersed in coastal waters were coming from coal, *fly ash* and *bottom ash*, and classified from 2 decaying series Uranium-Radium (^{238}U series) and Thorium (^{232}Th series)

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and ^{40}K. After burned, coal volume will decreased, so the natural radionuclides contains in *fly ash* and *bottom ash* will increased and contribute the increasing of environment radiation exposure and marine pollution in Muria coastal waters.

Validation for simulation model and observed shown in **Fig. 11** until **Fig. 18.**

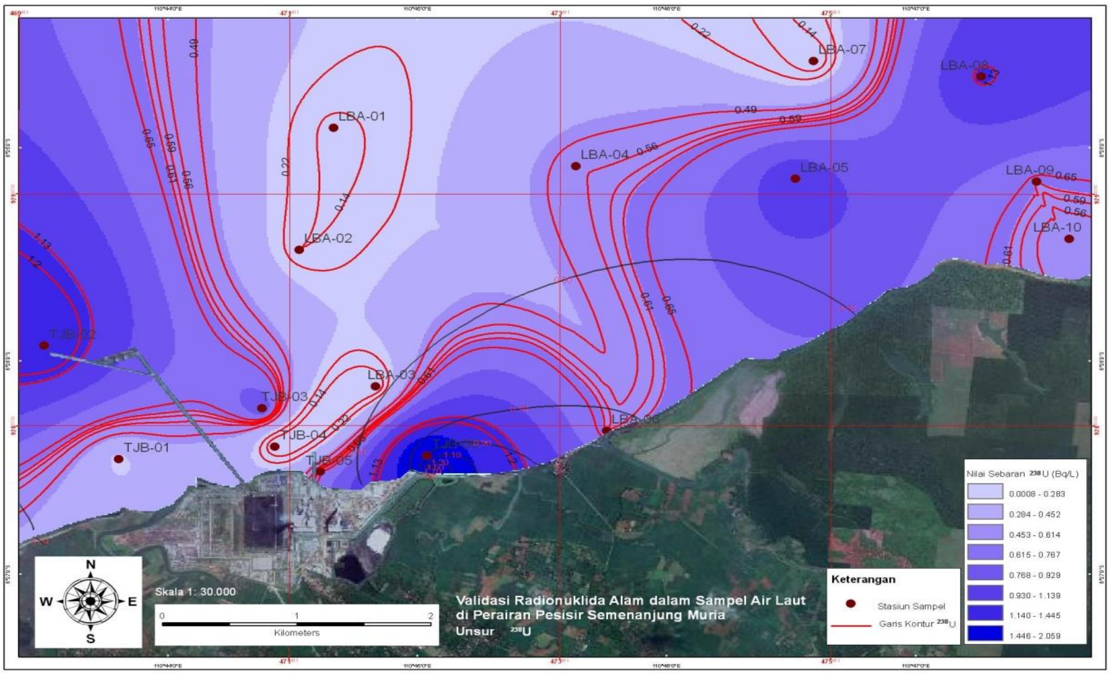

Fig. 11. Validation for Iso-Activity of ^{238}U in Sea Water Sample

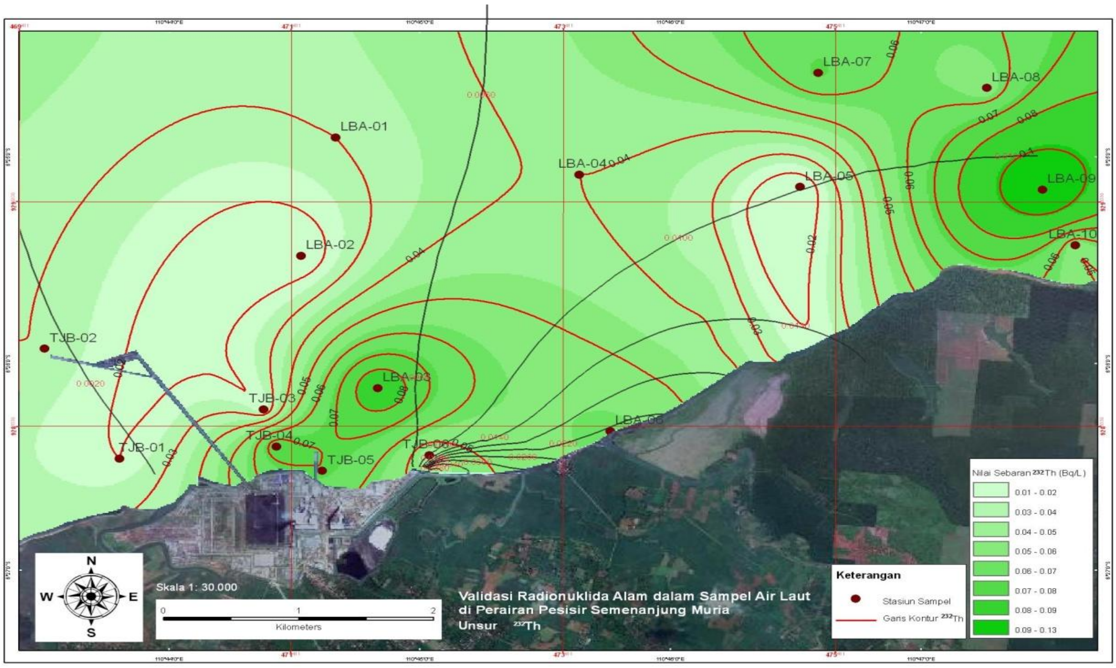

Fig. 12. Validation for Iso-Activity of ^{232}Th in Sea Water Sample

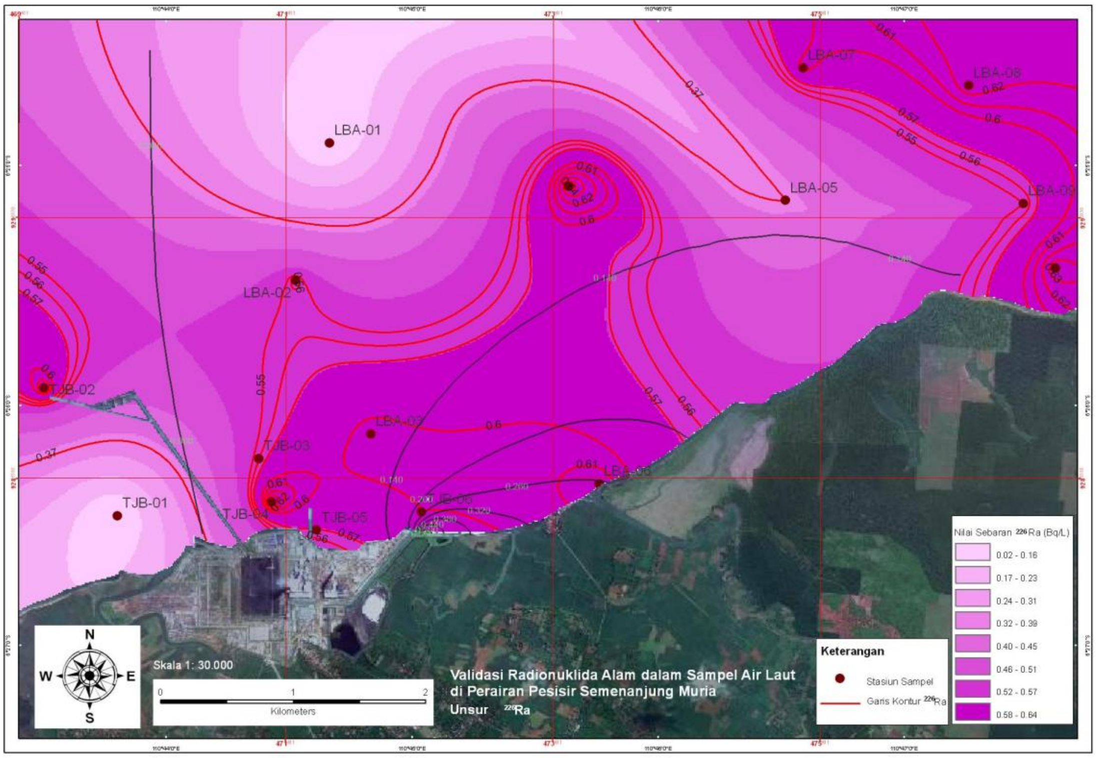

Fig. 13. Validation for Iso-Activity of ^{226}Ra in Sea Water Sample

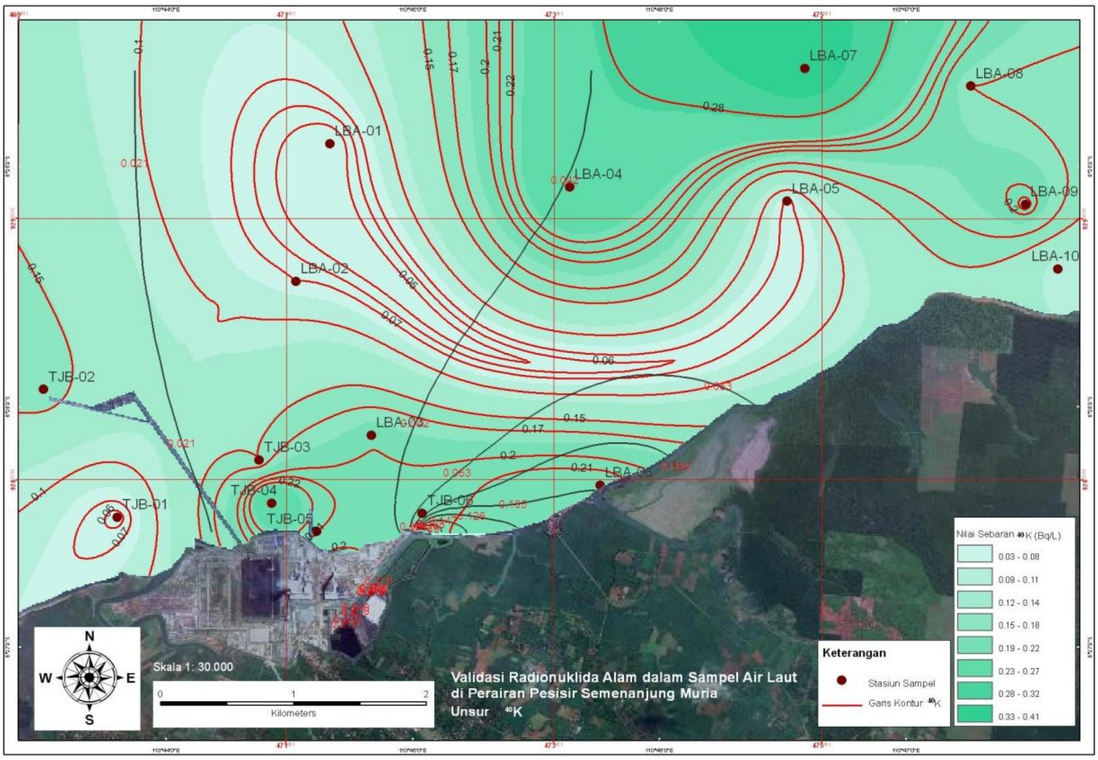

Fig. 14. Validation for Iso-Activity of ^{40}K in Sea Water Sample

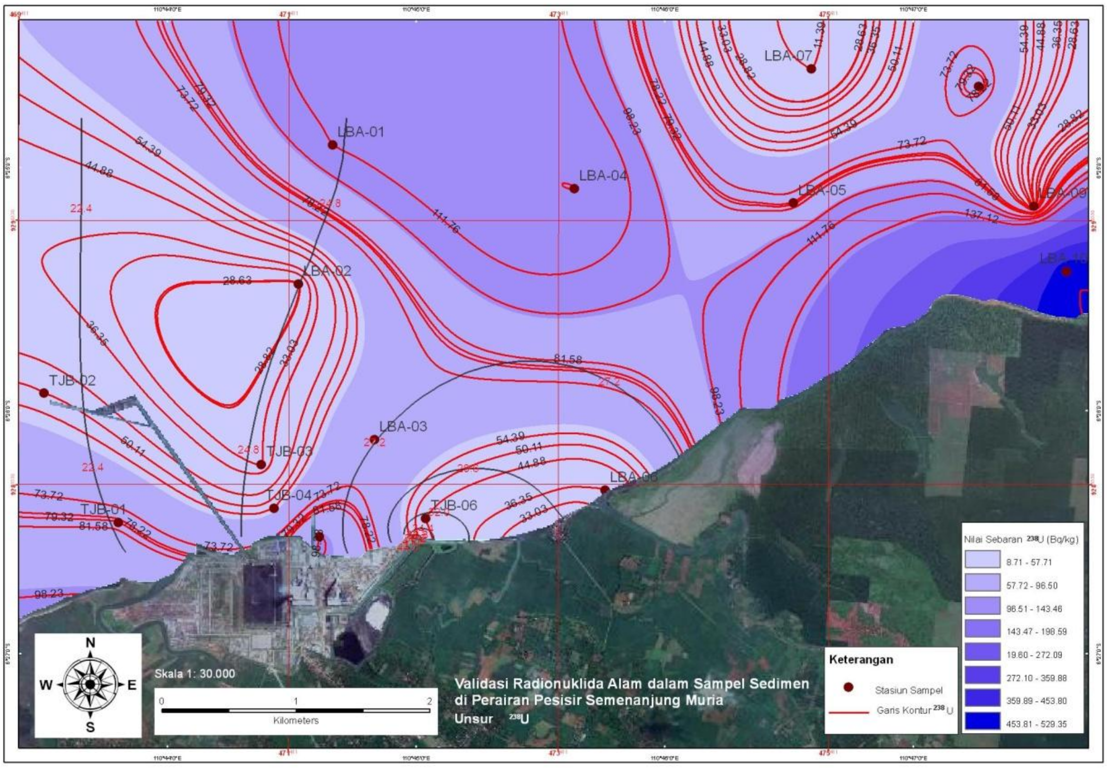

Fig. 15. Validation for Iso-Activity of ^{238}U in Sediment Sample

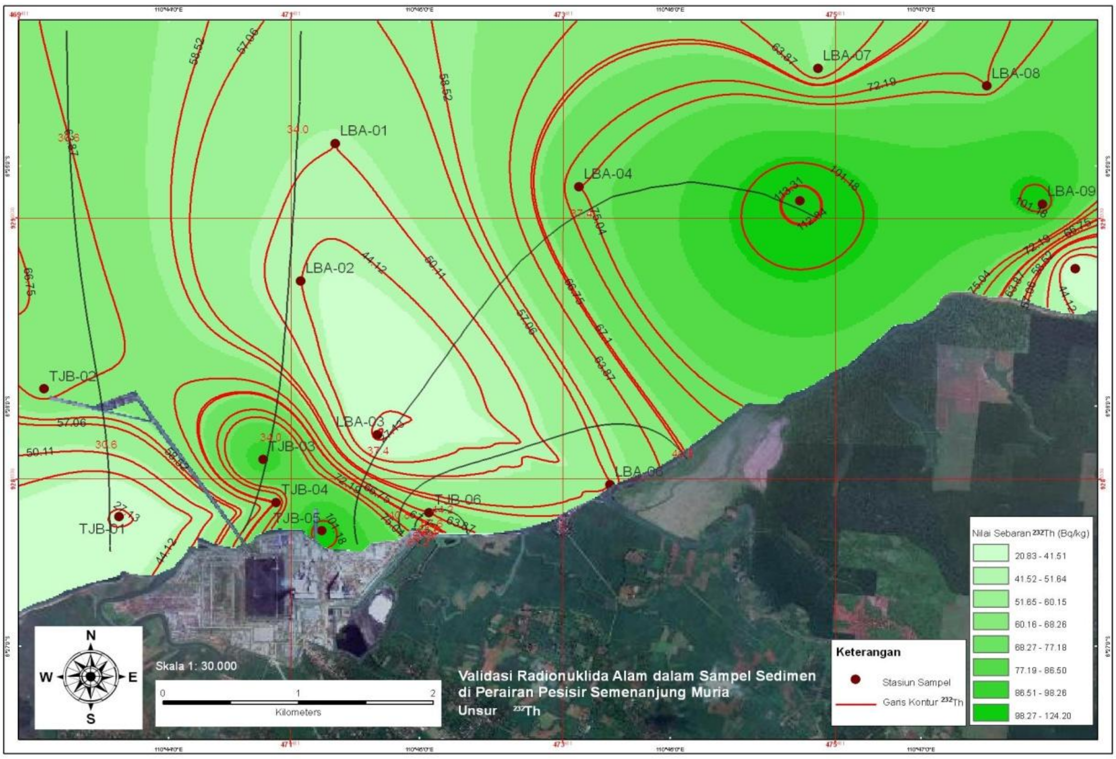

Fig. 16. Validation for Iso-Activity of ^{232}Th in Sediment Sample

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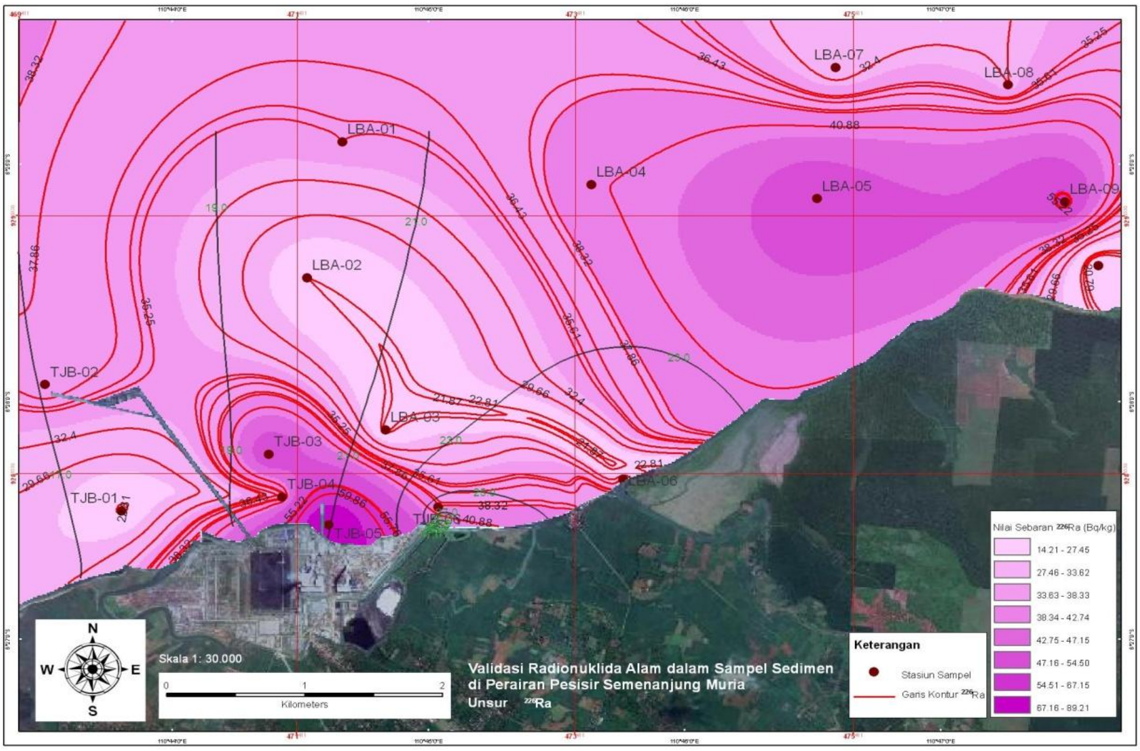

Fig. 17. Validation for Iso-Activity of ^{226}Ra in Sediment Sample

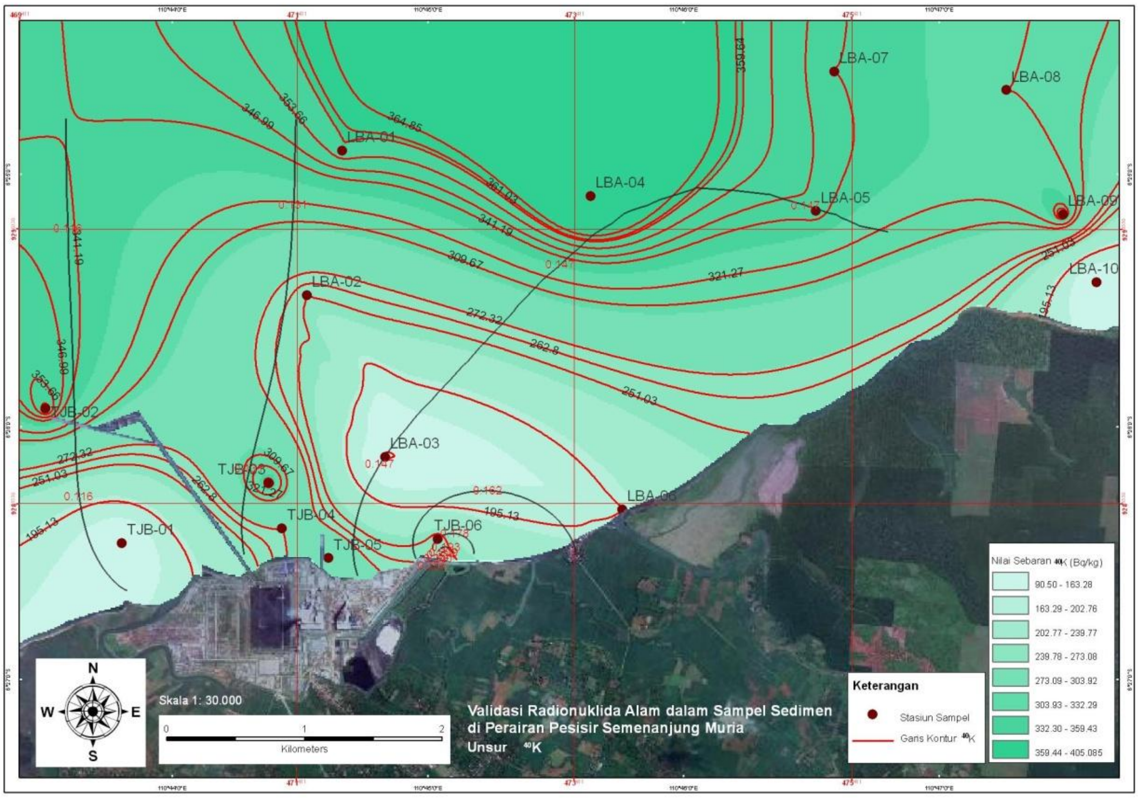

Fig. 18. Validation for Iso-Activity of ^{40}K in Sediment Sample

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CONCLUSION

The natural radionuclide of ^{238}U, ^{232}Th, ^{226}Ra and ^{40}K in sea water and bed-load sediment can be identified and measured from the radionuclides counting and spectrometri-γ analysis. Also it can be justified that natural radionuclides of ^{238}U, ^{232}Th, ^{226}Ra, ^{40}K was leachated from fly ash and bottom ash of coal burned Tanjungjati CPP to sea water. The natural radionuclides from leaching process will suspend in suspended-load and deposited in bed-load sediment.

The hydrodynamics model of unsteady 2-dimensional fexible grid system by using CD Oceanography software for current plotting, ArcView GIS 3.3 software for bathimetric contouring and SMS 8.1 software for modeling of natural radionuclides dispersion in coastal waters can be applied for dispersion analysis of ^{238}U, ^{232}Th, ^{226}Ra, ^{40}K in sea water and sediment samples from Muria coastal waters.

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