

Heavy Metals Content and Pollution in Tin Tailings from Singkep Island, Riau, Indonesia

(Kandungan Logam Berat dan Pencemaran di Lombong Timah dari Pulau Singkep, Riau, Indonesia)

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ABSTRACT

Instead of its economic impact, tin mining activities cause environmental problems. The tin occurrence which is related to tin-bearing alteration on S-type Muncung Granite and its mining history in Singkep Island describes in this study. This work assessed the heavy metals concentration in six tin tailings and two soils from Singkep using inductively coupled plasma - mass spectrometry in correlation to environmental hazard. Both primary and placer mining methods applied in tin excavations on the studied area. Concentration tendency of heavy metals in the six studied tailings samples is generally $Cr > Pb > As > Ba \approx V$. The identical heavy metals trend represented by the three samples from Bukit Tumang might reflect a similar primary tailings character. On the other hand, anthropogenic activities and different surrounding rocks caused the dissimilarity of heavy metals pattern on placer wastes. Higher environmental problems are detected on the primary wastes than the placer one, especially arsenic and chrome. Severe arsenic pollution degree is also indicated in the soil sample just outside the mining location at Betong Village. Conservation and amelioration programs are useful in improving the environmental condition on studied locations.

Keywords: Heavy metals; pollution index; S-type granite; tin tailings

ABSTRAK

Aktiviti perlombongan bijih timah lebih menyumbang kepada masalah alam sekitar berbanding memberi kesan kepada ekonomi. Berlakunya timah yang berkaitan dengan bijih timah-galas pindaan ke atas S-jenis Muncung granit dan sejarahnya perlombongan di Pulau Singkep diterangkan dalam kajian ini. Kajian ini menilai kepekatan logam berat dalam enam lombong bijih timah dan dua tanah dari Singkep menggunakan plasma induktif berganding - spektrometri jisim yang berkolerasi kepada membahayakan alam sekitar. Kaedah perlombongan utama dan plaser digunakan dalam penggalian bijih timah di kawasan yang dikaji. Kecenderungan kepekatan logam berat dalam enam sampel lombong yang dikaji secara umumnya adalah $Cr > Pb > As > Ba \approx V$. Trend logam berat yang diwakili oleh tiga sampel dari Bukit Tumang mungkin mencerminkan watak utama lombong yang sama. Selain itu, aktiviti antropogenik dan batu sekeliling yang berbeza telah menyebabkan ketidakerupaan pola logam berat pada sisa plaser. Masalah persekitaran yang tinggi terutamanya pencemaran arsenik dan kromium dikesan di dalam sisa utama daripada plaser satu. Tahap pencemaran arsenik yang tinggi juga dikesan di dalam sampel tanah di luar lokasi perlombongan di Kampung Betong. Program pemuliharaan dan ameliorasi penting dalam memperbaiki keadaan alam sekitar di lokasi yang telah dikaji.

Kata kunci: Granit jenis-s; indeks pencemaran; logam berat; lombong timah

INTRODUCTION

Indonesia has a long history of Tin (Sn) mining dating back to more than two centuries ago. It started in the Dutch colonial period, then continued by PT Timah as the National Enterprise and the private company PT Koba Tin in several islands: Karimun, Kundur, Singkep, Bangka, and Belitung. Indonesian Tin production was ever known as the top three in the world because of mining activities in these Islands and gave extensive impact on the national economy. The metal resource is related to the Southeast Asian Tin Belt which expands from China towards Thailand, Peninsular Malaysia to Indonesia (Asmarhansyah et al. 2017). Tin mineralizations are associated to peraluminous tin-bearing S-type granites

which typically spread in the main granite province of Southeast Asia (Cobbing 2005; Irzon 2015).

Mining projects demand land area for house and utility construction instead of mining area itself. The economy of Dabo, the capital of Singkep Island, ever grew well because of the tin industry. The reduction of market demand and the lowering of metal price resulted in the closure of tin mining activity in 1993. A large area of wastelands, ex-tin mining ponds and tailing dumps are some negative impacts of tin exploitations. Tin tailings is a waste portion of target mineral produced after tin separation and concentration from the rock mass and the ore. Fine grain tailing generates dusty pollution in a dry environment while sulfide one yields acidic runoff (Alshaeabi et al. 2009). Previous

studies showed that the disposed of tailings caused health and environmental problems that are hazardous to the local resident (Alshaebi et al. 2009; Ashraf et al. 2012, 2011; Asmarhansyah et al. 2017; Domingo and David 2014; Nurtjahya et al. 2017; Selinus 2004; Wapwera et al. 2015; Zulfahmi et al. 2012). Besides, as tin mineralization correlated with S-type granite which also contains REE, the tailing is proved to contain a valuable number of REE (Irzon et al. 2016).

Heavy metals are not only naturally contained in soils and rocks but also derived from anthropogenic sources, namely, industries, agricultural fertilizers, pesticides, waste incineration, road traffic and combustion of fossil fuels (Baharim et al. 2016; Onder et al. 2007). Chrome (Cr), lead (Pb), arsenic (As), barium (Ba) and Vanadium (V) are some heavy elements which are byproducts of tin mining and concentrated in tin tailing (Alshaebi et al. 2009; Daniel et al. 2014). Most of the heavy metals are brought into the human body via food and water in the diet, the air we breathe, tailing leakage and occupational exposure (Ray & Ray 2009; Teng et al. 2011). Body immunotoxicity, nasal irritation, skin illness, intractable vomiting, and gastrointestinal disease are health effects of lead, chrome, vanadium, barium, and arsenic pollutions, respectively (Cappuyns & Slabbinck 2012; Llugany et al. 2000; Ray & Ray 2009).

Environmental effects and suitability of former-tin mining lands for agricultural and resident areas portrayed in previous investigations (Alshaebi et al. 2009; Asmarhansyah et al. 2017; Domingo & David 2014; Nurtjahya et al. 2017; Usikalu et al. 2011; Wapwera et al. 2015). This study describes the geological view of tin occurrence and its mining history in Singkep Island. Although Pb, Cu, and Cd concentration on the freshwater trailer of former tin excavation on Singkep Island has been previously previewed (Eddiwan 2017), no investigation established about heavy metals on tailings and the impact to environmental issue. Pollution index of five heavy metals (Pb, As, Bi, Mn, and Cr) concentration in tin tailings at Singkep Island also evaluated in assessing the environmental impacts. The information could be useful for future appropriate land usage and development of the former mining sites.

MATERIALS AND METHODS

LOCAL GEOLOGY

The studied location is an island in Riau Islands Province at the east of Sumatra and northwest of Bangka Island. Actual coordinates of sampling sites were attained using Global Positioning System to reconfirm the location for future study in the Singkep Island. Regional geology of the studied location is a part of Geological Map of The Dabo Quadrangle, Sumatra. The Permo-Carbon Persing Metamorphic Complex and the Bukitduabelas Quartzite are the oldest rock units in Singkep Island. The metamorphic complex occupies the most area of the island whilst the

quartzite outcrops situated in the north. Two plutons are located in the Island: the S-type Triassic Muncung Granit which was divided into two facies geochemically on the northeast and the I-type Jurassic Tanjungbuku Granit at the southwest (Irzon 2015). The northern coast of Singkep is dominated by Quaternary Swamp Deposit whilst the Alluvium is located at both north and south shores the island. The climate of Singkep Island is tropical with dry and rainy seasons. The average temperature in the part of Lingga Regency is 28°C and the range of yearly rainfall is 1,000 - 3,000 mm (Irzon et al. 2016).

SAMPLE COLLECTION

Dumping sites are easily identified on the abundance of quartz sand on the surface because tailings are composed of quartz, Ca-K- and Na-feldspar, clay minerals, Ti-oxides and carbonates (Courtin-Nomade et al. 2015). The greater composition of quartz sand generally decreases metal composition in the soil. The samples of this study were taken from the surface to not more than 50 cm below the surface without any pre-concentration procedure. Two tailing stations are situated in Bukit Tumang at the area of Muncung Granite. The region was a mining site of PT Timah and nowadays is being re-mined by local residents using simple washing and filtering methods. TRE 51 H and TRE 51 I were attained in Bakong Village at the north of Bukit Tumang whilst TRE 59 was taken from the south of the district. TRE 51 H is dominantly composed of reddish clay with a small amount of quartz sand while TRE 51 I is new tailing from recent re-mined activity but duskier than the previous. No re-mining activity detected at the south of Bukit Tumang in which TRE 59 as a mixture of quartz sand and reddish clay was taken.

TRE 65 A was dominated with white-yellowish fine sand from 50 m near the three-way junction in Marok Tua. TRE 76 was attained about two km before the east shore of Singkep Island in Batu Berdaun Village. Alike at Bakong Village, local inhabitants conducted simple re-mining activities in the location. Singkep was a tin island in which more than forty mining activities were emplaced even in present Dabo city. TRE 66 was sampled not more than 1 km outside Dabo and majorly composed of reddish clay and a small quantity of quartz sand. Huge kolongs found around sampling location in Marok Tua and Batu Berdaun. Similar ex-tin mining pond might possibly was located around TRE 66 but had been buried on the city development project. Two soil samples from different sites in Singkep Island were taken for comparison. A mixture of quartz sand and pale red clay material was identified as TRE 50 which was grabbed 100 m outside a re-mining base in Bakong Village. Organic matters were detected in RGI 55 A as the topsoil cover of granite outcrop in Marok Kecil. Sampling locations in Singkep Island shown in Figure 1.

EXPERIMENTAL PROCEDURES

All samples were brought to Geology Laboratory of Indonesian Center for Geology Survey in Bandung to be

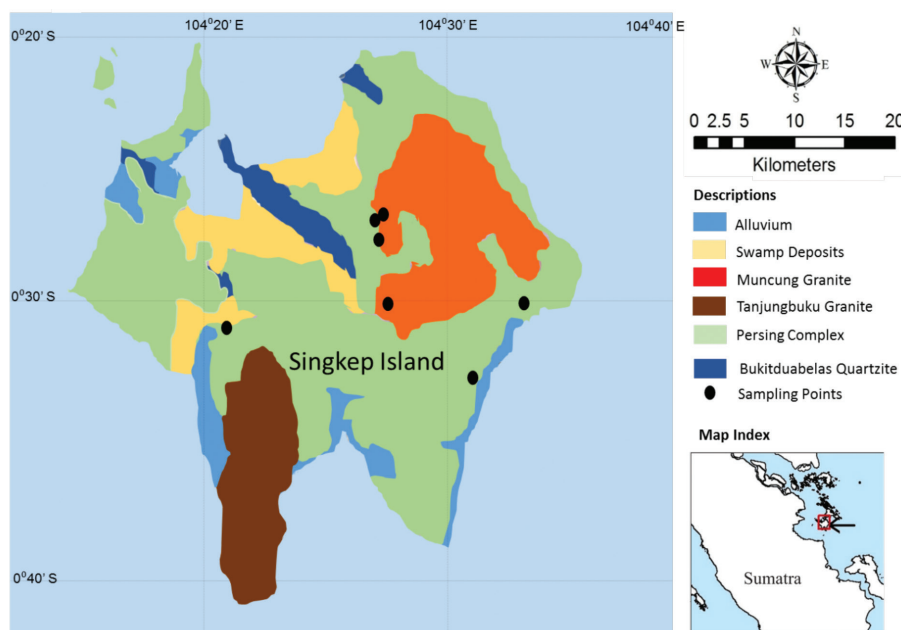


FIGURE 1. Geological map of Singkep Island and the sampling points (Sutisna et al. 1994)

analyzed using Thermo X-Series Inductively Coupled Plasma – Mass Spectrometry (ICP-MS). The equipment was applied on analyzing many environmental investigations because of its sensitivity (Ashraf et al. 2012; Courtin-Nomade et al. 2002; Garcia et al. 2009; Sakawi et al. 2013; Turunen et al. 2016;). After dried outdoor for one day in minimum, the samples were then crushed and milled to gain the fraction of <200 mesh. All laboratory equipment had been previously washed using 2% HNO₃ and rinsed with deionized water before used. Acid destruction was applied using hydrofluoric acid, perchloric acid, and nitric acid. ICP-MS was run adapting the recommended parameters of the previous study about weathered granite (Irzon et al. 2016). AGV2 and GBW 07113 were the two certified reference materials (CRM) to verify the measurement results. The recovery values of the CRMs are presented in Table 1.

RESULTS AND DISCUSSION

MEASUREMENT RESULT

The accuracy of preparation and measurement procedures were verified using two certified reference materials, namely, AGV-2 and GBW 7113. AGV-2 is andesite certified

reference material from Guano Valley whilst GBW 7113 is the Chinese rhyolite. All samples and CRMs were measured three times to establish the measurements stability on their relative standard deviations (RSD). The applied ICP-MS in this study was relatively stable with RSD <1.5%. Moreover, the measurement results are reliable on good accuracy ranging from 80.88% - 119.01% (Table 1).

Cr is the most abundant heavy metal with the content of all analyzed samples ranging from 26 to 316.58 ppm with an average of 103 ppm. Pb, Ar, Ba and V average compositions are 32.32, 27.92, 21.66 and 20.02 ppm, respectively. The new tailing from Bukit Tumang contains the most Cr (316 ppm), Pb (71 ppm) and As (68 ppm). On the other hand, the most clayey sample which taken not far from the Dabo city indicates the highest Ba (39 ppm) and V (46 ppm) concentrations. Overall, concentration tendency of heavy metals in the six studied tailing samples is Cr>Pb>As>Ba≈V. The heavy metals abundance in the studied samples shown in Table 2.

TIN MINING IN SINGKEP

More than forty mining sites of both primary and placer sources situated in Singkep Island. Tin resource in Singkep

TABLE 1. Results of analysis on certified reference materials (CRM) in comparison with certified values (ppm)

	Average analysed value				Certified CRM value		Accuracy (%)	
	AGV-2 + RSD		GBW 7113 + RSD		AGV-2	GBW 7113	AGV-2	GBW 7113
V	107	(0.7%)	4.41	1.34%	120±5	3.8	89.17	116.05
Cr	15.97	(0.46%)	5.57	1.11%	17±2	7.3	93.94	76.30
As	-	-	0.6	1.48%	n.a.	0.66	n.a.	90.91
Ba	922	1%	521	1.09%	1140±32	506	80.88	102.96
Pb	15.45	0.57%	39.63	1.35%	13±1	33.3	118.85	119.01

TABLE 2. Heavy metals content of the studied samples (in ppm)

	Primary Tailings			Placer Tailings			Soil samples	
	TRE 51 H	TRE 51 I	TRE 59	TRE 65	TRE 66	TRE 76	TRE 50	RGI 55 A
Cr	84.32	316.58	64.79	26.51	29.71	86.04	8.07	122.21
Pb	45.43	71.07	24.89	14.69	31.17	6.69	8.04	17.36
As	37.5	68.43	23.68	2.76	15.45	19.73	62.83	10.54
Ba	12.69	17.24	12.49	24.28	39.14	24.14	9.88	23.28
V	6.78	8.18	18.91	17.95	46.77	21.52	17.16	16.61

associated with Muncung Granite on the main range province of the Southeast Asian tin belt which mostly consists of S-type granite (Cobbing 2005; Irzon 2015). Tin mineralization mostly recognized in this sedimentary/supracrustal origin of granite (Chappell & White 2001; Ghani et al. 2014; Hutchison 2014; Zhao et al. 2005). Tin bearing-hydrothermal fluid filled the fractures on the Muncung Granite and became the primary source. Cassiterite which contains tin dioxide (SnO₂) is the most important tin mineral instead of sulfide stannite (Cu₂FeSnS₄), teallite (PbSnS₂) and kesterite (Cu₂(Zn, Fe)SnS₄). Kuantan-Dungun (Schwartz & Askury 1990), Belukar Semang (Sapari et al. 2016), Bangka - Belitung (Ng et al. 2017) and Sungai Isahan (Setiawan et al. 2017) are some locations with S-type granite associated tin mineralization in Sumatra and Peninsular Malaysia.

The abundant wealth of reserve, Indonesia is a world-class tin producing country. Tin mining activity in Singkep was started more than two century in the past. NV Singkep Tin Exploitatie Maatschappij (NV SITEM) was Dutch the tin mining company in the island during the colonial period. The corporation was then incorporated with Banka Tin Winning Bedrijf (BTW) and Gemeenschappelijke Mijnbouw Maatschappij Billiton (GMB) into PT Timah after the independence of Indonesia. Tin is mined both from primary and secondary/placer sources. A relatively complicated method is required for hard rock deposits tin recovery which associated with underground granitic rock. The primary source of tin in Singkep is derived from tin bearing-hydrothermal fluid filled the fractures on the S-type Muncung Granite at the northeastern part of the island. During long geologic time further since quartz- and cassiterite-rich veins mineralization, the host rock surely experienced continuous weathering. Placer deposits, either *in-situ* alluvial deposits, alluvial concentrations of cassiterite in streams and river gravels are generated due to the weathering process of the mineralized S-type granite. Placer minings are easier because the metal can be simply excavated using basic equipment and are recovered applying simple gravity separation technique (Gardiner et al. 2015; Nelson & Church 2012). Nevertheless, placer mines need massive flowing water to mobilize huge volumes of sediment resulting in downstream aggradation and environment instability (Nelson & Church 2012).

Bukit Tumang is located in west Singkep on which huge batholith of S-type granite identified. The Muncung Granite in Singkep is megascopically light grey,

holocrystalline, medium-grained, phaneritic and generally consists of quartz, K-feldspar and plagioclase (Irzon 2015). As located in Bukit Tumang, tin wastes of TRE 51 and TRE 59 were originated from primary minings. Without any catchments, the metals concentrations should be lower in the area farther from the mining site on describing their lower abundance in soil outside mining area (TRE 50) than tin tailings of TRE 51 H and TRE 59. Nevertheless, the low mobile Ar was accumulated on the upper part of the soil close to tailing location due to dust emission (Garcia et al. 2009; Turunen et al. 2016). TRE 51 I is a newly produced tailing from which minimum fraction leached out because of meteoric water and wind breeze to portray the more heavy metals abundance in the sample (Table 2).

As described previously, large water volume is required in placer mining to separate tin containing cassiterite from the sediments. TRE 65 and TRE 75 are most possibly the waste of placer mining on behalf of large waste ponds near them. On the other hand, no such large ponds situated close to TRE 51 and TRE 59. Relatively conformable heavy metals trend represented by the three samples from Bukit Tumang might be an indication of a similar source (Figure 2(a)). Tin containing minerals in TRE 65, TRE 66 and TRE 75 were transported far away from its primary resource and emplaced above different rock units, namely, the Swamp Deposit, the Alluvium and the Persing Metamorphic Complex, respectively. The Meteorological condition such as wind, rainfall and profile during the migration should surely influence the chemical abundance in the tin placer material (Onder et al. 2007). Moreover, the chemical composition of the surrounding rocks might have influenced the tin tailing composition of the three samples to illustrate the dissimilar pattern of the heavy minerals (Figure 2(b)).

POLLUTION INDEX

Besides giving economic growth, tin exploitation caused pollution which desire conservation, mitigation and protection to bring about a healthy environment (Wapwera et al. 2015). The ratio metal concentration in the studied sample against selected threshold values (C/P index) is calculated to assess significance heavy metal contamination range of the studied area (Chokor & Ekanem 2016; Hong et al. 2014). Detailed of ten contamination intervals were proposed using C/P value from <0.1 to >16 on describing very slight contamination to excessive pollution degree

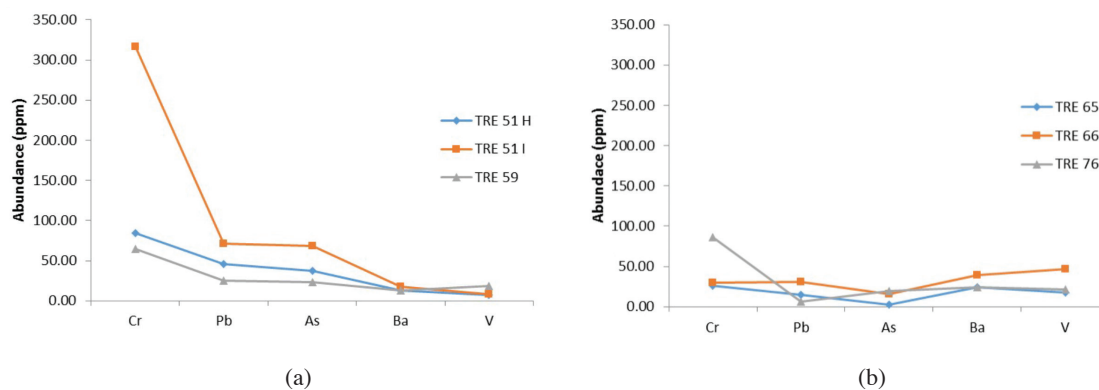


FIGURE 2. Heavy metals composition in primary tailings and placer tailings of the studied samples, a) primary tailings draw a similar trend to describe the identical source; on the other hand and b) several different factors influence the dissimilar tendency of heavy mineral on the placer tailings

(Lacatusu 2000). C/P above 1 might be harmful to soil, plant, and environment whilst the lower one is relatively secure (Hong et al. 2014). Adapting critical value from other parts of the world covering spatially significant areas is necessary because of the unavailability of local data. Five regulatory reference values (RRVs) of heavy metals in soil are compared on extending the contamination value internationally, namely, VROM, CCME, CEPA, SFT and NEPM from Netherland, Canada, China, Norway and Western Australia, respectively (Table 3). CEPA is special because it is set by the Chinese national authority not only based on the scientific view, but also on economic and political judgments (Wu et al. 2014). Canadian residential guideline is used in this study because of more the complete element threshold value and match with the purpose of the study on assessing the tailing area for a dwelling place. The C/P index of this study shown in Table 4.

The graph of C/P index on three groups of the studied sample, namely, primary tailings, placer tailings and soils

drawn in Figure 3. Tailing areas from the primary source of tin mining are moderately polluted by chrome and arsenic according to the pollution index. On the other hand, the three locations of the tailing site from tin placer mining only very severely polluted by Arsenic but indicate moderate Cr contamination. Lead concentration in primary tailing samples denotes a slight contamination degree whilst the other group with a moderate one. Barium and vanadium compositions in the studied location are not environmentally dangerous, except for TRI 66.

Heavy metals might be transported as leachate from tailing and even waste rocks to surrounding and cause pollutions on soils, surface and ground waters (Daniel et al. 2014). The transport process explains the higher contents of the six heavy metals in newly generated waste than the other tailing around Bukit Tumang. Moreover, the transmigration effect is also shown on the abundance of elements in soil outside the mining area (TRE 50) which is lower than old and new tin exploitation wastes except for

TABLE 3. Five standard limits of heavy metals in soil in ppm. 1 = VROM (2000), 2 = CCME (2007), 3 = CEPA (1995) in Wu et al. (2014), 4 = SFT (1995) and 5 = NEPM (2014)

	Netherlands ¹	Canada ²	China ³	Norway ⁴	Western Australia ⁵
Cr	230	64	200	100	100
Pb	290	140	300	50	300
As	40	12	30	20	100
Ba	625	500	-	-	-
V	-	130	-	-	-

TABLE 4. Contamination/Pollution index of studied samples based on CCME values

	Primary Tailings			Placer Tailings			Soil samples	
	TRE 51 H	TRE 51 I	TRE59	TRE 65	TRE 66	TRE 76	TRE 50	RGI 55 A
Cr	1.32	4.95	1.01	0.41	0.46	1.34	0.13	1.91
Pb	0.32	0.51	0.18	0.1	0.22	0.05	0.06	0.12
As	3.13	5.7	1.97	0.23	1.29	1.64	5.24	0.88
Ba	0.03	0.03	0.02	0.05	0.08	0.05	0.02	0.05
V	0.05	0.06	0.15	0.14	0.36	0.17	0.13	0.13

Arsenic and Vanadium. Instead of natural processes such as weathering, biological activity and volcanic emissions, As concentration in the environment is also influenced by wallpaper industry, mining activity, fossil fuels combustion and the used of arsenic pesticides (Ray & Ray 2009; Smedley & Kinniburgh 2002; Turunen et al. 2016; Wuana & Okiemen 2011). Situated nearby mining sites explain the relatively higher Ar concentration in primary tailings than the placer ones. Arsenic does not transport laterally and the element's vertical mobility is limited in soils (Tóth et al. 2016). Moreover, dust emission accumulates Ar on the upper part of the soil close to tailing location (Garcia et al. 2009; Turunen et al. 2016). During heavy metals movement from concentrated location to relatively uncontaminated one, arsenic was left in the nearby soil and accumulated in TRE 50.

Naturally, vanadium concentration in soils and sediment relies on the parent material and V-containing minerals. Moreover, anthropogenic activities such as using of pesticide, burning of fossil fuels, iron refining and dyeing release vanadium to the environment (Cappuyens & Slabbinck 2012; Teng et al. 2011; Wright & Belitz 2010). This element mainly associated with Fe(hydro)oxides, clay minerals and organic matter. Because the tailings might have been dumped since years ago, the surrounding condition of the site should also influence its heavy metals composition. TRE 50 and TRE 65 was attained close to the roads and are affected by fossil fuels combustion to reflect their vanadium abundance. TRE 66 situated 1 km outside Dabo City and 2.5 km southwest of Dabo Airport. Various anthropogenic activities might become the source of the relatively high vanadium concentration in TRE 66 than the other samples. Only slight chrome pollution is detected in RGI 55 A which reflects granite as the parent rock. However, none of these samples denote vanadium pollution problem according to C/P index <0.4 (Table 4).

Amelioration and conservation programs such as rehabilitation, regeneration, revegetation, and reforestation would improve the soil property of the selected waste

dumping locations before it is suitable for a residential place (Domingo & David 2014). Various substances have been implemented for amelioration programs with different benefits. Lime is adopted for neutralizing acidity and reducing the toxicity of metals in oxidized mine tailings while the phosphate-based material is effective for immobilizing Cd, Pd and Zn. Inorganic substances have been proven to improve the drainage of fine-grained tailings whereas organic materials should buffer the soil pH on improving the metals adsorption in mine tailing (Wang et al. 2017). The selection of suitable plant species based on acidity, salinity and alkalinity of the waste site is the most important consideration on amelioration through phytoremediation (Wu et al. 2013). The use of fertilizer with a better irrigation system is suitable for tailings on sandy environments such as in TRE 51, TRE 66 and TRE 76. On the other hand, good drainage system improvement with phytoremediation would be appropriate for clayey tailing as described in TRE 59 and TRE 65 (Ang & Ho 2002).

CONCLUSION

Tin industry in Singkep Island is related to the tin-bearing mineralization on the S-type Muncung Granite. The material was mined from both primary and secondary/placer type deposits in more than forty locations. In general, concentration tendency of heavy metals in the six studied tailing samples is Cr>Pb>As>Ba≈V. Similar heavy metals trend in the three samples from Bukit Tumang represents identical primary tailings character. On the other hand, transportation process and different surrounding rocks lead to the dissimilarity of heavy metals pattern on placer wastes. Primary tailings depict more heavy metals concentration than the placer ones. Although placer tailing cites depict lower pollution index than the primary ones, only TRE 65 station is suitable for a residential place. Severe arsenic pollution degree is detected in soil just outside of former-tin mining land at Betong Village while

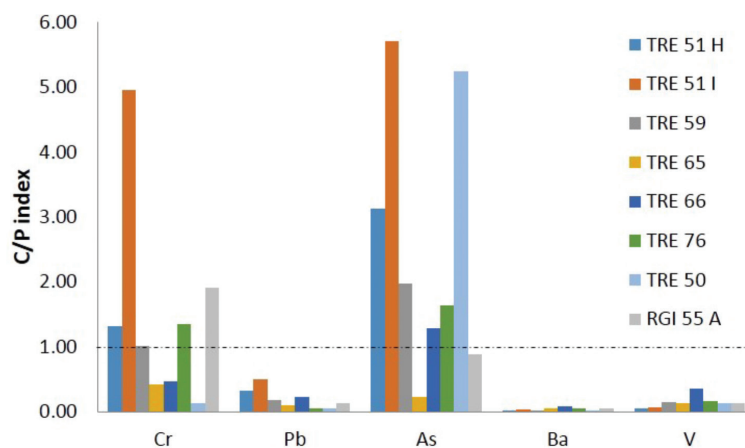


FIGURE 3. Contamination/pollution index of studied samples. Primary tailings are relatively more prone to environmental damage than the placer ones

slight chrome pollution is indicated in soil derived from weathered granite at Marok Tua. More conservation and amelioration projects should be utilized at the former-tin mining locations and surrounding area on improving its environmental condition.

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