

SPATIAL DISTRIBUTION AND ECOLOGICAL RISK OF POTENTIALLY TOXIC ELEMENTS IN MAROS REGENCY, INDONESIA

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Abstract: Air pollution is a major environmental problem in Indonesia. Potentially toxic elements (PTEs) which bounded to particulate matter (PM) samples were collected from Maros karst area, the second largest karst in the world. The seasonal levels of PTEs and ecological risk assessment were used to investigate the pollution levels near the residential areas using pollution load index (PLI) and potential ecological risk index (PERI). Spatial distributions of ecological risks based on the PTEs concentration have been explored with Empirical Bayesian Kriging method. The result indicated the mean concentration of PTEs (Cr, Pb, Cu, As and Zn) were significantly higher in dry season than wet season. Based on the PERI and PLI values, the PTEs accumulation are more severe near industrial activities and traffic roads. Hotspots of the PTEs were located in the East and Southeast area. The implications of this study could be used to optimize the management strategies in controlling the PTEs pollution and become a scientific reference for taking environmental protection policies.

Keywords: air pollution, ecological risk, Maros karst, pollution index, trace metals

1. INTRODUCTION

Air pollution is caused by the accumulation of pollutants released into the atmosphere (Manisalidis et al., 2020). These pollutants are generated from natural events such as volcanoes, wildfires and dust storms or human activities including coal combustion, vehicle emissions and waste burning (Yin et al., 2021; Jelea, et al., 2007a; Rauf et al., 2020). The problem of particulate matter (PM) in ambient air has received a lot of attention as the social economy, industrialization, and urbanization have grown. The PM can reduce visibility and have a severe negative impact on climate change in global and regional areas (Lertxundi et al., 2010). The trace elements represent a small fraction of the total PM mass. Among the trace elements, arsenic (As), lead (Pb), cadmium (Cd), chromium (Cr) and mercury (Hg) are classified as potentially toxic elements (PTEs) or poisonous even at low concentrations. Moreover, nickel

(Ni), iron (Fe), and zinc (Zn) are micronutrients, but at certain concentrations and exceeding the specified limits, they can cause adverse impact on human health and organisms (Pourret & Hursthouse, 2019; Vitó et al., 2020). Thus, these elements are also included as PTEs. Several studies have proved that PTEs are associated with adverse health effects (Ziyadeh et al., 2019).

Increasing anthropogenic activities in Indonesia, which elevated the number of vehicles, fossil fuel combustion, construction, and stone crushing, has been reported to produce PM that possibly contains PTEs (Kurniawan et al., 2021; Santoso et al., 2020). These activities potentially degrade the environment, resulting the decreased environmental quality and human health problems (Jelea, et al., 2007b). Some reports confirmed the high levels of PTEs that accumulated in ambient air around anthropogenic activities such as road dust and industrial processes (Celo et al., 2021; Mallongi et al., 2021). Tehran, the most industrialized city in Iran, had

high enrichment levels of Cd, Cu, Pb, and Zn, with mean concentrations 6.6, 6.4, 7.5, and 8.4 times higher than background values due to road dust and industrial activity (primarily the cement industry) (Ali-Taleshi et al., 2021). Another study with the same results confirmed the impact of meteorological conditions on Pb contamination in atmospheric particles from industrial sources (Zhou et al., 2020). The accumulation of PTEs in the environment will bioaccumulate in the food chain that affecting animals and humans (Thalassinou & Antoniadis, 2021). This is in accordance with the findings reported in the target hazard quotient (THQ) of Pb and Cr in zucchini and spinach stems that exceeded the safe limit in Bangladesh (Al Amin et al., 2020).

Elements of the earth's crust, such as Zn, Mn and Ni, usually come from natural enrichment such as rock weathering and soil resuspension. A study in Qatar found the accumulation of Cd, Cu, Pb, and V is attributed to parent rock material and the natural weathering process (Alsafran et al., 2021). Geogenic inputs should not be neglected since local geology plays a substantial role (Marszałek et al., 2014). In this present study, the area is mostly surrounded by karst rocks consisting of basalt, limestone, alluvial soil and intrusive rocks (Astuti et al., 2021a; Rauf et al., 2021a). Moreover, this area is included as a national park and tourism destination, where the famous art painting caves from prehistoric times lie in this region (Huntley et al., 2021). In the last two decades, the activity of karst quarries in this area has been increasing. The largest private cement factory in Eastern Indonesia, marble factories and karst mining were established. This condition can be a serious threat to the environment and human.

Dry and wet deposits can dissolve rock and cause a wind-assisted distribution of pollutants into the air. Hence, the soil resuspension and the blowing winds are able to distribute PTEs in the air. The accumulation of PTEs in PM is strongly associated with ecological problems in water and soil (Astuti et al., 2021b; Damian et al., 2018). Previous studies recorded high levels of Hg and Cr in soil near residential areas around a cement plant (Mallongi et al., 2020), while the presence of Cr (VI) and SiO₂ found in groundwater wells (Rauf et al., 2021b; Astuti et al., 2021c). However, there are no recent studies regarding the presence and accumulation of PTEs in ambient air across the region. So, it is necessary to evaluate the levels of PTEs in the air and the ecological risk estimation in the study area for preparing the policy adjustments to reduce PTEs exposure in environments and residential areas.

In this work, the seasonal variations of PTEs concentration are used to determine the ecological

risk through pollution load index (PLI) and potential ecological risk index (PERI). Deeper knowledge of the distribution and the levels of PTEs in Eastern Indonesia will be beneficial to the investigation of chemical potential effects in Maros karst area and human well-being.

2. METHODS

2.1. Study Area

The sampling location is located around Maros regency in South Sulawesi, Indonesia, which is a very famous for limestone karst area, cave complex and prehistoric art that covers an area of ~ 450 km² between 4°7'S and 5°1'S (Huntley et al., 2021). This area is a nationally protected as a national park in Indonesia because it has abundant natural resources for the unique cone karst and hosts the richest hot-spot of tropical cave biodiversity (Hoch et al., 2011). Maros karst has rock layers or river underground in the almost 300 caves. The groundwater resources are used for drinking water and domestic purposes for the surrounding community. Unfortunately, during the last few decades there have been exploitation activities around this area. Several industrial activities operating around this area such as marble factories, cement plant and local rock mining. The local emission and vehicle smoke from the loading activities near residential areas also possible to affect the conditions of vegetations, air quality and even affect the health of the local residents. The sampling site is surrounded by karst hills, agricultural land and wetlands.

2.2. Sampling Collection

The particulate matter was collected from 9 to 30 November 2020 (wet season) and 23 July to August 2021 (dry season). The sampling location is located in Bantimurung sub-district, approximately 35 km North of Makassar city. Sampling was carried out around residential areas and located at least 300-500 m from the roads. A high-volume air sampler (HVAS) (Staplex TFIA 2) was used to collect the PTEs samples in the form of particulate matter for 24 hours in (seven days in each season). Ambient air that contains particulate matter is drawn by a fan into the HVAS with the help of a suction pump. Particulates trapped in the filter paper will be wrapped in aluminium foil before being analysed in the laboratory. The PTEs, including Cr, Pb, Cu, Al, As, Ni and Zn were measured by inductively coupled plasma optical emission spectrometry (ICP-OES). The calibration curves were linear and greater than 0.995).

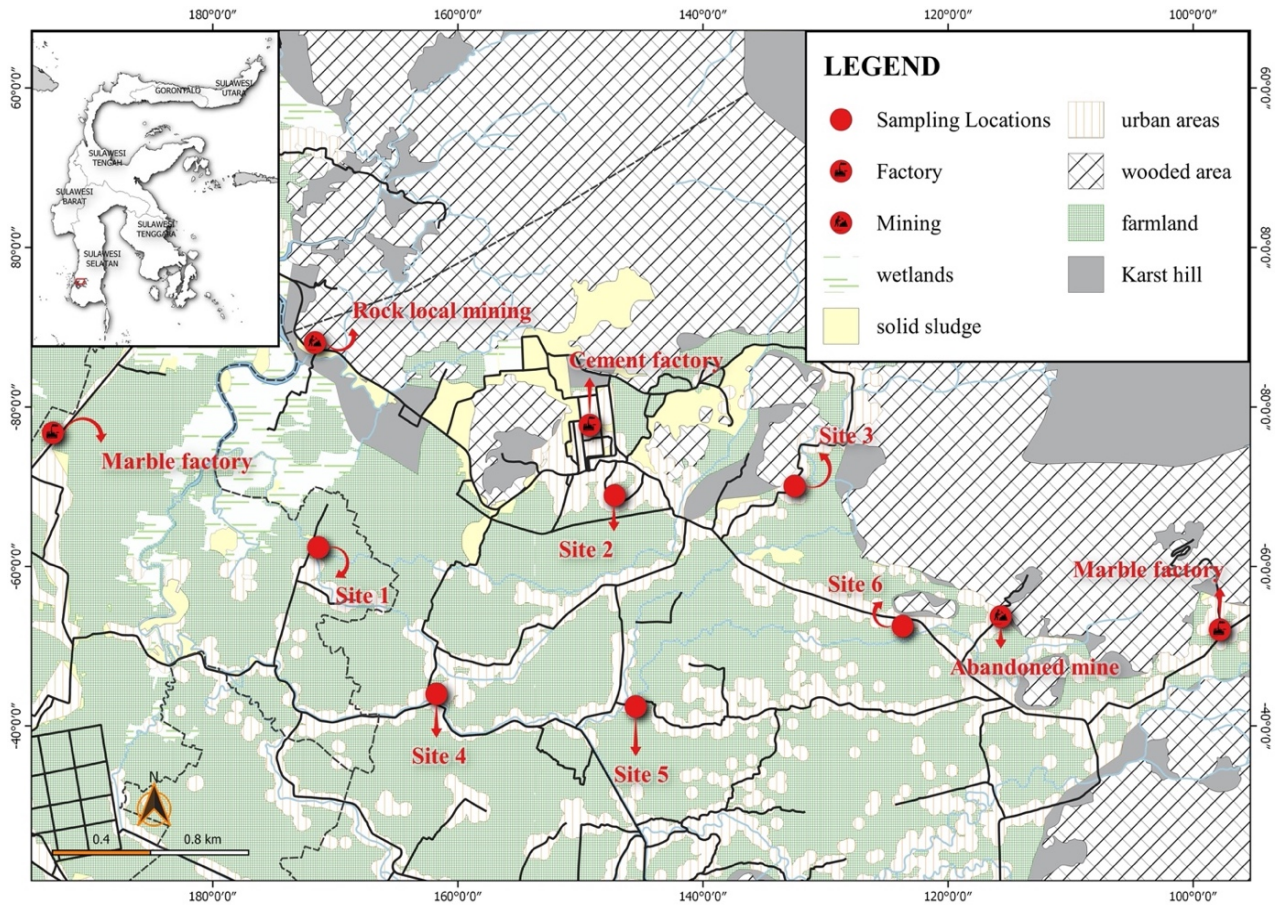


Figure 1. Study area and sampling locations in Maros Regency.

2.3. Pollution Level Assessment

2.3.1. Potential Ecological Risk Index (PERI)

This index developed by Hakanson (1980). The potential ecological risk in ambient air were calculated by using E_r and risk index (RI) (Hakanson, 1980). The following equations were used to calculate potential risks:

$$C_f^i = \frac{C_d^i}{C_r^i} \quad E_r^i = T_r^i \times C_f^i$$

$$PERI = \sum_{i=1}^n E_r^i$$

where C_f^i is the metal pollution for a single element, C_d^i is the metal(s) concentration from the site, C_r^i is the background concentration of metal in examined environment. The level of metal contamination was classified as low for $C_f^i < 1$, moderate for $1 \leq C_f^i < 3$, considerable for $3 \leq C_f^i < 6$, and very high for $C_f^i \geq 6$. E_r^i is the potential ecological risk for a single metal, T_r^i is the toxicity coefficient for metal i , and RI is a multiple metals ecological risks at a single site. The toxicity coefficients of Pb, Zn, Ni, Cu, Cr, Cd, Mn and As were 5,1,5,5,3,30,1 and 10, respectively.

PERI can be categorized as follows, $PERI < 40$, low risk; $40 \leq PERI < 80$, moderate potential risk; $80 \leq PERI < 160$, considerable potential risk; $160 \leq PERI < 320$, potential risk; $PERI \geq 320$, very high risk.

2.3.2. Pollution Load Index (PLI)

The degree of metal pollution from the ambient air was evaluated by applying Pollution Load Index (PLI) (Tomlinson et al., 1980). PLI values of measured metal contamination in sediment, soil and dust across the study site and control area generally found less than 1 (Ekwere & Edet, 2021). PLI calculated by the following equation.

$$PLI = (C_f^1 \times C_f^2 \times C_f^3 \times \dots \times C_f^i)^{1/n}$$

Where i is the number of pollutants and C_f^i is the value of the contamination factor from the previous equation. The degree of PLI can be categorized as follows, no pollution for $PLI < 1$, moderate pollution for $PLI = 1$ and deterioration of the site quality $PLI > 1$.

2.3.3. Statistical Analysis

All statistical and mathematical analysis were performed by using SPSS version 24.0 and Microsoft Excel 2018. For the spatial distribution of PERI and PLI, we applied gridding method from Golden

3. RESULT AND DISCUSSION

3.1. PTEs concentration

The average concentration of PTEs in the study area is presented in Figure 2. This data adapted from our previous work and proved that particulate matter was enriched by PTEs (Rauf et al., 2021c). In this study, Zn is the most abundant element among the metals studied in particulate matter. The presence of Zn in the atmosphere is the result of mining, coal burning and burning waste (ATSDR, 2005). A similar result was discovered in Southern China, where Zn accounts for roughly 60% of the total trace element (Sun et al., 2015). The significance of high amounts of Zn agrees with earlier observation in Nigeria (Olumayede & Ediagbonya, 2018), that far above the standard limits prescribed by WHO for respirable dust. This element is widely distributed in the earth's crust and mainly occurs from *sphalerite* rock, a major source of Zn that strongly associated with Pb and Fe (USEPA, 1969). Combustion processes around industrial areas can cause isotope fractionation of Zn due to redox effects (Schleicher et al., 2020). Karst quarries, limestone mining for cement production and marble factories around study sites may contribute to the high Zn and Al through combustion or crushing process and the release of dust through the stacks.

Arsenic (As) is one of the most dangerous and deadly metals on earth. The presence and high concentration of As in the air around residential areas indicates further degradation of the environment. The environmental-friendly method for As removal, strict regulations and emission control of anthropogenic activities around the study area are highly recommended. The existence of industrial activities correlated with biomass burning will produce dust from combustion. In Mazarrón, Spain, waste mines are the main source of several metals in dust, including As, Fe, Pb and Zn (Gabarrón, et al, 2018).

Transportation and distribution activities of industrial products also contribute to the high silt load, resuspension and accumulation of PTE in the air (Alshetty et al., 2022). A similar study in China proved that PM was driven by the air mass dispersion during the winter and spring seasons, which was mostly caused by traffic and fossil fuel emissions, particularly at night (Liu et al., 2022). The condition of the study area that is passed by transportation of cement production with high intensity every day can cause a high ecological risk, as happened at a cement factory in Ewekoro, Nigeria (Laniyan & Adewumi, 2020) and Dalian, China (Fan et al., 2021).

Spatial variations show that the number of PTEs was significantly higher in the dry season, except for Ni and Al. The dry season will increase the air temperature so that the particulates can be transported

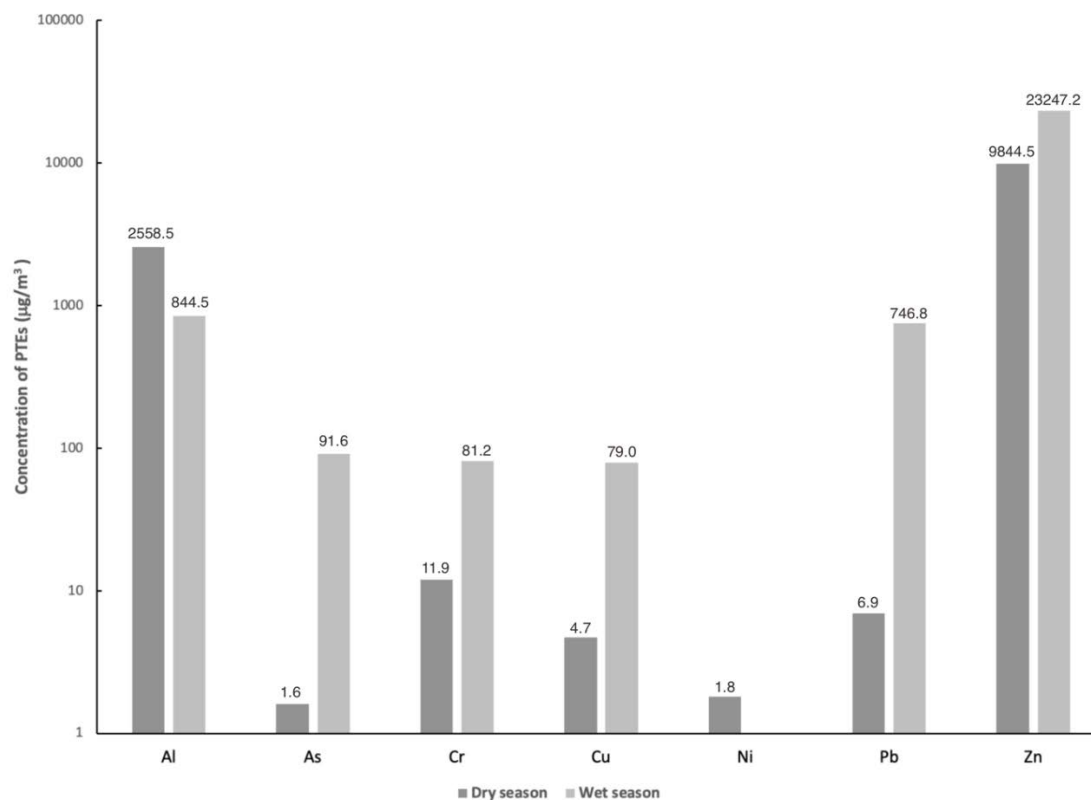


Figure 2. Potentially toxic elements concentration ($\mu\text{g}/\text{m}^3$) in wet and dry season.

further from the source (Rauf et al., 2021c). However, in both seasons, the concentrations of As, Cr and Pb exceeded the minimum limits set by WHO. The high concentration of these metals is possibly correlated with the increase of anthropogenic activity in this region. The cement plant and traffic roads that can produce smoke and particulate matter are associated with the high levels of PTEs (Kolo et al., 2018). These results were in accordance with a previous study in Ibadan city, where concentrations of Pb, Zn, and Cd were greater than their corresponding background values during dry seasons (Odediran et al., 2021). In India, the crustal elements, including Al, Si, Ca, Fe, and Ti were high during the pre-monsoon seasons, potentially due to high wind speeds (Nirmalkar et al., 2021). Cement factories produce harmful pollutants due to coal combustion, which at high temperatures will release harmful metals such as Hg, Cd, Co and Cr and pollute the environment around the factory (Fan et al., 2021; Kolo et al., 2018).

3.2. Pollution Level Assessment

The determination of PERI and PLI values was carried out for PTEs (As, Cr, Cu, Ni and Pb). Since Al does not have a background value, this element

was not considered in the ecological risk assessment of this paper. The PERI values in residential areas descended in order as follows: site 3 > site 4 > site 1 > site 6 > site 2 > site 5. PERI values showed extremely high potential risks in three residential areas; site 2 (163.76), site 5 (201.64) and site 6 (163.67). Site 5 and site 6 are located in the Southeast area of the cement factory and surrounded by the karst hills that lie to the North and Northeast. On the other hand, PERI values classified as considerable risk for site 1 (133.31), site 3 (100.24) and site 4 (115.70). These findings indicate the accumulation of PTEs in this area was extremely polluted and harmful for ecosystem. The high accumulation of PTEs in an area can result the disrupting in ecosystem. For example, dry deposition from the air can fall to the ground and accumulate over a long period of time. The presence of PTEs interferes the plant growth around the contaminated sites. Some plants can uptake large quantities of Cd, Pb and Cr in their root systems (Ibrahim et al., 2021).

A study in Sri Lanka found that most of the accumulated PTEs are mainly concentrated in the leaves of the grape tree rather than in the fruit, which will inhibit the process of plant growth and photosynthesis (Prabagar et al., 2021).

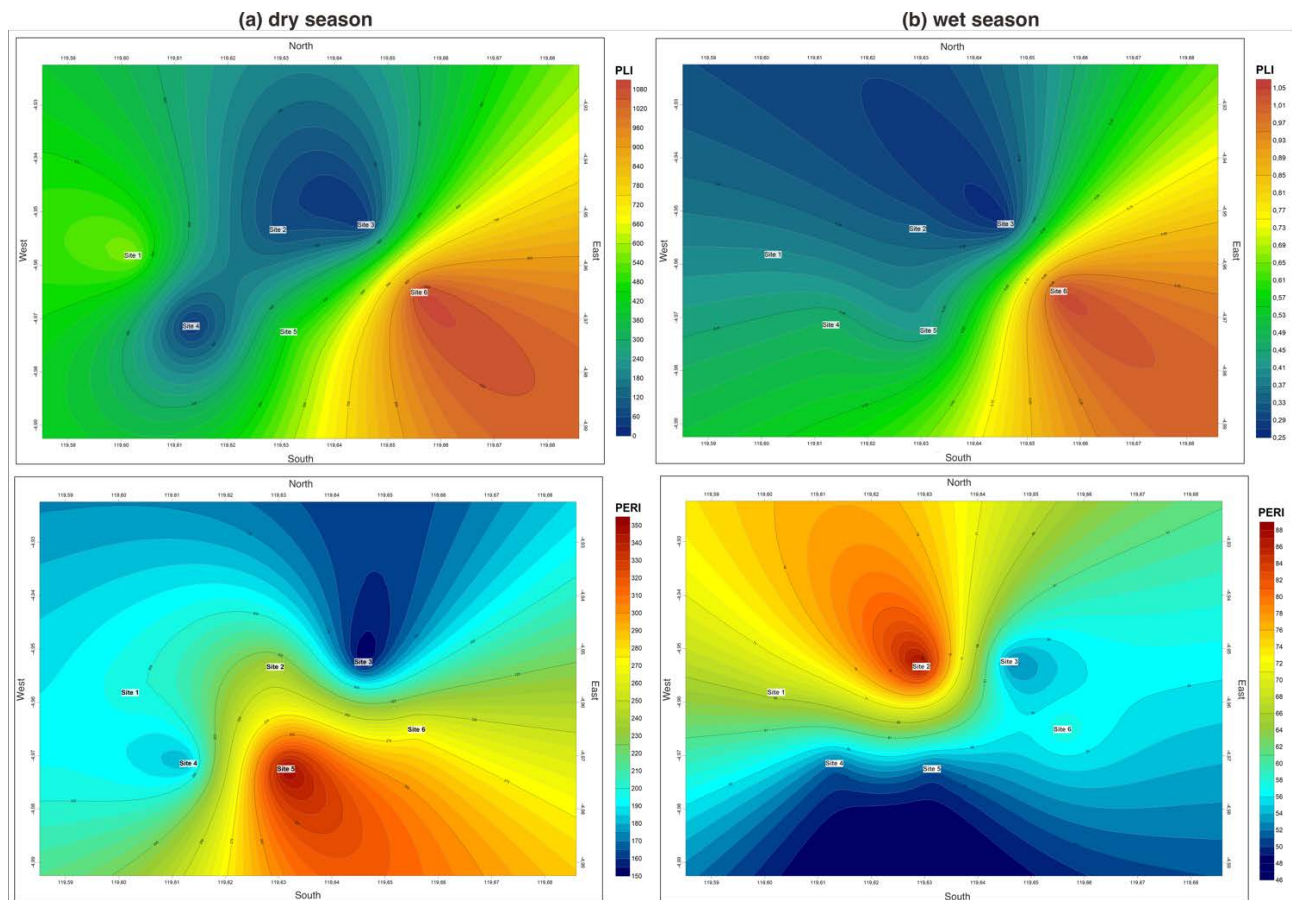


Figure 3. The PERI and PLI of PTEs accumulation in study area.

The PLI values of the metal in all sites were greater than 1; site 1 (21), site 2 (3.9), site 3 (2.0), site 4 (6.3), site 5 (63) and site 6 (346), indicating the accumulation of PTEs in PM can decreasing the ecological quality in all sites. The organisms around those locations may highly exposed by PTEs, where the highest values were recorded in site 6. The contribution of biomass burning aerosols that potentially emitted during dry season may play an important role. Moreover, the local emission from the industrial activities needs to be considered. A study conducted by Zhao (Cai & Li, 2019), revealed that the strength of precipitation in rainy days and the falling dust reflected the background characterization of particulates in the air which attributed to the regional anthropogenic activity and low atmospheric circulation. The spatial distribution of PERI and PLI of the PTEs in the study area is using Empirical Bayesian Kriging. Figure 3 shows the spatial distribution of pollution levels (PERI and PLI) in dry and wet season. These results are consistent with the previous study (Rauf et al., 2021c), where the particulate matter is higher and concentrated in the East and Southeast area. This is influenced by the prevailing winds which carry dust and other particles towards the settlements area.

The areas with the highest level of ecological risk (PLI and PERI) in the dry season are Baruga and Tukamasea village. This area are residential areas surrounded by karst towers, hills, rice fields and inhabited by residents before industrial activities began more than three decades ago. Dry season with less humidity and winds that blow predominantly towards the East and Southeast greatly contribute to the distribution of PTEs in the study area. This result proves the relationship with previous studies where the distribution of wind and dry deposition will bring particulates to move and fall to the ground near the area where people live (Rauf et al., 2020). Different results were obtained from PERI value in the wet season, which indicated that the level of ecological risk was more concentrated and higher in the Northwest. This area is the main route for transportation of cement products and traffic roads

4. CONCLUSION

In this study, the ecological risks and seasonal levels of PTEs, including Cr, Pb, Cu, Al, As, Ni and Zn in PM samples were determined. The mean concentrations of Cr, Pb, Cu, As and Zn were extremely higher in dry season than wet season, whereas the concentration of Ni is not detected or below the detection limit in dry season. The PERI and PLI values indicate that the closest location to

industrial activities and traffic roads posed higher ecological contamination. The spatial distribution of PERI and PLI values showed the hotspot maps of PTEs were located in the East and Southeast areas. This study demonstrates the power of spatial machine learning techniques to show the environmental condition of residential areas around Maros karst. Thus, preventive efforts to reduce the concentration of PTEs must be carried out by the government and the environmental department of all related industries.

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Conflicts of Interest

The authors declare no conflicts of interest.

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