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ASSESSMENT OF SURFACE RADIATION DOSE RATE AND ACCUMULATION OF CADMIUM, NICKEL AND PLUMBUM IN THE ROADSIDE SOILS ALONG BANDAR PUSAT JENGKA TO CHENOR TOLL, PAHANG

(Penilaian Kadar Dos Radiasi Permukaan dan Penumpukan Kadmium, Nikel dan Plumbum di dalam Tanah Pinggir Jalan Sepanjang Bandar Pusat Jengka Ke Tol Chenor, Pahang)

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Abstract

Studies have been carried out along the road of Bandar Pusat Jengka to Tol Chenor, Pahang to determine surface radiation dose rate and heavy metals concentration in the roadside soils as a result of transportation of mining wastes. The in situ surface radiation dose and concentration of Cd, Ni and Pb were studied and compared with the results obtained from an area with no transportation of mining wastes. The outcomes of this research were compared with the tolerable radiation dose and maximum allowable metal concentration in soils recommended by regulators. Result shows that, radiation dose at the surface was in a range of 0.146 μSv hr^{-1} to 0.467 μSv hr^{-1} with a mean value of 0.227 μSv hr^{-1} while radiation dose at 1 m above the ground was in a range of 0.101 μSv hr^{-1} to 0.322 μSv hr^{-1} with an average value of 0.151 μSv/hr. Accumulation of Cd, Ni and Pb in soil samples at study areas yield an average concentration of 1.90±0.54 mg kg^{-1}, 61.45±14.71 mg kg^{-1} and 104.41±8.62 mg kg^{-1} respectively. The result indicates elevated doses of radiation which is above the recommended value. Meanwhile, the distribution of heavy metals in the roadside soils at the sampling areas noted a lower level of metal concentration than their background limit. The findings of this preliminary study indicates the importance of radiological and heavy metals studies on the side of preventing further dispersion and distribution of these toxicants in the environment besides monitoring and protecting the ecosystem balance for present and future's generation.

Keywords: surface radiation, heavy metals

Abstrak

Kajian penentuan kadar dos radiasi permukaan dan kepekatan logam berat dalam tanah pinggir jalan akibat pengangkutan sisa perlombongan telah dibuat sepanjang jalan dari Bandar Pusat Jengka ke Tol Chenor, Pahang. Kadar dos radiasi permukaan in situ dan kepekatan Cd, Ni and Pb dibandingkan dengan kawasan yang tiada pengangkutan sisa perlombongan. Hasil kajian dipadankan dengan dos radiasi dan kepekatan maksimum logam dalam tanah yang dibenarkan mengikut yang disyorkan dalam peraturan. Keputusan menunjukkan dos radiasi di permukaan adalah dalam julat 0.146 μSv hr^{-1} hingga 0.467 μSv hr^{-1} dengan nilai min 0.227 μSv hr^{-1} sementara dos radiasi permukaan 1 m daripada tanah ialah dalam julat 0.101 μSv hr^{-1} hingga 0.322 μSv hr^{-1} dengan nilai purata 0.151 μSvhr^{-1}. Penumpukan Cd, Ni dan Pb dalam sampel tanah di kawasan kajian menunjukkan kepekatan purata 1.90±0.54 mgkg^{-1}, 61.45±14.71 mg kg^{-1} dan 104.41±8.62 mg kg^{-1} masing-masing. Keputusan ini menunjukkan peningkatan dos radiasi yang melebihi nilai yang dibenarkan. Walau bagaimanapun, taburan logam berat dalam tanah pinggir

jalan kawasan persampelan mencatatkan kepekatan logam yang lebih rendah berbanding had sekelilingnya. Hasil daripada penyelidikan permulaan ini menunjukkan kepentingan kajian radiologi dan logam berat bagi tujuan mencegah penyebaran lanjutan dan taburan toksid ini di samping memantau dan memelihara keseimbangan ekosistem untuk generasi hari ini dan akan datang.

Kata kunci: radiasi permukaan, logam berat

Introduction

Soil is a specific and dynamic component of natural resources for the needs of human life. Soil contamination resulted when hazardous substances or chemical either solid or liquid are mixed together with the locality soil in that particular area. On the other hand, soil contamination occurs as the concentration of a harmful substances exceed or higher than background concentration. [1,2] demonstrated that, intensive human economics activities and industrial development led to environmental pollution as a result of increasing in production and emission of industrial waste, wastewater, various radioactive substance and chemical or pesticides used in agriculture. Yet, the most considerable problem related to soil pollution is heavy metals and radioactive waste. Heavy metal pollution was visualized as undesirable changes in the physical, chemical and biological characteristics of soil, water and air in which later will pose a significant threat to human, animals and vegetation. Some of the metals, for instance, Manganese (Mn), Zinc (Zn), Copper (Cu), and Nickel (Ni) are essential and beneficial for human, microorganism, plant and animal in minute amount but excessive intake of these metal can have strong toxic effect on human and other living organisms.

An infamous hazard event due to heavy metals was the fatal Itai-itai disease in Japan caused by cadmium toxicity which claimed many lives. The cadmium due to mining in Toyama Prefecture polluted the Jinzu River which was the main source for drinking water apart from paddy planting. The disease that causes renal damage [3] was officially recognized in 1968 as the first disease induced by environmental pollution in Japan after legal proceedings. Nickel on the other hand has been identified as a toxin that severely damages reproductive health which could lead to infertility and miscarriage as well as interfering with the nervous system [4]. Perhaps, many of the elder citizens have actually experienced the era where lead is the main constituent in the water plumbing system. These had brought about a very serious illness related to nervous system. The lead poisoning can cause severe mental and physical impairment. According to Roger [5], among the common source of lead is soil polluted by car exhaust or chipping house paint especially those near to a highway. The consumer of leaded petrol fortunately had deliberately turned to unleaded fuel, under government policy to limit emissions of lead, and hence reduces the exposure of the population to the metal.

Acknowledged by Li et al [6], soil heavy materials contamination has become a serious and widespread woe in many parts of the world due to the rapid social and economic development over the past several decades. Heavy metals contamination may occur naturally in soil as a result of additional contributions derived from human activities such as agriculture, urbanization, industrialization and mining. Among these, mining has been considered as one of the most significant sources of environmental contaminations by radioactive waste and heavy metals. In addition, natural radioactivity in soil comes from the earth crust which derived essentially from the natural Potassium-40 (^{40}K), and the progenies of Uranium-238 (^{238}U) and the Thorium-232 (^{232}Th) decay series such as radium, radon, actinium, protactinium, lead and polonium. These progenies are the primary source of terrestrial gamma radiation received by human and lead to radiation hazard to human beings [7]. In processing industry, only part of mineral is used and the rest of it is rejected radioactive residues which are known as waste. As in many industrial processes, the unusable and unwanted waste product such as the residues may turn out to be hazardous. Therefore, mining wastes including those radioactive and heavy metal are in great concern since it will affect the environment as well as the health of human beings due to the short term and long term exposure. Any effective control strategy requires knowledge of the sources of environmental exposure and an understanding of the pathways of this metal in the environment.

The main goal of this research is to determine the radiation surface dose rate and to study the accumulation of heavy metals namely cadmium, nickel and lead in the roadside soils along Bandar Pusat Jengka to Tol Chenor daily access road as well as verifying the possible resources of pollution.

Materials and Methods

The research was conducted in a distance of 26 km along major roadside area of Bandar Pusat Jengka to Tol Chenor lying in the latitude of 3°45.094'N to 3°32.396'N and longitudes of 102°32.487'E to 102°29.960'E. These locations are frequented by the trucks carrying mining wastes suggested due to the good road condition and closer to their dispensation point. The control soil sample was taken at Jengka 13 (3°45.600'N, 102°32.968'E) on the ground that during daylight, no activity related to the transportation of mining waste are sighted. This location with the same natural soil profile is also far from the federal main road. The sampling areas of this study are shown in Figure 1.

Figure 1**.** The map of Bandar Pusat Jengka to Tol Chenor and Jengka 13 showing the location of sampling including control. The inset boxes are the study areas and the dots represent the sampling points.

In order to do a consistent mapping, the samples were taken every 3 kilometres. Ten sampling locations were chosen with each site was 1 meter from the roadside. For every point, five soil samples were collected by digging in 10 cm depth using hand auger (Figure 2). The control sample was collected at the roadside of Jengka 13. This method improved the representative homogeneity of sample from every sampling point [8]. The position of each sampling points were determined using the Global Positioning System (GPS). Surface radiation dose rate was measured in situ at the surface and at about one meter height from the surface of selected points. The measurements were then conducted using the portable radiation survey meter Ludlum Model 2241. In the laboratory, all the contaminants such as roots, leaves, sticks and non-soil debris were removed manually from the soil samples. The samples were then oven dried at a temperature of 100°C for two days to remove the moisture content (MC). Samples were crushed into a fine powder by using grinder and filtered using 250 μm sieve to get a homogenous sample [9]. About 0.500 g of soil sample was weighed and transferred into a digestion vessel to which 10 mL of nitric acid (HNO_3) was added. The vessel was swirled gently so that all material came in contact with HNO_3. 1 mL of hydrogen peroxide (H_2O_2) was then added to each sample solution. After all the bubbling action ended, 2 mL of hydrochloric acid (HCl) was added to the sample solution. The digestion vessels were then laden on the turntable and placed in the microwave oven (Milestone Ethos One acid microwave digestion). Samples were digested for about one hour and half at 540 W of power setting. The carousel was placed in the fume hood and the vessel units are allowed to cool at room temperature for 5 minutes once the heating cycle and digestion process was completed. The sample solutions were then filtered through the Whatman (No.42) filter and diluted into 100 mL volumetric flasks with Deionized Water Type II (18.3 MΩ/cm resistivity). The vessels were rinsed several times to ensure that

the residue solution from the digestion vessel was completely dissolved and quantitatively transferred to the funnels. The filtrate was made up to volume. After thorough mixing, a portion of solution was poured into polyethylene bottles for analysis by Atomic Absorption Spectrophotometry (AAS). All the process of mixing and filtrate were carried out in a fume hood [10].

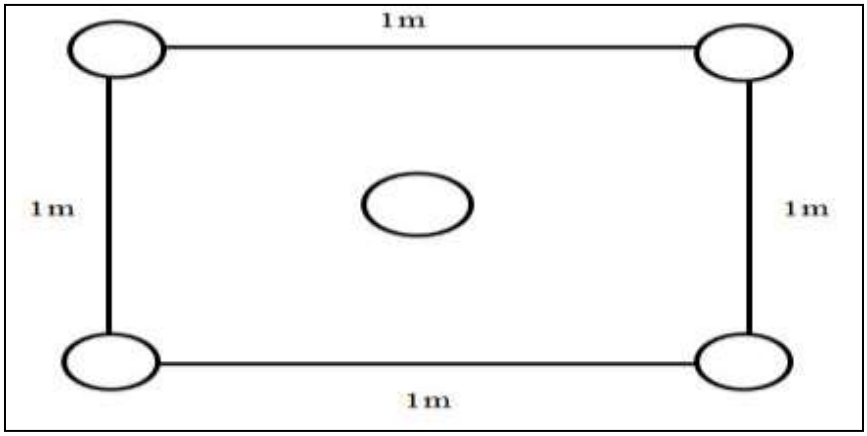

Figure 2. Illustration of sampling technique

Results and Discussion

Table 1 provides the position coordinates of the soil samples taken along the roadside of Bandar Pusat Jengka to Tol Chenor which started at Kilometer 2 from Sultan Ahmad Shah Mosque at Bandar Pusat Jengka until Kilometer 24. Sampling point number 10 as a control sample is located at Jengka 13. From the geological point of view, Jengka 13 represents an area with similar characteristic with respect to age, soil condition and petrochemical. Jengka 13 is markedly different since this area does not have any transportation of mining waste activity.

Table 1. Location of sampling stations

Location	Distance from Bandar Jengka	Latitude (N) ($\pm 0.0005'$)	Longitude (E) ($\pm 0.0005'$)
1	0	3°45.094’	102°32.487’
2	3	3°43.623’	102°32.433’
3	6	3°42.319’	102°32.514’
4	9	3°40.825’	102°32.580’
5	12	3°37.241’	102°32.164’
6	15	3°36.740’	102°32.110’
7	18	3°35.345’	102°31.252’
8	21	3°33.014’	102°31.596’
9	24	3°32.396’	102°29.960’
10	Jengka 13 (Control)	3°45.600’	102°32.968’

Surface Radiation Dose Rate in Soil Samples

The results of the radiation surface doses measured at the surface and in air at 1m above the ground investigated at various sampling points along Bandar Pusat Jengka to Tol Chenor are presented in Table 2.

Table 2. Radiation doses on surface and at 1m from the surface at sampling points

Location	Surface (µSv/hr) (± 0.0005)	1m above surface (µSv/hr) (± 0.0005)
1	0.188	0.136
2	0.191	0.128
3	0.230	0.165
4	0.235	0.118
5	0.179	0.101
6	0.222	0.131
7	0.183	0.131
8	0.146	0.130
9	0.467	0.322
10	0.061	0.050
*Mean	0.227	0.151
*SD	0.089	0.062

The results revealed that the measured surface dose rate is higher at the surface compared to that 1 m above ground. As mentioned earlier, there is no transportation of mining waste activity in the control area, Location 10 whereas the transport frequented other locations are visibly sighted at an average of 20 trucks per day. Thus, it is suggested that the radiation may be attributed mainly from the sources in the soil which may be due to the fallout or deposition of mining waste during migration. Table 2 shows that the radiation doses at the sampling points varied in a range of 0.146-0.467 µSv/hr with a mean value of 0.227 µSv/hr and 0.101-0.322 µSv/hr with an average value of 0.151 µSv/hr at the surface and 1m from the surface respectively. The highest dose rate was found at location 9 which is 0.467µSv/hr while the lowest dose rate was measured at location 10 which is 0.061µSv/hr. The soil sample at point 9 was collected at the area where the truck for transportation of mining waste routinely stopped by for meal breaks. As a result, some of the mining waste may be leaked out and deposited on the soil. Even though, the amount of deposited waste is in minute amount but frequent transportation of hazardous waste at the same route may lead to greater or explosive radiological effect in the future.

This observation again enhances the belief that the radiation at 1m above the ground is mainly derived from the soil. Moreover, the fact that heavy metals are not easily mobilised, made it impossible to be moved from the soil by wind or rain. Saat et al. [11] reported that the general observation regarding radiation doses at 1m are always lower than on the ground surface can be explained by the fact that beta radiation from the soil has been absorbed by air and unable to be detected by the meter. Therefore, this suggested that the prominent source of soil contamination by radionuclides is due to the fallout of mining waste during migration or transportation.

Distribution of Heavy Metals in Soil Samples

Table 3 shows the heavy metals concentration of cadmium, nickel and lead accumulated in the roadside soil samples. The respective Anthropogenic Factor (AF), calculated relative on the control location (Location 10) are also shown.

Table 3. Illustration of concentration of Cd, Ni and Pb and their respective Anthropogenic Factor (AF) in the soil samples

Location	Cd (mg/kg)	AF	Ni (mg/kg)	AF	Pb (mg/kg)	AF
1	2.5±0.25	8.33	68.8±2.86	1.74	115.0±1.23	1.53
2	2.1±0.25	7.00	49.0±0.89	1.24	103.9±3.52	1.38
3	1.3±0.09	4.33	48.7±2.45	1.23	101.3±2.94	1.35
4	2.3±0.09	7.67	57.2±0.71	1.45	109.2±2.78	1.45
5	1.4±0.28	4.67	53.5±0.89	1.35	91.3±2.75	1.21
6	1.7±0.09	5.67	45.7±3.11	1.16	92.6±3.39	1.23
7	1.3±0.09	4.33	60.7±1.97	1.54	104.7±1.96	1.39
8	1.6±0.16	5.33	76.0±2.54	1.92	102.8±1.14	1.37
9	2.9±0.09	9.67	93.6±3.86	2.37	118.9±6.87	1.58
10	0.3±0.19	-	39.5±1.51	-	75.3±4.47	-
*Mean	1.90±0.54	6.33	61.45±14.71	1.56	104.41±8.62	1.39

Table 3 points out that the highest concentration of Cd, Ni and Pb were found in the soil sample taken at location 9 which are 2.9 ± 0.09 mg/kg, 93.6 ± 3.86 mg/kg and 118.9 ± 6.87 mg/kg respectively and while the lowest concentration of Cd, Ni and Pb were measured at location 10 which 0.3 ± 0.19 mg/kg, 39.5 ± 1.51 mg/kg and 75.3 ± 4.47 mg/kg respectively. The significant difference in these measurements at both locations may be due to the fallout and leakage of mining waste during transportation. Furthermore, most of the sampling points which is frequently used by truck for transportation of mining waste shows higher concentration of Cd, Ni and Pb compared to location 10. Table 3 shows that, the concentration of metals in all soil samples collected at the study area gives lower levels of Cd, Ni and Pb than the maximum acceptable concentration of Pb 400 mg/kg, Ni 100 mg/kg and Cd 3 mg/kg in soil [2]. However, the concentration of Cd in sampling point 9 is almost nearing the maximum allowable concentration in soil.

Anthropogenic Factor (AF) for each element based on the concentration relative to the control concentration of the element calculated using a method described by Adamu and Nganje [12] is also presented. Based on the single factor analysis an AF < 1indicates no pollution, a value of between 1 and 2 indicates potential pollution, while between 2 and 3 represents slightly polluted, and if AF > 3 polluted by the respective element is concluded [13]. In the table, Cd from all locations showed the value of AF > 3 indicating the sampling points were polluted by Cd. The main sources of Cd in air of the area could be attributed to incineration of solid waste, fossil fuel burning and application of phosphate fertilizer [14,15]. As for Ni, except for location 9, all other locations fall into potential pollution category. For location 9, the the AF = 2.37 indicated slight pollution. For Pb, based on the AF, all the sampling points showed potential pollution. Based on AF, the degree of anthropogenic pollution by the elements in soil follows decreasing order of Cd>Ni>Pb. This shows that Cd is most enriched in comparison to Ni and Pb.

Lead enrichment in roadside soils due to the vehicular emissions may lead to soil contamination by heavy metal. According to Mmolawa et al. [16], the origin source of pollutants characterized by Pb and Ni are vehicular emissions while the other common heavy metals contamination are caused by lithogenic occurrences of human activities. Since the samples were taken from the roadside, thus there are possibilities that the measured concentration levels of metals may be due to the vehicular emission. On the other hand, it is important to note that the results of this preliminary study are comparable to the results of earlier study at several mines worldwide. Documented by Li et al. [6], the average level of Cd, Ni and Pb concentration related to mining activities show a relative correlation to this present study in which, the mean concentration of Cd in South Korea and Iran mines are

1.99 mg/kg and 1.49 mg/kg respectively. For Ni concentration, China (urban road dust) recorded a mean value of 56.75 mg/kg and average concentration of Pb at same mines in South Korea is 111.1 mg/kg. Therefore, the outcomes of this research proposed that transportation of mining waste along the road of study area is the potential contributor of soil contamination by heavy metal.

Conclusion

The surface radiation dose rate measured in the soil samples collected along Bandar Pusat Jengka to Tol Chenor shows an average value of 0.210 µSv/hr. The in situ measurement displayed that the dose is higher than the suggested dose limit of 0.079–0.190 µSv/hr recommended by UNSCEAR [17] In terms of heavy metal contamination, highest distribution of Cd, Ni and Pb at the study area were found at the concentration of 2.9±0.09 mg/kg, 93.6±3.86 mg/kg and 118.9±6.87 mg/kg respectively. Comparing this result to the maximum allowable background trace elements in soil provided by regulators, the concentration of Cd, Ni and Pb, however, shows a lower distribution of associated metals in soil samples. Based on the results, nonetheless, this study proposed the presence of radionuclides activity and Cd, Ni and Pb in the study area as a result of leakage and deposition of mining waste on the soil surface during transportation since radiation doses and concentration of metals are higher than those at Jengka 13 (control sample).

The results of this present study can be used as a baseline to further elucidate the soil contamination by distribution and accumulation of radionuclides due to the transportation of mining waste along the study area. Using the data of this present study, any changes in environmental with respect to the distribution and dispersion of radionuclides caused by mining activity can be observe and determine. Note that, once the radioactive materials exposed to the surface, deposition might introduce the real problem especially related to health effect. In the process of radioactive decay, the radioisotope is continuously emitting the radiation until they achieves their nuclei stability since radioisotope decaying process can last from hundreds to thousands of years. Timely, the radiation emitted can poses an adverse threat to human beings, animal, food crops and ecosystems balance as well as other living organisms. Therefore, it seems necessary to study and investigate the radionuclides concentration with respect to ^{238}U and ^{232}Th which will provide an indicator of determining external radiation risk index related to health risk. The outcomes can also be used to provide a reference data for monitoring possible radioactivity contamination in future.

References

1. Matchavariani, L. and Kalandadze, B. (2012). Pollution of Soils by Heavy Metals from Irrigation near Mining Region of Georgia. *Forum Geografic* XI(2): 127–136.
2. Tahar, K. and Keltoum, B. (2011). Effects of Heavy Metals Pollution in Soil and Plant in the Industrial Area, West Algeria. *J. Korean. Chem. Soc.* 55(6): 1018–1023.
3. Bernard, A. (2008) Cadmium and its adverse effect on human health; *Indian J Med Res* 128: 557-564.
4. Forgacs Z, Massányi P, Lukac N, Somosy Z. (2012). Reproductive toxicology of nickel–review. *J Environ Sci Health A Tox Hazard Subst Environ Eng* 47(9): 1249-60.
5. Roger, H. (2013). Lead Pollution: Causes and control; Chapman & Hall, New York, 55-57.
6. Li, Z., Ma, Z., Van der Kuijp, T. J., Yuan, Z., and Huang, L. (2014). A review of soil heavy metal pollution from mines in China: pollution and health risk assessment. *Sci. Total Environ.* 468-469: 843–853.
7. Yii, M.W., Jaffary, N.A.M. and Ahmad, Z. (2011). Radiation hazard from natural radioactivity in the sediment of the East Coast Peninsular Malaysia Exclusive Economic Zone (EEZ), *J. Anal. Sci* 15(2): 202–212.
8. Hamzah, Z. Alias, M., Saat, A., Wood, A. K., Tajuddin, Z. (2007). Surface Radiation Dose Measurement and Mapping of Jengka 15 Oil Palm Land, *Malay. J. Anal. Sci* 11: 237-245.
9. Alias, M., Hamzah, Z., Saat, A., Omar, M. and Wood, A. K. (2008). An assessment of absorbed dose and radiation hazard index from natural radioactivity, *J. Anal. Sci*. 12(1): 195–204.
10. Kalra, Y. P., Maynard, D. G., and Radford, F. G. (1989). Microwave digestion of tree foliage of multi-elements analysis. *J. For. Res.* 19: 981–985.
11. Saat, A., Hamzah, Z., Wood, A. K., and Thomas, F. F. (2013). Assessment of radiation and heavy metals concentration in soils along trails of Gunung Ledang, Johor; Prosiding Seminar XPDC Gunung Ledang; 1 – 3 September 2013, Muar, Johor.
12. Adamu C. I. & Nganje, T. N. (2010) Heavy metal contamination of surface soil in relationship to land use pattern: A case study of Benue State, Nigeria; Materials Science and Application 1: 127-134.

13. Deng Hong-gui, Gu Teng-feng, Li Ming-hui, Deng Xu (2012) Comprehensive assessment model on heavy metal pollution in soil; *International Journal of Electrochemical Science* 7: 5286-5296.
14. Dragovic S., Mihailovic N. (2009) Analysis of mosses and topsoil for detecting sources of heavy metal pollution: multivariate and enrichment factor analysis; *Environ Monit Assess*157: 383-390.
15. Bajpai R., Mishra G.K., Upreti D.K., Nayaka S. (2011) Determination of atmospheric heavy metals using two lichen species in Katni and Rewa cities, India. *Journal of Environmental Biology*, 32: 195-199.
16. Mmolawa, K. B., Likuku, A. S., and Gaboutloeloe, G. K. (2011). Assessment of heavy metal pollution in soils along major roadside areas in Botswana, Afr. *J. Environ. Sci. Technol.* 5: 186–196.
17. UNSCEAR, United Nations Scientific Committee on the Effects of Atomic Radiation (2000). Exposure from Natural Sources. Report to General Assembly. Annex B, New York.