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Risk identification of Hg and Pb in soil: a case study from Pangkep Regency, Indonesia

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Abstract

Lithogenic and anthropogenic activities can increase the concentration of heavy metals in the soil and degradation of environmental quality. Pangkajene dan kepulauan (Pangkep) regency is one of the areas in South Sulawesi Province which has severe environmental pressure. Twenty two surface soils of Pangkajene dan Kepulauan (Pangkep) regency, South Sulawesi, Indonesia were collected in order to determine the contamination status and potential ecological risks. The geo-accumulation index (I_{geo}) and potential ecological risk index (RI) were used to evaluate the contamination and risk level. The metal content in soils was determined using cold vapor atomic absorption spectrophotometer (CV-AAS) for Hg and flame atomic absorption spectrophotometer (F-AAS) for Pb. The results showed that the concentration of Hg and Pb ranged from 20.81- 223.47 mg kg^{-1} and 25.98-108.68 mg kg^{-1} respectively. Pb concentration in studied soil was below the quality standard for soil, whereas the Hg concentration was exceeded the soil quality standard. Agriculture field in Bungoro sub-district has the highest Hg concentration. The I_{geo} value showed that the soil in Pangkajene was extremely enriched with Hg and moderately enriched by Pb. The ecological risk index showed comprehensively the watershed area was at extreme risk level in need of effective monitoring and pollution control, and Hg is the important risk factor of Pangkajene watershed area.

1. Introduction

Heavy metals pollution in the soil is the severe problem worldwide which attracts public attention, especially concerning food security. Hg and Pb are the trace elements which may persist and accumulate in the soil. Its contamination does not only reduce the quality of the soil but also affects the growth of crop. Moreover, it can possibly transmit to the human body via dust and water resources by direct contact and ingestion (Chen and Zheng, 1996; Christoforidis and Stamatis, 2009; Li et al., 2016; Suryawanshi et al., 2016; Tchounwou et al., 2012; Wuana and Okieimen, 2011). Also, it can penetrate the food chain through bio-uptake by plants (Dowdy and Volk, 1983) and therefore affects animal and human health. Transfer of Hg and Pb to soil includes wet and dry deposition from atmosphere and direct discharge of pollutant (Ebinghaus et al., 1999). Waste incineration, mining, mineral processing, coal combustion, industries (electronics, pharmaceutical, caustic soda, petrochemical, cement), sewage sludges irrigation and application of fertilizer and pesticides are sources of Hg emission to the soil (Higueras et al., 2014; Li et al., 2016; Tangahu et al., 2011; Wang et al., 2020; Zhong et al., 2016). While, the Pb sources may come from leaded gasoline, mining, manufacturing, industry (paint, batteries, ammunition, metal production) and agriculture (fertilizer, manure, and pesticides) (Atafar et al., 2010; Tchounwou et al., 2012).

Hg and Pb are classified as the second and third highly toxic heavy metals, which are substances prioritize for control by U.S Agency for Toxic Substances and Disease Registry (ATSDR) in 2019 (U.S. Department of Health & Human Services, 2019). Like other heavy metals, Hg and Pb can accumulate in the soil and are bioaccumulated through food chain, which may pose a risk to ecological and human health (Masindi and Muedi, 2018; Rzymski et al., 2015; Wijayawardena et al., 2016). There is no level of exposure to Hg and Pb which is known to be without harmful effects (Bjørklund et al., 2019; Wani et al., 2015; World Health Organization (WHO), 2019). The high concentration of mercury or methylmercury primarily will affect to the neurological and renal system (Agency for Toxic Substances and Disease Registry (ATSDR), 1999; Bjørklund et al., 2019; El-Ansary et al., 2017; Fields et al., 2017; Jackson, 2018). Mercury also produces teratogenicity, especially in organic form (Methylmercury) (Bose-O'Reilly et al., 2010b; Inouye, 1989; Manduca et al., 2014; Mobarak, 2008). While the high concentration of Pb will affect to the nervous system, reproductive system and bone (Agency for Toxic Substances and Disease Registry (ATSDR), 2007; El-Ansary et al., 2017; Jackson et al., 2008; Nan et al., 2017; Nieboer et al., 2013; Slivkova et al., 2009; Sun et al., 2019; Telišman et al., 2007; Wu et al., 2012; Yang et al., 2013). Exposure to Pb will interrupt heme synthesis by inhibiting porphobilinogen synthase enzyme (Wijayawardena et al., 2016). World Health Organization (WHO), (2019) estimating long term Pb exposure accounted for 1.06 million deaths and 24.4 million years of healthy life lost worldwide in 2017. Besides, long term exposure Hg and Pb in soil have adverse effects to the microbiome, animal (earthworm), and plant (Bücker-Neto et al., 2017; Da Silva et al., 2016; Frossard et al., 2018; Kushwaha et al., 2018; Li et al., 2016; Xie et al., 2016). Therefore, it is important to monitor the concentration of Hg and Pb in order to provide information for ecological risk assessments.

Maros and Pangkep regency are well-known as the largest and the second most beautiful karst area in the world after the karst area in China (Ahmad and Hamzah, 2016). An area of land built up of limestone called a Karst area. This area has complex activities, which includes industry, domestic, fisheries, agriculture, and tourism. Furthermore, this area has severe environmental pressures from limestone mining activity (WALHI, 2018), and the impact of the mining activity is not only a threat to the availability of groundwater but also to biodiversity (WALHI, 2018). Streams and coastal area in Pangkajene watersheds are increasing-degraded by the heavy inflow of metal, nutrient, and bacterial pollutants due to rapid land use and land cover shifts. Several studies showed that there were contamination of metals in river water (Bahar et al., 2012; Bugis et al., 2012; Fadirubun et al., 2012), biota (shellfish, fish, prawn) (Daud et al., 2015; Haeriah, 2019; Haspullah et al., 2018; Sunti et al., 2012; Usman et al., 2015) and sediment (Fitriani and Dini, 2014; Fitriani et al., 2013) in Pangkep regency. Pangkajene river is the source of irrigation water to the farmland. Within a decade, there was a degraded of total agriculture area in Pangkep regency. The total reduction of the agricultural area in Minasatene sub-district 31% while in Pangkajene sub-district was 22% (Idris et al., 2015).

Agricultural land to non-agricultural land shifting may alter significantly natural biogeochemical cycle, hydrological pattern, and soil quality and thus contributing to trace elements and nutrients run-off into streams. Moreover, the geological complexity of Pangkep regency makes this area consisting of mineral and mining material. Limestone, clay/loam, silica sand/quartz, gravel, gems, alluvial gold, chert, feldspar, kaolin, basalt, slate, coal, trachea, propylite, diorite, sandstone, and radioactive mineral from weathering of rocks and soil make up the natural source of contamination. Related to the same parent rock that developed soil in Pangkep regency, previous research conducted by Rauf et al., (2020) indicated that soil Hg contamination is harmful for karst area. Hg soil contamination can infiltrate the water body, which is harmful to the population. Studies related the deposition and sources of heavy metals in soil and the potential ecological risk have seen a growing pattern in many countries such as China (Cui et al., 2018; Zhou et al., 2016), India (Raj et al., 2017; Suryawanshi et al., 2016), USA (Sridhar et al., 2020), Iran (Rostami et al., 2020), Nigeria (Ihedioha et al., 2017), Colombia (Marrugo-Negrete et al., 2017). Whereas, the heavy metals contamination in terrestrial habitat (soil) has not been assessed yet in Pangkep regency, Indonesia. Therefore, the study aims (1) to assess the concentration and spatial distribution of Hg and Pb in topsoil around Pangkajene watershed area, (2) to identify the level of Hg and Pb contamination in topsoil using I_{geo}, (3) to evaluate the potential ecological risk associated with exposure to these metals.

2. Materials and methods

2.1 Study Area

This study was carried out in an area near the Pangkajene river, Pangkajene dan Kepulauan (Pangkep) regency, South Sulawesi, Indonesia (Fig. 1). The selected areas include three sub-districts (Bungoro, Minasatene and Pangkajene), which represented the area from the upstream river to the estuary. Pangkajene is located in the north of Makassar City, South Sulawesi (110° longitude and 4°.40'–8°.00' latitude). The area of Pangkep regency is divided into two, which includes 898.29 km^2 land area and 11.464 km^2 sea. The topography of the region is dominated by plain and karst hills, with elevation ranging from 100 to 1000 metres. This area has a typical tropical monsoon climate with an average annual temperature and rainfall of 26.4°C and 2500–3000 mm/year, respectively. This region has mining resources, which includes coal, marble and cement. Also, it has developed agriculture and fisheries activities, such as rice, corn, cassava, soybean, peanuts, sweet potato, milkfish, and shrimp. In addition, Pangkep regency has a large number of mining, and industry, and those activities may be responsible for pollution, which has a significant impact on soil environmental quality in Pangkajene watershed area

2.2. Sample collection and analysis

The soil sampling was carried out in April 2020, and the sample was obtained from Pangkajene watershed area (Fig. 1). A total of 22 sampling sites with different land use, including (1) agriculture soil (13 samples), and (2) non-agriculture soil (9 samples). Seven samples (P01-P05 and P08-P09) were taken from upstream area of Pangkajene river, eight sample soil (P06-P07, P10, P14-P18) were taken from middle stream area, and other samples (P11-P13, P19-P22) were taken from the downstream area of Pangkajene river. The boundary of the sampling sites was areas which are less than 5 km from the Pangkajene river include three sub-districts (Bungoro, Minasatene and Pangkajene). Only three sub-districts selected as the sampling site because they are the largest area with largest total populations in the Pangkep regency. Pangkajene river is one of the irrigation sources water to the farmland. The method of soil samples was taken by composite with 10 individuals per sampling site. The distance between the individual samples was 200 m in a random pattern. The composite soil samples were taken using a shovel from the depth of 0–20 cm in the surface of the soil and then samples were cleared from rubbish, gravel, grasses and plant root. Therefore, 500-gram soil sample was placed into clean polyethylene bags and transported to the laboratory.

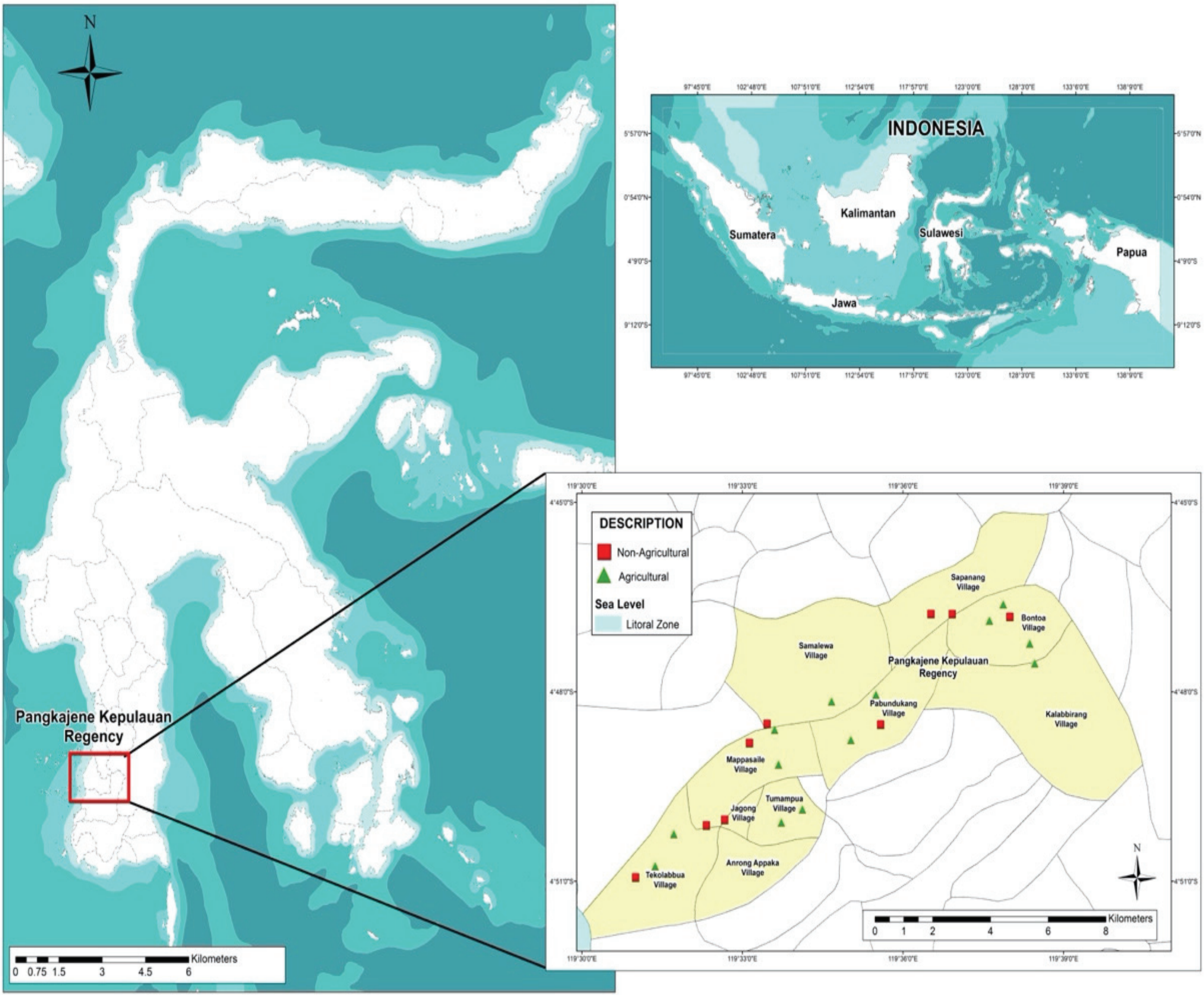


Fig. 1. Soil sampling sites in Pangkep Regency, Indonesia

All samples collected were analyzed at the center of plantation-based industry (BBIHP) laboratory in Makassar, South Sulawesi Province, Indonesia. The soil samples were dried at room temperature. The air-dried soil samples were sieved through a 2 mm Nylon sieve and homogenized. To determine Hg content 0.5 g of soil sample was weighed and placed in volumetric flask. Samples were digested by adding 2 ml of HNO_3 : $HClO_4$ (1:1) to the volumetric flask. Then added 5 ml H_2SO_4 to the mixtures and heated at 250°C for 20 minutes. Then cooled and added distilled water until the volume limit. The Hg content were analysed using cold vapour atomic absorption spectrophotometer (CV-AAS) (SHIMADZU, AA-7000 with mercury vaporizer unit (MVU-1A)) Hg analyser according to Indonesia Standard for Hg analysis (SNI 06-6992.2-2004). CV-AAS has been extensively used to measure mercury content in environmental samples (Han et al., 2006).

To measure Pb, the soil samples were air-dried, crushed, homogenized and placed in polyethylene bottle before analysis. An air-dried soil sample was weighed for 3 gram in analytical balance and placed in Erlenmeyer glass. Added and stirred the samples with 25 ml distilled water. Then, added 5 ml of HNO_3 and heated at 105°C until the mixture volume is ±10 ml. After that, cooled the mixture at room temperature. Then, added 5 ml HNO_3 and 1 ml $HClO_4$ to the mixture. Heated the mixture until it transparent and filtered the mixture using filter paper whatmann no.42 then measured using flame atomic absorption spectrophotometer (F-AAS) (PerkinElmer, PinAAcle 900H) at wavelength 217.0 nm. This method is based on the Indonesian Standard for Pb analysis (SNI 06-6992.3-2004). The QA/QC used to avoid cross-contaminant and gain accurate results. The QA/QC included standard operating procedures, and the standard material (NIST 1646a estuarine sediment), all were analysed attentively.

2.3. Assessment method

2.3.1. Geo-accumulation index

This research used geo-accumulation index (I_{geo}) to determine the heavy metal contamination in the soil by comparing current concentration and the background concentration. This method was introduced by Müller, (1969). Geo-accumulation index can be calculated using this formula:

$$I_{geo} = \log_2 \left(\frac{C_n}{1.5 B_n} \right)$$

Where, C_n was the measured concentrations of heavy metals in the soil, and B_n was the concentrations of background heavy metals in the soil. The background concentration can be used to determine the damage of toxic element in the environment (Chernova and Bezuglova, 2019). The local soil background concentrations for Pb and Hg were not available, therefore we used average crustal concentration from study conducted by Taylor (1964). The Hg and Pb background concentration are 0.08 and 12.5 mg/kg, respectively (Taylor, 1964). The constant 1.5 is often used to account for the feasible variations in the reference value as they are affected by natural phenomena and anthropogenic activities (Aiman et al., 2016). The I_{geo} classified the level of contamination as follows; practically uncontaminated ($I_{geo} \leq 0$), uncontaminated to moderately contaminated ($0 < I_{geo} \leq 1$), moderately contaminated ($1 < I_{geo} \leq 2$), moderately to heavily contaminated ($2 < I_{geo} \leq 3$), heavily contaminated ($3 < I_{geo} \leq 4$, heavily to extremely contaminated ($4 < I_{geo} \leq 5$), extremely contaminated ($I_{geo} > 5$) (Xu et al., 2017).

2.3.2. Potential ecological risk index

This index is a comprehensive method for assessing the potential ecological risk of heavy metals in the soil/sediment considering several factors, such as concentration of contaminant, toxicity level, type of pollutant and sensitivity of soil to heavy metals pollution (Hakanson, 1980). Not only can it be used to identify the potential ecological risk of heavy metals in soil to humans, animals and plants, but also to illustrate the potential ecological risk from overall level of contaminations. The formula used to compute ecological risk is as follows (Hakanson, 1980):

$$RI = \sum E_r = \sum T_r \times C_f = \sum T_r \times \frac{C_n}{B_n}$$

Where, RI referred to the total individual potential ecological risk for all heavy metals. E_r was the potential ecological risk index for single heavy metal, T_r was the toxic-response factor for specific heavy metal, C_f was contamination factor for specific heavy metal. The toxic-response factor for Hg and Pb are 40 and 5 respectively (Ahamad et al., 2020; Wang et al., 2019). The ecological risk status are classified as follows: low risk ($E_r < 40$; $RI < 150$), moderate risk ($40 \leq E_r < 80$; $150 \leq RI < 300$), considerable risk ($80 \leq E_r < 160$; $300 \leq RI < 600$), high risk ($160 \leq E_r < 320$; $600 \leq RI < 1200$), very high risk ($E_r \geq 320$; $RI \geq 1200$) (Hakanson, 1980; Wang et al., 2019).

2.4. Data analysis

Hg and Pb concentrations data were collected after analysis in the BBIHP laboratory then the statistical analysis was carried out using SPSS 24.0 version software package. Data presented was the result of descriptive analysis (mean, standard deviation, minimum and maximum value). ArcGIS 9.2 Software was used to analyze the geostatistical model. The kriging interpolation method for spatial variations of Hg and Pb concentrations was used across the whole study area.

3. Results and Discussion

3.1. Hg and Pb distribution in the soil

Table 1 showed the overall values of the Hg and Pb concentrations in the soil around Pangkajene watershed area. While Table 2 showed the Hg and Pb concentrations of the soils based on land use. The abundance of heavy metals in study area is Hg > Pb. Overall, Pb concentration in studied soils was lower than permissible concentration. While Hg concentration in soils was exceeded the permissible concentration governed by the Canadian Council of Ministers of the Environment (Table 1).

Our study was conducted in Pangkep regency, South Sulawesi Province, Indonesia. This study revealed that Hg concentration in soils was higher than the background concentration. Based on regional geological mapping of Maros-Pangkajene regency, carbonate rocks (Tonasa formation) formed in Eocene – Miocene (51 – 16 million years ago) were soil parent rocks for this region (Fatinaware et al., 2019; Ramli et al., 2009; Taslim, 2014). The soils in Pangkep regency are formed by alluvium/sediment material, limestone, breccias, lava, tuffs, conglomerates, basalt, ultra-basalt, trachyte, and mixed rock (Ramli et al., 2009). Type of soils in Pangkep regency is divided into four ordo including entisols, inceptisols, mollisols and alfisols (Putra, 2017; Ramli et al., 2009). This study location was enriched with mineral and mine resources such as limestone, clay/loam, silica sand/quartz, gravel, gems, alluvial gold, chert, feldspar, kaolin, basalt, slate, coal, propylite, diorite, marble, trachyte, sandstone, and radioactive mineral from weathering of rocks and soil make up the natural source of contamination (Dinas Pekerjaan Umum Kabupaten Pangkajene dan Kepulauan, 2017; Priyono et al., 2004). Hg may concentrated naturally high in coal and argillaceous sediments (Morgan et al., 2009; Ottesen et al., 2013). Moreover, the igneous and metamorphic rocks in Pangkep tend to accumulate Hg by natural mechanism, as explained in Ottesen et al (2013) (Ottesen et al., 2013). This happens maybe caused these soils developed by sediment material containing high lime (Ca2+) and high soil cation exchange capacity, as explained in the study conducted by Hikmatullah and Suparto (2014). The type of soil in Pangkep regency also has low nutrients (P and K) level (Ramli et al., 2009), thus fertilizer is required in this region. Furthermore, the residual soils on limestone and dolomite might be elevated Hg concentration in karst soil (Gosar et al., 2016). Carbonate-derived soils (elevated Ca and Sr) are poor drainage and leaching, thus it can be stored trace element (Hasan et al., 2020). The transporta-

Table 1
Hg and Pb concentration in studied soils (mg kg^{-1})

Element	Mean ±SD	Range	Background value*	Permissible concentration**
Hg	61.9 ±47.2	20.8–223.5	0.08	12
Pb	47.1 ±17.9	25.9–108.7	12.5	70

*Hg and Pb background were obtained from study conducted by Taylor (1964)
**Canadian Soil Quality Guidelines (CCME, 1999a, 1999b)

Table 2
Descriptive statistics of soil Hg and Pb concentrations based on land use (mg kg^{-1})

Location	Land Use Type	Heavy metals	Number of samples	Heavy metal concentration			Standard Deviation	Reference
				Mean	Min	Max		
Pangkajene Watershed	Agriculture land	Hg	13	61.9	25.8	223.5	51.7	Present study
		Pb	13	41.6	25.9	57.1	9.9	
	Non-agriculture land	Hg	9	61.8	20.81	166.5	42.5	
		Pb	9	55.8	30.84	108.7	23.7	
Yangtze river, Eastern China	Agriculture land	Hg	78	0.04	0.03	0.25	0,03	(Zhou et al., 2016)
		Pb	78	37.20	19.68	113.84	16.67	
	Non-agriculture land	Hg	53	0.07	0.01	0.21	0.06	
		Pb	53	55.97	27.01	185.59	30.77	
Yellow river watershed, China	Agriculture land	Hg	107	0.14	0.02	0.29	0.07	(Zhang et al., 2018)
		Pb		32.1	17.87	63.72	8.06	
The Hun-Taizi River Watershed, China	Agriculture land	Hg	272	0.12	0.01	0.67	0.09	(Zhang et al., 2020)
		Pb		30.18	0.32	93.58	13.51	
Xi River Watershed, China	Agriculture land	Hg	411	1.36	0.05	24.20	na	(Lian et al., 2019)
		Pb		73.62	14.50	215.00	na	
Huixian Wetland, South China	Agriculture land	Hg	6	0.14	-	-	0.14	(Huang et al., 2020)
		Pb	6	50.55	-	-	20.7	
Maros regency, Indonesia	Non-agriculture land	Hg	10	42.2	13	83	25.1	(Rauf et al., 2020)
	Agriculture land	Hg	10	77.3	29	163	43.5	
The Beke-Cave Watershed (Aggtelek-Karst, Hungary)	Non-agriculture land	Pb	98	34.64	-	-	-	(Kaszala and Bárány Kevei, 2015)
Danube River, Hungary	Non-Agriculture land	Hg	68	0.03	<0.001	0.5	0.1	(Pavlović et al., 2016)
		Pb	68	42.1	17.7	85.0	3.3	
Tropical river watershed, Terengganu, Malaysia	Non-agriculture	Hg	51	0.09	0.011	0.25	0.08	(Sultan et al., 2011)
		Pb	51	29.23	0.110	73.96	19.458	
Kinshasa watershed, Congo	Agriculture land	Hg	8	2.15	-	-	0.21	(Ngweme et al., 2020)
		Pb	8	251.05	-	-	5.02	
Sinu River Basin, Colombia	Agriculture land	Hg	83	0.177	0.050	0.410	0.080	(Marrugo-Negrete et al., 2017)
		Pb	83	0.066	0.02	0.10	0.020	

tion of trace elements in soil-derived carbonate can be through the mechanism of zinc carbonate precipitation formation with low solubility and absorption of metals via colloids, clay minerals and hydroxide colloids of iron and manganese. The alkaline environment in karst area also restrict the transportation and activation of toxic heavy metals, thus they can accumulate in the soil, plants and river sediment (Wu et al., 2020). The mechanism of metals accumulation in karst soil was quite complex where the heavy rainfall and high groundwater velocity may contribute to Hg transportation (Qiao et al., 2019; Tao et al., 2020; Zhu et al., 2019).

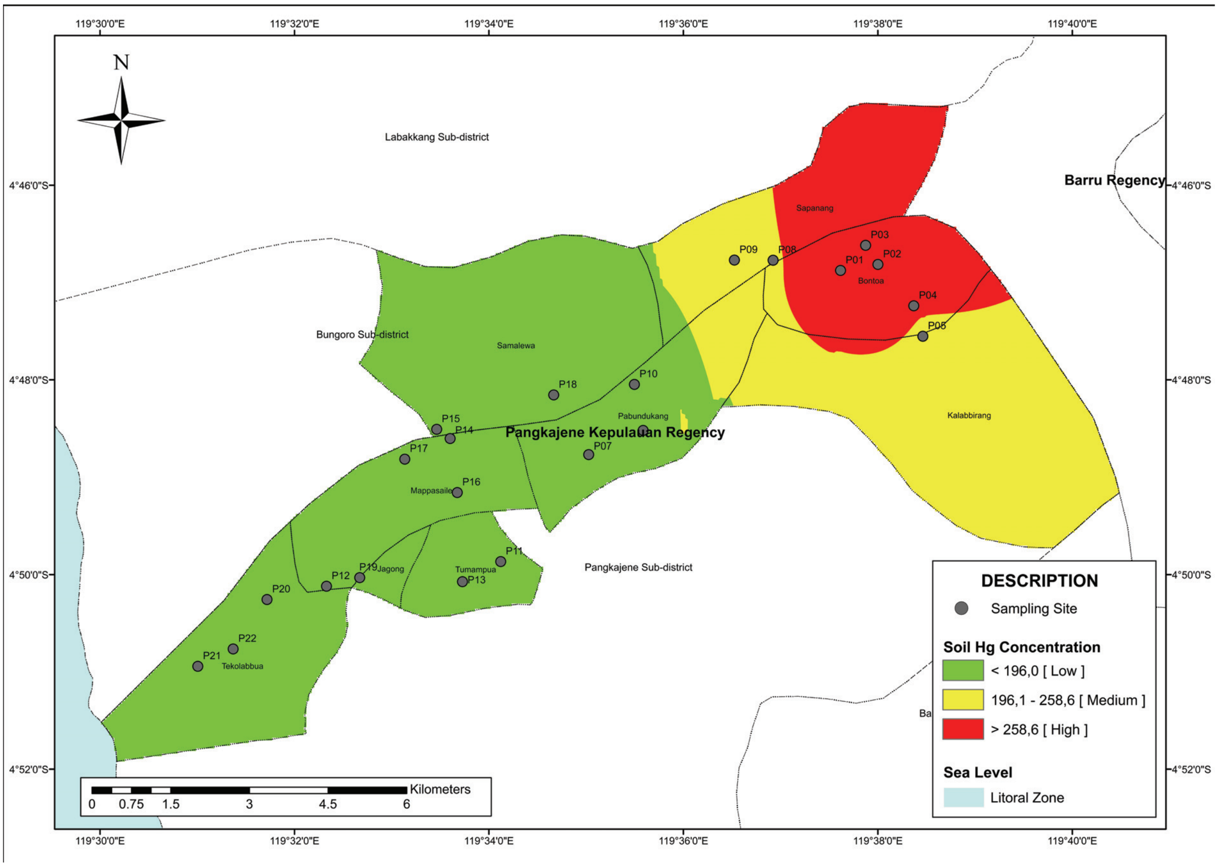
The anomalous Hg concentrations in studied area may come from gold and coal deposit in Pangkep region. There is a potential gold mining in South Sulawesi Province scattered in several districts such as Luwu, Palopo, Maros, Pangkep, Barru, Tanatoraja, Bone, Jeneponto, Selayar and Wajo (Kompas.com, 2008; Pemkab Maros, 2008). Gold mining is the second largest mining in South Sulawesi Province with total area 68271.4 Ha (17.38%) (Pemerintah Provinsi Sulawesi Selatan, 2014). Whereas the total areal of coal mining in Maros-Pangkep regency is 4639.03 Ha (Yusuf, 2014). A prior study showed that there is anomalous Hg concentration in Talawaan watershed soil, Sulawesi related to natural contamination from inactivating volcano of Mount Kablat which the soil was formed by carbonates (Filho et al., 2004). As explained by Filho et al. (2004), carbonates have abnormal amounts of Hg, As, Sb and Tl concentration and low base metals including Zn, Cu, and Pb. They are also undergone decalcified, silicified, dolomitizes process. Several studies showed that Hg contamination in Sulawesi was related to gold mining and processing (Abbas et al., 2020; Arifin et al., 2020; Bose-O'Reilly et al., 2010a; Castilhos et al., 2006; Limbong et al., 2004; Filho et al., 2004; IPEN, 2013; Limbong et al., 2003; Lusantono and Hantari, 2020; Mallongi et al., 2014; Mallongi, 2014; Mallongi et al., 2015; Mallongi and Herawaty, 2015; Nakazawa et al., 2016). A study from Tao et al. (2020) presented that there is an interaction between lithology factors and watershed to the accumulation of Hg in karst soil where local mining activities and the mechanical transport of pollutant in the watershed has a strong dependence to Hg accumulation. Since Hg has characteristic as volatile substance and soluble in water, it can be distributed long distance by air and water. Hg may transported via Hg-contaminated sediment and consequent accumulate in the areas of slow river flow, as showed in Gosar *et al.* (2016). It also tend to attach to suspended solid particles and organic matter where it can rapidly fall into river bottom, as stated in Limbong et al. (2003). The distribution of Hg into watershed influenced by several factors such as topography of watershed, climate, and ore deposit type (carbonate or sulphate) (Limbong et al., 2004; Ottesen et al., 2013). Usually, Hg-rich sediment presented as elemental and amalgam phases because the ore was from silica-carbonate deposit in the soil (Limbong et al., 2004). Relating to the atmospheric deposition, the lower Hg concentration in downstream than in upstream indicating that Hg emission from mining site to atmosphere present a relatively short residence time as Hg vapour, and it mostly precipitated on soil surrounds the source, as explained in Filho et al. (2004).

Compared to other watershed area, Hg concentration in this study was higher than the Hg concentration in watershed from other countries. We compared our results to Maros karst area, Indonesia and Beke-Cave watershed, Hungary (Table 2). Maros and Pangkep regency has the same structure of soil because they were developed by similar parent rocks. The Hg concentration of Pangkajene agriculture soil is lower than Maros agricultural soil. In contrast, Hg concentration of non-agricultural soil in Pangkajene regency is higher than in Maros regency. The high concentration of Hg in Maros agriculture soil may be influenced by the application of fertilizer and soil metal absorption factor (Rauf et al., 2020). Furthermore, the spreading of Hg may be influenced by the wind. Hg can disperse and stay in the atmosphere within 7 days (Penzias et al., 2003). Study from Hatcher and Filippelli, (2011) presented that there is a relation of wind-driven Hg deposition in soil surrounds watershed area in Indiana, USA. Wind direction and the geometry of waterways flows are related to Hg deposition in soil and sediment indicating Hg can spread to the far place from the pollution sources. Compared to the concentration of Pb in the Beke-Cave watershed, the Pangkajene watershed has a high Pb concentration than the Beke-Cave watershed. The concentration of heavy metal in soil can be influenced by the texture of soil (Kaszala and Bárány Kevei, 2015). Another study by Gosar et al. (2016) showed that karst area in Slovenia which has an extreme Hg contaminated area (19,900 mg·kg^{-1} in soil) are known influenced by chimney emission, smelting and mining processes.

Land use can significantly affect the accumulation and spatial distribution of heavy metals in the soil, which will endanger human health and the security of the ecosystem (Li et al., 2019; Smith et al., 2016; Tian et al., 2020; Wang et al., 2017). According to this study, the mean concentration of Pb in non-agricultural soil was higher than the Pb concentration in agricultural soil. While the mean of Hg concentration in agricultural soil was higher than in non-agricultural soil. The non-agricultural soil mentioned in this study was the residential soil or construction land. This result correspond with the study conducted by Tian et al., (2020), in which the Pb concentration accumulated mostly in construction land. The higher concentration of Hg in farmland can be caused by anthropogenic activities, such as mining and industry activities surrounding the farmland. In addition, the use of pesticides, fertilizers and feed additives has increased rapidly in the 1990s for aquaculture and agriculture activities by farmers in Pangkep regency (Paena et al., 2015). The Phosphate fertilizer is a type of fertilizers that is applied in rice field in Pangkep regency. Urea, SP36 and ZA are the common fertilizers used. The application of phosphate fertilizers also contributing to the deposition of heavy metals in the soil (Mendes et al., 2006). Phosphate fertilizers (P20) contains Pb and Hg as much as 9.1 and 0.024 mg.Kg -1 (Kabata-Pendias, 2011). Same study by Rauf et al. (2020a) showed that Hg content is higher in agriculture land than in non-agriculture land. Long term use of fertilizers may affect the accumulation of heavy metals especially Cr, Ni, Cu, Zn, Pb, Hg, As and Cd in the surface soil (Guo et al., 2018; Huang et al., 2020; Li et al., 2019; L. Wu et al., 2012; Zhou et al., 2015). Kabata-Pendias (2011) mentioned that the agriculture may generate higher inputs of trace metals than livestock land.

Based on the spatial distribution in the Fig. 2, it was shown that the concentration of Pb in the soil is relatively low to mod-

(a)



(b)

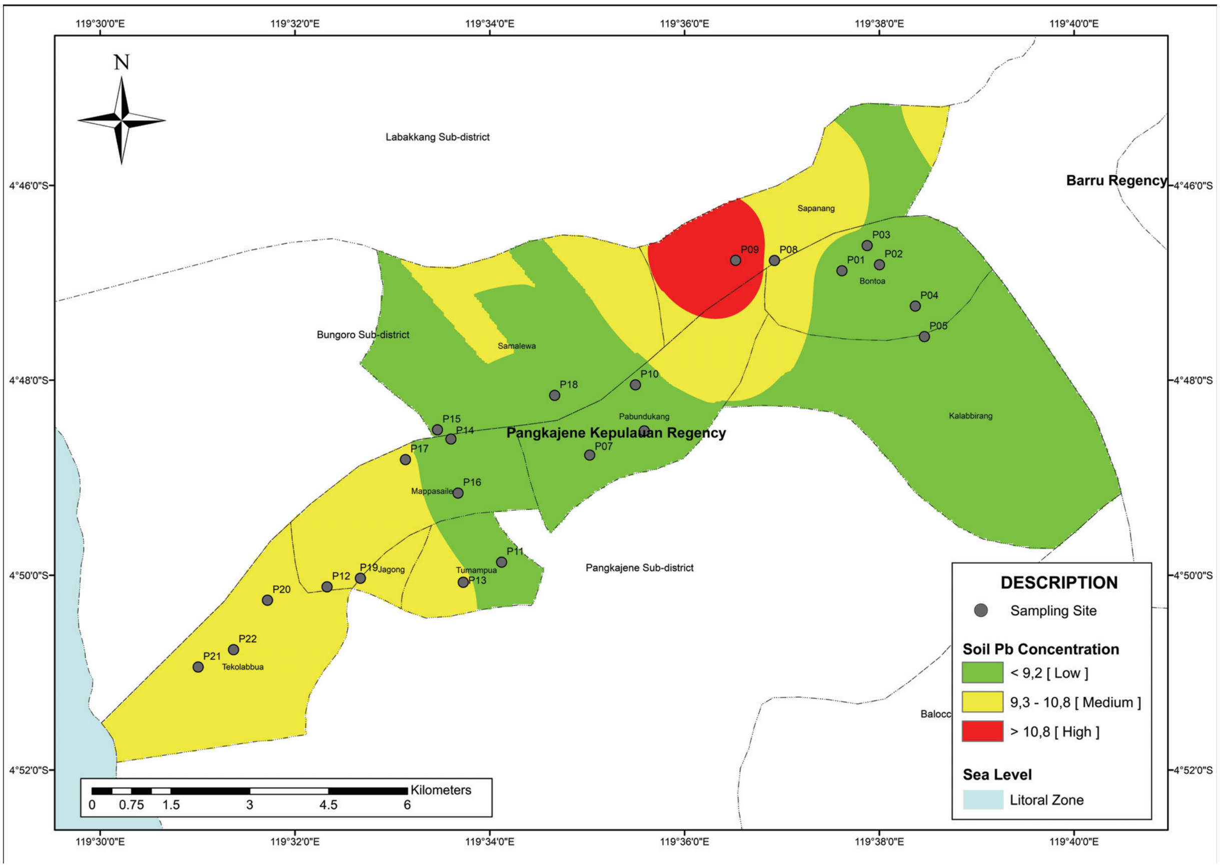


Fig. 2. Distribution of Hg(a) and Pb(b) in soil at Pangkajene Watershed, Indonesia

erate in all sampling sites. The highest concentration of Pb is only located in Bungoro subdistrict (sampling site 9). While the concentration of Hg in the soil is relatively high only at upstream and low at downstream towards the coast. In the upstream area, there is mining and industrial zone, which uses coal as fuel including cement and marble industry. The combustion of fossil fuel and cement production contributes to the deposit of Hg in the environment by 65% and 6% respectively (Kim and Zoh, 2012). A previous study showed that there is contamination of Hg in air, soil, and surface water surrounding the cement plant of Bungoro sub-district, Pangkep regency (Mallongi et al., 2020). Another study by Rauf et al., (2020), showed that there is high accumulation of Hg and Cr in soil Maros karst area, which has active cement industry and limestone mining activities. The heavy metals concentrations in limestone soil is formerly not too high. Barany-Kevei et al. (2001) also mentioned that the Pb contents in limestone and dolomite is approximately 3–10 ppm. When the concentration of Pb dan Hg exceeds the natural or background concentrations, the pollutants may come from anthropogenic sources. It can be related to air pollution through metals wet and dry deposition in the form of particles (Kabata-Pendias, 2011; Akbar et al., 2006; Miko et al., 2003).

There are several parameters related to binding process of heavy metals in the soils, such as pH, organic matter and clay contents, which are called soil buffering capacity (Barany-Kevei, 2001). High buffering capacity in the soil can accumulate toxic metals and do not allow the metals infiltrate into the soil solution and reach the limestone bedrock and karst water (Barany-Kevei, 2001). In this study, those parameters are not included and the data only indicate the Hg and Pb content in the studied soil which can be used as basic point for further research.

3.2. Contamination assessment

The I_{geo} was used to determine the degree of pollution in the soil of individual heavy metals. I_{geo} values are widely used and helpful for dividing soil into quality classes also, which can be compared to the present contamination and prior contamination (Kowalska et al., 2018). As shown in table 3. In agricultural and non-agricultural soil, the I_{geo} values showed that soil was classified as extremely enriched with Hg. When the I_{geo} values for Pb in agricultural and non-agricultural soil were classified as moderate enriched. From Fig. 3, all sampling sites exceeded the critical value of $I_{geo} > 0$, which indicated that the soil in Pangkajene watershed area are contaminated by metals (Hg and Pb) derived from anthropogenic sources. The I_{geo} was less than zero means that the soil was uncontaminated with Hg and Pb. The status of contamination varies from one another sampling site. Both agriculture (sampling site 1) and non-agriculture (sampling site 2) soil in Bungoro sub-district has the highest value of I_{geo} for Hg. I_{geo} value for Pb is located in sampling site 9, and it is the non-agricultural soil near cement industry of Bungoro sub-district. The previous study by Mallongi et al. (2020) show that Hg contamination occurs in air, water and surface soils around the cement industry in Tonasa, Bungoro Subdistrict. The highest to lowest value of Hg found in soil>air>water. The concentration of Hg in the soil and surface water is closely related to the concentration of Hg in the air.

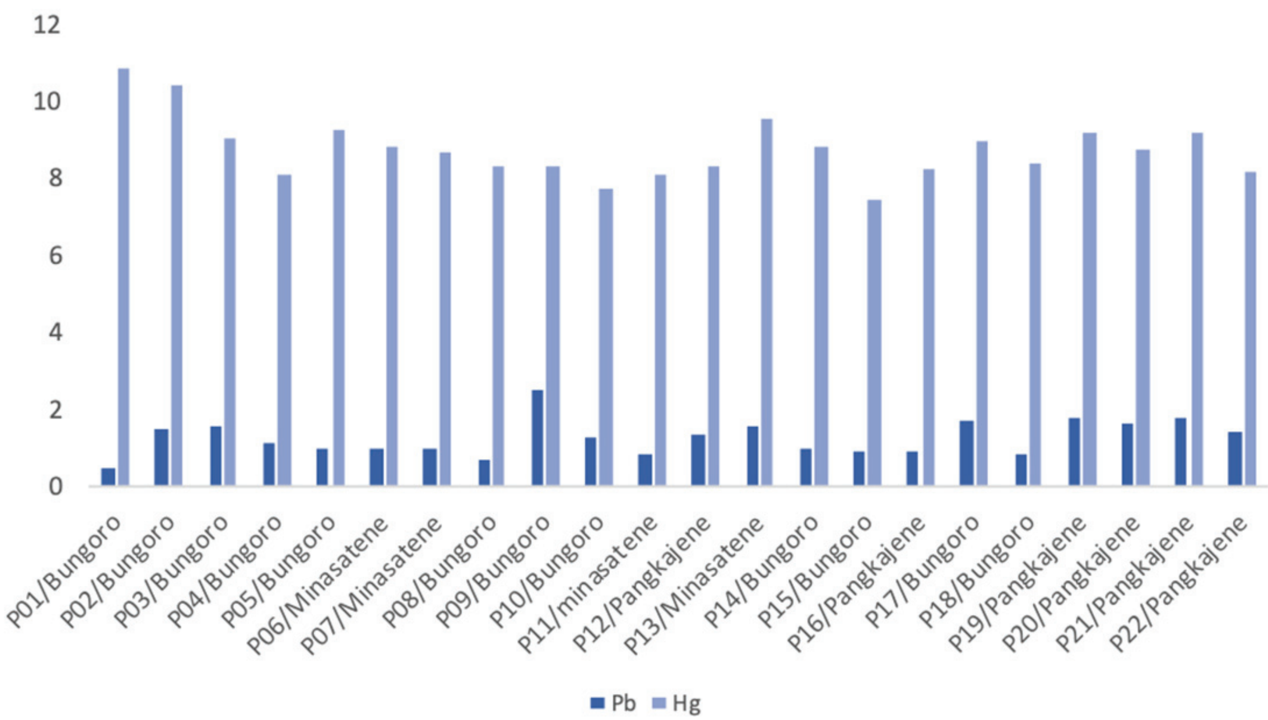


Fig. 3. Geo-accumulation index of Hg and Pb in Pangkajene watershed area

Table 3
Geo-accumulation index (I_{geo}) value of studied soil

Land use	Heavy metals	Mean	Min	Max	SD	Contamination status
Agricultural land	Hg	8.73	7.74	10.86	0.82	Extremely contaminated
	Pb	1.10	0.47	1.61	0.34	Moderately contaminated
Non-agricultural land	Hg	8.77	7.44	10.40	0.82	Extremely contaminated
	Pb	1.47	0.72	2.53	0.56	Moderately contaminated
Overall	Hg	8.75	7.44	10.86	0.80	Extremely contaminated
	Pb	1.25	0.47	2.53	0.46	Moderately contaminated

Table 4
Potential ecological risk index of soil heavy metals in studied soils

Element	Ecological risk index			Percentage of area (%)				
	Mean	Min	Max	Low risk	Moderate risk	Considerable risk	High risk	Very high risk
Hg	30928	10409	111739	-	-	-	-	100
Pb	19	10	43	95	5	-	-	-
RI	30947	10423	111749	-	-	-	-	100

This condition may be related to the large amounts of silica emissions, wind flow and the production of cement plants using silica as a raw material (Mallongi et al., 2020).

3.3. Potential ecological risk of soil

E_r was used to evaluate the potential ecological risk of a single metal in the soil. The potential ecological risk (RI) can determine the environmental risk of heavy metal contamination in the soil, as it not only reflects the influence of pollutants, but also the overall effect of pollutants (Harikumar et al., 2010; Santos-Francés et al., 2017). This index estimates showed that different heavy metal have different ecological toxicity, and therefore there is a different ecological risk to the environment (Wang et al., 2019). Moreover, it can use as the baseline information to take measures for managing the environment. According to the mean value of Hg and Pb potential ecological risk index (E_r) of each soil sample, the order of the mean value of potential ecological risk is Hg > Pb. The highest E_r value for Pb was in non-agricultural soil, while the highest value of Hg was in agricultural soil (Fig. 4). Mean value of ecological risk (E_r) Hg was 30928, it indicated that the soil in this study area has a very high ecological risk from Hg contamination. Whereas Pb mean value of ecological risk (E_r) was 19 implied that the soil of Pangkajene watershed has low ecological risk from Pb contamination. Overall, the risk status for soil in Pangkajene watershed area is extreme (RI = 30947). The highest value of RI is located in the upstream area (Fig. 5). This area is dominated by mining, industry and agriculture activities. Overall, the soil in Pangkajene area has very high risk from heavy metals and the most contributing factors are from Hg. Therefore, the monitoring and pollution control needs to be carried out in this area.

4. Conclusions

The result indicates that soil in this study area was a very high ecological risk from Hg and moderate risk from Pb. The upstream area dominated by mining, industry and agriculture activities was the area with the highest potential ecological risk score (RI). In conclusion, this study showed that Hg is the most important risk factor in Pangkajene watershed area. The results can provide baseline information for the management of Pangkajene dan Kepulauan regency's environment and will drive the attention to further research. There are some related topics

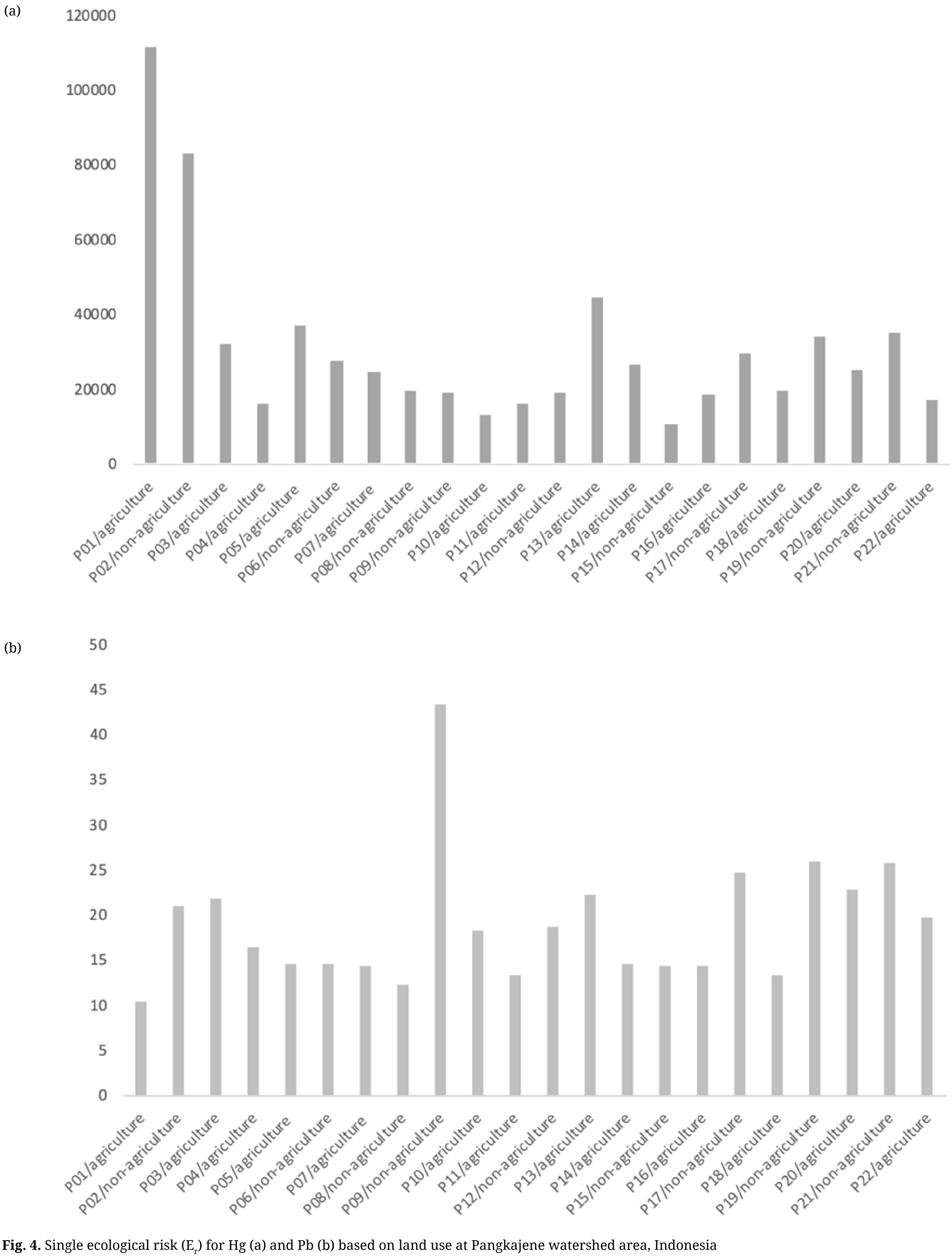


Fig. 4. Single ecological risk (E_r) for Hg (a) and Pb (b) based on land use at Pangkajene watershed area, Indonesia

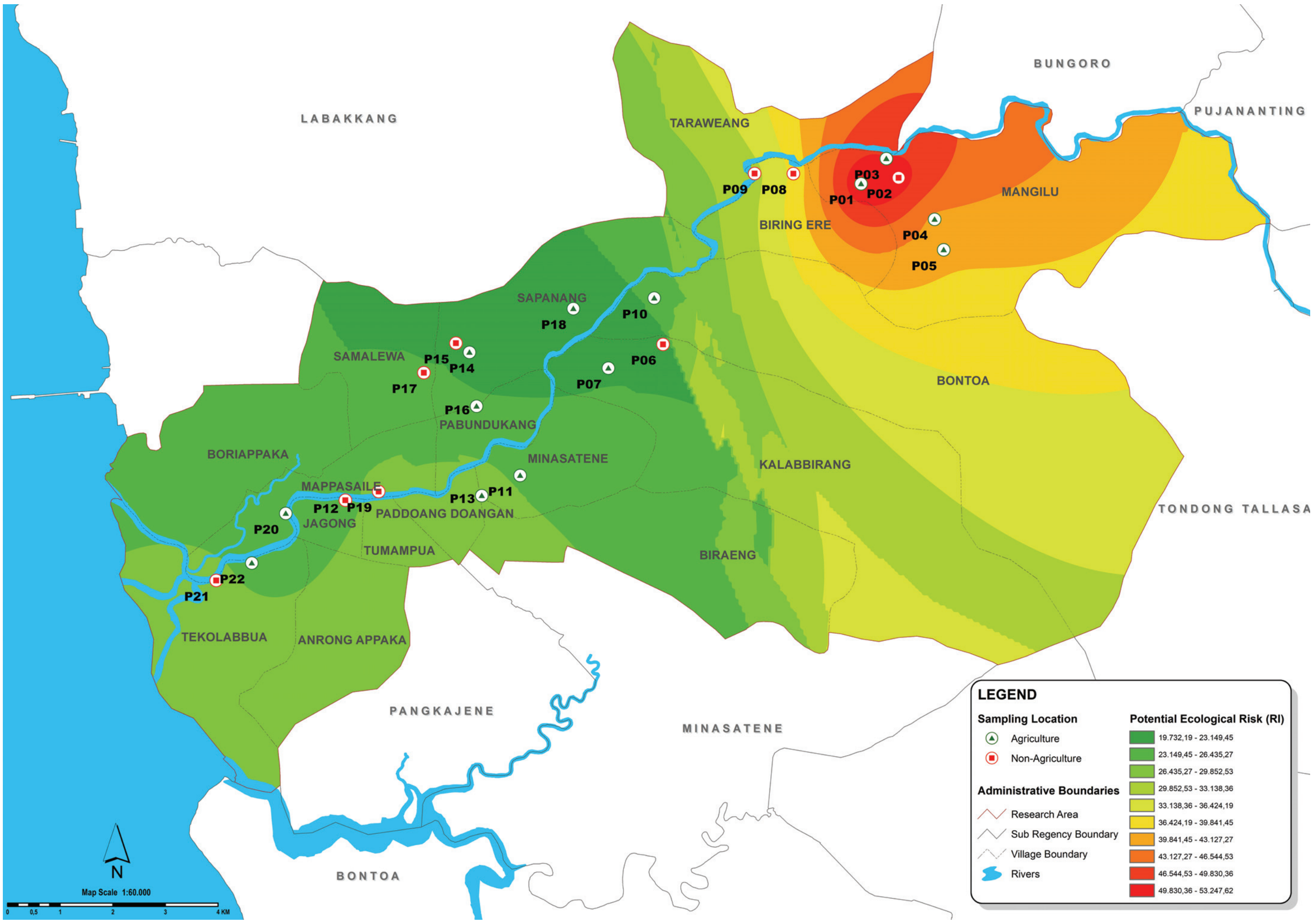


Fig. 5. Potential ecological risk (RI) at Pangkajene watershed area, Indonesia

to the research that are needed to be performed in the future. First, the need for physical and chemical properties of the soil that influence the content of heavy metal. Second, the accumulation of Hg and Pb in rice. Finally, further studies are needed to analyse the effects of contamination on human health through the food chain

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